Surface Science Division
Fachverband Oberflächenphysik (O)

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Overview of Invited Talks and Sessions

(Lecture halls H 0105, H 1012, HE 101, HL 001, MA 004, MA 005, MA 041, MA 042, MA 043, MA 141, MA 144, and TC 006; Poster A, C, and D)

Plenary Talks

PLV I	Mon	8:30– 9:15	H 0105	**Opportunities from single magnetic adatoms on superconductors** — •Katharina J. Franke
PLV IV	Tue	8:30– 9:15	H 0105	**Capturing light induced phase transitions with femtosecond movies** — •Nuh Gedik

Laureate of the Gaede Prize 2024

O 64.1	Wed	17:30–18:00	MA 004	**Molecular spin switches on surfaces** — •Manuel Gruber

Overview Talks

O 1.1	Mon	9:30–10:15	HE 101	**(Surface-)science-driven machine learning** — •Johannes T. Margraf
O 19.1	Tue	9:30–10:15	HE 101	**Quasiparticle dynamics and spin-orbital texture of 2D electron liquids at surfaces** — •Felix Baumberger
O 41.1	Wed	9:30–10:15	HE 101	**Extreme interaction of photons and electrons in metallic nanocavities and STM tunnelling gaps** — •Javier Aizpurua
O 73.1	Thu	9:30–10:15	HE 101	**Ice structures and dynamics on surfaces investigated on the local scale** — •Karina Morgenstern
O 100.1	Fri	9:30–10:15	HE 101	**Momentum microscopy & 2D materials: Excitons in space and time** — •Marcel Reutzel
O 112.1	Fri	13:15–14:00	HE 101	**A surface science approach to thermal and electrochemical ammonia synthesis** — •Ib Chorkendorff

Invited Talks of the joint Symposium "Frontiers of Electronic-Structure Theory - Advances in Time-Dependent and Nonequilibrium Ab Initio Methods" (SYES)

SYES 1.1	Tue	9:30–10:00	H 0105	**Light control of charge transport and phase transitions** — •Sheng Meng
SYES 1.2	Tue	10:00–10:30	H 0105	**Probing the transport of the interacting electron-phonon system self-consistently and *ab initio*** — •Nakib Protik
SYES 1.3	Tue	10:30–11:00	H 0105	**In- and out-of-equilibrium ab initio theory of electrons and phonons** — •Gianluca Stefanucci
SYES 1.4	Tue	11:15–11:45	H 0105	**Phonon screening of excitons in semiconductors and insulators from first principles** — •Marina Rucsandra Filip
SYES 1.5	Tue	11:45–12:15	H 0105	**Light-matter control of quantum materials: from Floquet to cavity engineering** — •Michael Sentef

Topical Talks of the Focus Session "Spins on Surfaces studied by Atomic Scale Spectroscopies"

O 3.1	Mon	10:30–11:00	MA 004	**Magnetic molecules on superconductors: from sensing experiments to novel molecular candidates for quantum platforms** — •Giulia Serrano
O 11.1	Mon	15:00–15:30	MA 004	**Unusual magnetism of closed-shell molecules on metal substrates** — •Alexander Weismann
O 21.1	Tue	10:30–11:00	MA 004	**Theory of Electron Spin Resonance in Scanning Tunneling Microscopy** — •Juan Carlos Cuevas
O 52.1	Wed	15:00–15:30	MA 004	**Locally driven quantum phase transitions in a strongly correlated molecular monolayer** — •Markus Ternes

Topical Talks of the Focus Session "Molecular Nanostructures on Surfaces: On-Surface Synthesis and Single-Molecule Manipulation"

O 20.1	Tue	10:30–11:00	HE 101	**Generation and Characterization of Cyclocarbons** — •Leo Gross
O 51.1	Wed	15:00–15:30	HE 101	**Exploring the Magnetic and Topological Properties in Carbon-based Nanomaterials** — •Ping Yu
O 87.1	Thu	15:00–15:30	HE 101	**Towards the third dimension: Organic molecular architectures synthesized on solid surfaces by means of solution-based click chemistry** — •Michael Dürr
O 103.1	Fri	10:30–11:00	HE 101	**Toward nanofabrication with molecular building blocks** — •Christian Wagner

Topical Talks of the Focus Session "Spin Phenomena in Chiral Molecular Systems"

O 26.1	Tue	10:30–11:00	MA 141	**The Electron's Spin and Chirality - a Miraculous Match** — •Ron Naaman
O 26.2	Tue	11:00–11:30	MA 141	**Electrons, Vibrations and Chirality** — •Martin B. Plenio
O 26.3	Tue	11:30–12:00	MA 141	**Electrical Dipole Moment Governs Spin Polarization in Charge Transport in Single α-helical Peptides Junctions** — •Ismael Diez-Perez
O 48.1	Wed	10:30–11:00	MA 141	**Chiral-induced Spin Selectivity in Hybrid Chiral Molecule/ Metal Systems** — •Angela Wittmann
O 48.2	Wed	11:00–11:30	MA 141	**Chirality-induced spin selectivity at the single-molecule scale** — •Daniel Emil Bürgler

Topical Talks of the Focus Session "Novel Approaches to Surface Plasmon Generated Charges and Heat for Photocatalysis (joint session O/CPP)"

O 58.1	Wed	15:00–15:30	MA 141	**Plasmonic photothermal chemistry on single nanoparticles** — •Andrea Baldi
O 58.6	Wed	16:30–17:00	MA 141	**Disentangling plasmonic catalysis contributions by time-resolved spectroscopy** — •Holger Lange
O 83.1	Thu	10:30–11:00	MA 141	**Designing Plasmonic Photocatalysts** — •Emiliano Cortes

Topical Talks of the Focus Session "Ultrafast Processes in Organic Semiconductors and Perovskites"

O 78.1	Thu	10:30–11:00	MA 004	**Transport and trapping in molecular materials at the picosecond time scale** — •Marina Gerhard
O 78.4	Thu	11:30–12:00	MA 004	**Ultrafast charge-transfer dynamics in organic and hybrid interfaces from first principles** — •Caterina Cocchi
O 88.1	Thu	15:00–15:30	MA 004	**Time-resolved chiroptical probes to track spin & light polarization in solution-processable semiconductors** — •Sascha Feldmann

Topical Talks of the Focus Session "Proximity Effects in Epitaxial Graphene"

O 94.1	Thu	15:00–15:30	MA 141	**Tailoring the electronic structure of epitaxial graphene on SiC** — •Kathrin Küster
O 94.6	Thu	16:30–17:00	MA 141	**Proximity spin-orbit coupling and topological interfaces in graphene / alloyed transition metal dichalcogenide heterostructures** — •Stephen Power, Zahra Khatibi
O 109.1	Fri	10:30–11:00	MA 141	**Heavy fermion quantum matter and topological superconductivity in artificial van der Waals heterostructures** — •Jose Lado
O 109.5	Fri	11:45–12:15	MA 141	**Superconducting quantum devices in twisted graphene layers** — •Klaus Ensslin

General Invited Topical Talks

O 4.7	Mon	12:00–12:30	MA 005	**Focused and coherent X-rays for the study of thin films and surfaces** — •Ivan Zaluzhnyy
O 13.1	Mon	15:00–15:30	MA 041	**Photocatalysis at the surface the TiO_2 on its real time** — •Heshmat Noei
O 14.3	Mon	15:30–16:00	MA 042	**Time-resolved interaction of the electron system with strong surface plasmon polariton fields** — •Frank Meyer zu Heringdorf
O 22.2	Tue	10:45–11:15	MA 005	**Bias free extraction of orbital angular momentum from two-dimensional materials by dichroic photoemission** — •Simon Moser
O 47.4	Wed	11:15–11:45	MA 043	**Adsorbate motors for unidirectional translation and transport** — •Grant J. Simpson
O 53.3	Wed	15:30–16:00	MA 005	**Tuning quantum electronic transport in nanoporous graphene** — •Aran Garcia-Lekue
O 54.1	Wed	15:00–15:30	MA 041	**Ultrafast scanning tunneling microscopy as a local probe of femtosecond electron and coherent phonon dynamics** — •Melanie Müller
O 54.6	Wed	16:30–17:00	MA 041	**Ultrafast scanning tunneling microscopy of a phonon-driven atomic vacancy in a monolayer crystal** — •Yaroslav Gerasimenko
O 60.5	Wed	16:00–16:30	MA 042	**Ab initio insights into atomistic processes at electrified solid/liquid interfaces** — •Mira Todorova
O 81.7	Thu	12:00–12:30	MA 042	**Trapping single atoms of noble gases in nanocages: from fundamental studies to applications** — •Jorge Anibal Boscoboinik
O 91.1	Thu	15:00–15:30	MA 041	**SPM on the path to direct nano-optical measurements** — •Martin Svec
O 96.4	Thu	15:45–16:15	TC 006	**Importance of charge transfer descriptor for the computational screening of electrocatalysts** — •Stefan Ringe
O 106.4	Fri	11:15–11:45	MA 041	**Characterization of just one atom using synchrotron x-rays** — •Volker Rose

Sessions

O 1.1–1.1	Mon	9:30–10:15	HE 101	**Overview Talk Johannes T. Margraf**
O 2.1–2.10	Mon	10:30–13:00	HE 101	**Focus Session: Frontiers of Electronic-Structure Theory – Advances in Time-Dependent and Nonequilibrium Ab Initio Methods I**
O 3.1–3.6	Mon	10:30–12:15	MA 004	**Focus Session: Spins on Surfaces studied by Atomic Scale Spectroscopies I**
O 4.1–4.7	Mon	10:30–12:30	MA 005	**New Methods: Experiments**
O 5.1–5.8	Mon	10:30–12:30	MA 041	**Ultrafast Electron Dynamics at Surfaces and Interfaces I**
O 6.1–6.10	Mon	10:30–13:00	MA 042	**Organic Molecules on Inorganic Substrates I: Adsorption & Growth**
O 7.1–7.10	Mon	10:30–13:00	MA 043	**Plasmonics and Nanooptics I**
O 8.1–8.10	Mon	10:30–13:00	MA 141	**Surface Reactions**
O 9.1–9.7	Mon	10:30–12:15	MA 144	**Semiconductor Substrates I: Adsorption of Small Molecules, Metallic Nanowires, Overlayers**
O 10.1–10.9	Mon	15:00–17:15	HE 101	**Focus Session: Frontiers of Electronic-Structure Theory – Advances in Time-Dependent and Nonequilibrium Ab Initio Methods II**

O 11.1–11.10	Mon	15:00–17:45	MA 004	**Focus Session: Spins on Surfaces studied by Atomic Scale Spectroscopies II**
O 12.1–12.10	Mon	15:00–17:30	MA 005	**2D Materials I: Electronic Structure (joint session O/TT)**
O 13.1–13.9	Mon	15:00–17:30	MA 041	**Ultrafast Electron Dynamics at Surfaces and Interfaces II**
O 14.1–14.9	Mon	15:00–17:30	MA 042	**Plasmonics and Nanooptics II: Light-Matter Interaction and Spectroscopy**
O 15.1–15.12	Mon	15:00–18:00	MA 043	**Organic Molecules on Inorganic Substrates II: Electronic, Optical and other Properties**
O 16.1–16.10	Mon	15:00–17:30	MA 141	**Surface Dynamics & Electron-Driven Processes**
O 17.1–17.8	Mon	15:00–17:00	MA 144	**Semiconductor Substrates II: Structure, Epitaxy, Growth**
O 18.1–18.12	Mon	15:00–18:00	H 1012	**Solid-Liquid Interfaces I: Structure and Spectroscopy**
O 19.1–19.1	Tue	9:30–10:15	HE 101	**Overview Talk Felix Baumberger**
O 20.1–20.9	Tue	10:30–13:00	HE 101	**Focus Session: Molecular Nanostructures on Surfaces: On-Surface Synthesis and Single-Molecule Manipulation I**
O 21.1–21.10	Tue	10:30–13:15	MA 004	**Focus Session: Spins on Surfaces studied by Atomic Scale Spectroscopies III**
O 22.1–22.6	Tue	10:30–12:15	MA 005	**2D Materials II: Electronic Structure (joint session O/TT)**
O 23.1–23.11	Tue	10:30–13:15	MA 041	**Ultrafast Electron Dynamics at Surfaces and Interfaces III**
O 24.1–24.10	Tue	10:30–13:00	MA 042	**Plasmonics and Nanooptics III: Light-Matter Interaction and Spectroscopy**
O 25.1–25.10	Tue	10:30–13:00	MA 043	**Organic Molecules on Inorganic Substrates III: Adsorption & Growth**
O 26.1–26.7	Tue	10:30–13:15	MA 141	**Focus Session: Spin Phenomena in Chiral Molecular Systems I (joint session O/TT)**
O 27.1–27.8	Tue	10:30–12:30	MA 144	**Tribology**
O 28.1–28.11	Tue	10:30–13:15	H 1012	**Solid-Liquid Interfaces II: Structure and Spectroscopy**
O 29.1–29.35	Tue	12:30–14:30	Poster A	**Poster: 2D Materials**
O 30.1–30.7	Tue	12:30–14:30	Poster A	**Poster: Proximity Effects in Epitaxial Graphene**
O 31.1–31.17	Tue	18:00–20:00	Poster C	**Poster: Organic Molecules on Inorganic Substrates**
O 32.1–32.17	Tue	18:00–20:00	Poster C	**Poster: Solid-Liquid Interfaces**
O 33.1–33.10	Tue	18:00–20:00	Poster C	**Poster: Supported Nanoclusters and Catalysis**
O 34.1–34.5	Tue	18:00–20:00	Poster C	**Poster: Surface Dynamics & Electron-Driven Processes**
O 35.1–35.7	Tue	18:00–20:00	Poster C	**Poster: Surface Reactions**
O 36.1–36.25	Tue	18:00–20:00	Poster C	**Poster: Ultrafast Electron Dynamics at Surfaces and Interfaces**
O 37.1–37.12	Tue	18:00–20:00	Poster D	**Poster: Electronic Structure of Surfaces and Spectroscopy of Surface States**
O 38.1–38.3	Tue	18:00–20:00	Poster D	**Poster: Electronic Structure Theory**
O 39.1–39.11	Tue	18:00–20:00	Poster D	**Poster: Metal Substrates**
O 40.1–40.6	Tue	18:00–20:00	Poster D	**Poster: Topological Materials**
O 41.1–41.1	Wed	9:30–10:15	HE 101	**Overview Talk Javier Aizpurua**
O 42.1–42.8	Wed	10:30–12:30	HE 101	**Focus Session: Frontiers of Electronic-Structure Theory – Advances in Time-Dependent and Nonequilibrium Ab Initio Methods III**
O 43.1–43.10	Wed	10:30–13:00	MA 004	**Focus Session: Spins on Surfaces studied by Atomic Scale Spectroscopies IV**
O 44.1–44.10	Wed	10:30–13:00	MA 005	**2D Materials III: Electronic Structure (joint session O/TT)**
O 45.1–45.9	Wed	10:30–12:45	MA 041	**Ultrafast Electron Dynamics at Surfaces and Interfaces IV**
O 46.1–46.10	Wed	10:30–13:00	MA 042	**Plasmonics and Nanooptics IV: Fabrication and Applications**
O 47.1–47.6	Wed	10:30–12:15	MA 043	**Metal Substrates I**
O 48.1–48.6	Wed	10:30–12:30	MA 141	**Focus Session: Spin Phenomena in Chiral Molecular Systems II (joint session O/TT)**
O 49.1–49.10	Wed	10:30–13:00	MA 144	**Oxide and Insulator Interfaces I**
O 50.1–50.8	Wed	10:30–12:30	TC 006	**Supported Nanoclusters: Structure, Reaction, Catalysis**
O 51.1–51.10	Wed	15:00–17:45	HE 101	**Focus Session: Molecular Nanostructures on Surfaces: On-Surface Synthesis and Single-Molecule Manipulation II**
O 52.1–52.8	Wed	15:00–17:15	MA 004	**Focus Session: Spins on Surfaces studied by Atomic Scale Spectroscopies V**
O 53.1–53.11	Wed	15:00–18:00	MA 005	**2D Materials IV: Graphene (joint session O/TT)**
O 54.1–54.9	Wed	15:00–17:45	MA 041	**Scanning Probe Microscopy: Light Matter Interaction at Atomic Scales I**

O 55.1–55.11	Wed	15:00–17:45	HL 001	**Topology and Symmetry protected Materials (joint session O/TT)**
O 56.1–56.4	Wed	15:00–16:00	MA 043	**Metal Substrates II**
O 57.1–57.7	Wed	16:15–18:00	MA 043	**New Methods: Theory**
O 58.1–58.8	Wed	15:00–17:30	MA 141	**Focus Session: Novel Approaches to Surface Plasmon Generated Charges and Heat for Photocatalysis I (joint session O/CPP)**
O 59.1–59.10	Wed	15:00–17:30	MA 144	**Oxide and Insulator Interfaces II**
O 60.1–60.10	Wed	15:00–17:45	MA 042	**Solid-Liquid Interfaces III: Reactions and Electrochemistry**
O 61.1–61.6	Wed	15:00–17:00	A 053	**Focus Session: 2D Transition Metal Carbides, Nitrides and Carbonitrides I (joint session DS/MM/O)**
O 62.1–62.7	Wed	15:00–18:00	Poster E	**Poster: $SrTiO_3$: A Versatile Material from Bulk Quantum Paraelectric to 2D Superconductor (joint session TT/KFM/MA/O)**
O 63.1–63.31	Wed	17:00–19:00	Poster B	**Poster DS (joint session DS/MM/O)**
O 64.1–64.1	Wed	17:30–18:00	MA 004	**Laureate of the Gaede Prize 2024**
O 65.1–65.17	Wed	18:00–20:00	Poster C	**Poster: Molecular Nanostructures on Surfaces**
O 66.1–66.22	Wed	18:00–20:00	Poster C	**Poster: Nanostructures at Surfaces**
O 67.1–67.10	Wed	18:00–20:00	Poster C	**Poster: Oxide, Insulator and Semiconductor Surfaces**
O 68.1–68.12	Wed	18:00–20:00	Poster C	**Poster: Scanning Probe Techniques: Method Development**
O 69.1–69.5	Wed	18:00–20:00	Poster C	**Poster: Spin Phenomena in Chiral Molecular Systems**
O 70.1–70.16	Wed	18:00–20:00	Poster C	**Poster: Spins on Surfaces & Surface Magnetism**
O 71.1–71.22	Wed	18:00–20:00	Poster D	**Poster: Plasmonics and Nanooptics**
O 72.1–72.13	Wed	18:00–20:00	Poster D	**Poster: Scanning Probe Microscopy: Light Matter Interaction at Atomic Scales**
O 73.1–73.1	Thu	9:30–10:15	HE 101	**Overview Talk Karina Morgenstern**
O 74.1–74.7	Thu	9:30–12:45	H 0104	**Focus Session: $SrTiO_3$: A Versatile Material from Bulk Quantum Paraelectric to 2D Superconductor I (joint session TT/KFM/MA/O)**
O 75.1–75.5	Thu	9:30–11:00	H 0107	**Focus Session: Wetting on Adaptive Substrates I (joint session CPP/DY/O)**
O 76.1–76.8	Thu	9:30–12:15	A 053	**Focus Session: 2D Transition Metal Carbides, Nitrides and Carbonitrides II (joint session DS/MM/O)**
O 77.1–77.5	Thu	10:30–13:00	HE 101	**Gerhard Ertl Young Investigator Award Competition**
O 78.1–78.8	Thu	10:30–13:00	MA 004	**Focus Session: Ultrafast Processes in Organic Semiconductors and Perovskites I (joint session O/CPP)**
O 79.1–79.10	Thu	10:30–13:00	MA 005	**2D Materials V: Growth, Structure and Substrate Interaction**
O 80.1–80.8	Thu	10:30–12:30	MA 041	**Scanning Probe Microscopy: Light Matter Interaction at Atomic Scales II**
O 81.1–81.8	Thu	10:30–12:45	MA 042	**Nanostructures at Surfaces I**
O 82.1–82.8	Thu	10:30–12:30	MA 043	**Electronic Structure Theory I**
O 83.1–83.10	Thu	10:30–13:15	MA 141	**Focus Session: Novel Approaches to Surface Plasmon Generated Charges and Heat for Photocatalysis II (joint session O/CPP)**
O 84.1–84.10	Thu	10:30–13:00	MA 144	**Electronic Structure of Surfaces I: Spectroscopy, Surface States**
O 85.1–85.9	Thu	10:30–12:45	TC 006	**Heterogeneous Catalysis I**
O 86.1–86.5	Thu	11:30–13:00	H 0107	**Focus Session: Wetting on Adaptive Substrates II (joint session CPP/DY/O)**
O 87.1–87.11	Thu	15:00–18:00	HE 101	**Focus Session: Molecular Nanostructures on Surfaces: On-Surface Synthesis and Single-Molecule Manipulation III**
O 88.1–88.11	Thu	15:00–18:00	MA 004	**Focus Session: Ultrafast Processes in Organic Semiconductors and Perovskites II (joint session O/CPP)**
O 89.1–89.7	Thu	15:00–17:30	A 053	**Focus Session: 2D Transition Metal Carbides, Nitrides and Carbonitrides III (joint session DS/MM/O)**
O 90.1–90.12	Thu	15:00–18:00	MA 005	**2D Materials VI: Growth, Structure and Substrate Interaction**
O 91.1–91.10	Thu	15:00–17:45	MA 041	**Scanning Probe Microscopy: Light Matter Interaction at Atomic Scales III**
O 92.1–92.11	Thu	15:00–17:45	MA 042	**Nanostructures at Surfaces II**
O 93.1–93.12	Thu	15:00–18:00	MA 043	**Scanning Probe Techniques: Method Development**

O 94.1–94.10	Thu	15:00–18:00	MA 141	**Focus Session: Proximity Effects in Epitaxial Graphene I**
O 95.1–95.10	Thu	15:00–17:30	MA 144	**Electronic Structure of Surfaces II: Spectroscopy, Surface States**
O 96.1–96.11	Thu	15:00–18:00	TC 006	**Solid-Liquid Interfaces IV: Reactions and Electrochemistry**
O 97.1–97.11	Thu	15:00–18:00	H 0104	**Focus Session: $SrTiO_3$: A Versatile Material from Bulk Quantum Paraelectric to 2D Superconductor II (joint session TT/KFM/MA/O)**
O 98	Thu	19:00–19:30	H 0105	**Members' Assembly**
O 99	Thu	19:30–20:30	H 0105	**Post-Deadline Session**
O 100.1–100.1	Fri	9:30–10:15	HE 101	**Overview Talk Marcel Reutzel**
O 101.1–101.11	Fri	9:30–12:30	H 0104	**Focus Session: $SrTiO_3$: A Versatile Material from Bulk Quantum Paraelectric to 2D Superconductor III (joint session TT/KFM/MA/O)**
O 102.1–102.10	Fri	9:30–12:45	H 0110	**Focus Session: Wetting on Adaptive Substrates III (joint session CPP/DY/O)**
O 103.1–103.9	Fri	10:30–13:00	HE 101	**Focus Session: Molecular Nanostructures on Surfaces: On-Surface Synthesis and Single-Molecule Manipulation IV**
O 104.1–104.5	Fri	10:30–11:45	MA 004	**Focus Session: Ultrafast Processes in Organic Semiconductors and Perovskites III (joint session O/CPP)**
O 105.1–105.9	Fri	10:30–12:45	MA 005	**2D Materials VII: Heterostructures (joint session O/TT)**
O 106.1–106.8	Fri	10:30–12:45	MA 041	**Scanning Probe Microscopy: Light Matter Interaction at Atomic Scales IV**
O 107.1–107.7	Fri	10:30–12:15	MA 042	**Nanostructured Surfaces and Thin Films**
O 108.1–108.8	Fri	10:30–12:30	MA 043	**Electronic Structure Theory II**
O 109.1–109.8	Fri	10:30–13:00	MA 141	**Focus Session: Proximity Effects in Epitaxial Graphene II**
O 110.1–110.9	Fri	10:30–12:45	MA 144	**Surface Magnetism**
O 111.1–111.8	Fri	10:30–12:30	TC 006	**Heterogeneous Catalysis II**
O 112.1–112.1	Fri	13:15–14:00	HE 101	**Overview Talk Ib Chorkendorff**

Members' Assembly of the Surface Science Division

Thursday 19:00–19:30 H 0105

- Report of the Chairman
- Election of a new Vice-Chair
- Presentation of the Gerhard Ertl Young Investigator Award
- Miscellaneous

O 1: Overview Talk Johannes T. Margraf

Time: Monday 9:30–10:15 Location: HE 101

Invited Talk O 1.1 Mon 9:30 HE 101
(Surface-)science-driven machine learning — •Johannes T. Margraf — University of Bayreuth

This talk discusses research towards the establishment of a science-driven approach to machine learning (ML) for surface science and chemistry.[1] In many fields, ML is a fundamentally data-driven endeavor, meaning that specific databases and benchmark problems (i.e. big data) are at the center of methodological development. While this has certainly led to tremendous advances in recent years (e.g. in image generation and natural language processing), the full diversity and complexity of surface chemistry cannot be adequately represented by static predefined databases. We therefore aim to build accurate data-efficient models which do not require enormous reference datasets for training. This way, our methods can be applied to a wide range of problems of scientific interest and not just to those for which big data happens to be available. To this end, we explicitly incorporate chemical and physical information into the ML models[2] and integrate the data generation or selection process with the model training[3]. Several examples of this in the context of the atomistic simulation of catalytic processes on surfaces will be discussed.

[1] J.T. Margraf, Angew. Chemie, 62, e202219170 (2023). [2] K. Chen et al. Chem. Sci., 14, 4913-4922 (2023). [3] H. Jung et al. NPJ Comput. Mater., 9, 114 (2023).

O 2: Focus Session: Frontiers of Electronic-Structure Theory – Advances in Time-Dependent and Nonequilibrium Ab Initio Methods I

Time-dependent and non-equilibrium phenomena in condensed matter involve a wide and diverse landscape of excitations (excitons, polarons, magnons, polaritons, etc.) and fundamental interaction mechanisms spanning different time and length scales as well as different levels of complexity. There has been enormous progress in the experimental probes of a variety of time-resolved phenomena with high precision, motivating further advancements in the ab initio description of ultrafast phenomena with electronic-structure calculations, explicit quantum dynamics, mixed quantum-classical dynamics methods, and many-body theories. This interdisciplinary Focus Session will cover recent progress in this area, with a primary focus on novel theoretical and computational paradigms for the description of time-dependent and nonequilibrium phenomena in quantum materials.

Organizers: Fabio Caruso (U Kiel), Claudia Draxl (HU Berlin), Reinhard Maurer (U Warwick)

Time: Monday 10:30–13:00 Location: HE 101

O 2.1 Mon 10:30 HE 101
Electron-phonon coupling in photoexcited graphene — •Nina Girotto and Dino Novko — Institute of Physics, Zagreb, Croatia

Understanding the out-of-equilibrium interactions and how they evolve in time is one of the main goals of ultrafast experiments. Here, we present an extensive study of the phonon spectrum and electron-phonon coupling in pristine and doped graphene during a strong optical excitation, calculated from first principles density-functional perturbation theory with adjusted occupations of the electronic states. By promoting a substantial number of electrons from the valence to the conduction band with a laser pulse, the thermalization process of photoexcited carriers includes three distinct electron distributions; photoexcited state is achieved exactly after the laser pulse and is followed by the photo-inverted state (cca 100 fs), and finally at timescales at the order of the electron thermalization time, the electrons follow a hot Fermi-Dirac distribution. In the three time regimes, we find renormalizations of the real and imaginary parts of the phonon self-energy, resulting from the interplay between ultrafast scatterings between nonequilibrium electrons and strongly coupled optical phonons and changes in the electron-phonon coupling matrix elements. Some of the compelling features are the photo-induced negative phonon linewidth, or 'phonon gain', found for some wave vectors, as well as anomalous phonon frequency shifts of strongly-coupled modes. Our results shed light on the intricate effects of photoexcitation on electron-phonon coupling, and track their changes in time.

O 2.2 Mon 10:45 HE 101
***Ab initio* theory of coherent phonons in semimetals** — •Christoph Emeis, Yiming Pan, and Fabio Caruso — Institut für Theoretische Physik und Astrophysik, Christian-Albrechts-Universität zu Kiel, 24118 Kiel, Germany

The spectral signatures of coherent phonons in pump-probe experiments can disclose novel insight into electron-phonon interaction and its influence on ultrafast phenomena. We developed an ab initio framework to describe the coherent phonon dynamics in photo-excited semimetals based on the time-dependent Boltzmann equation and its extension [1]. To illustrate the predictive capability of this approach, we investigate the displacive excitation of coherent phonons in Sb and $MoTe_2$ and discuss its fingerprints in frequency-domain ARPES, a powerful experimental technique to probe coherent phonons and electron-phonon coupling in semimetals [2]. [1] F. Caruso and D. Novko. Adv. Phys. X **7**, 2095925 (2022). [2] P. Hein et al. Nat. Commun. **11**, 2613 (2020).

O 2.3 Mon 11:00 HE 101
First-principles light-driven molecular dynamics through equivariant neural networks — •Elia Stocco[1], Christian Carbogno[2], and Mariana Rossi[1] — [1]MPI for the Structure and Dynamics of Matter, Hamburg, Germany — [2]Fritz Haber Institute of the MPS, Berlin, Germany

Recent experiments have shown a rich phenomenology in solids, liquids and molecules when driven by ultra-fast THz pulses. However, simulation techniques that can describe the nuclear dynamics of these processes for all material classes, without relying on dimensionality reduction, and going beyond perturbation theory, are very challenging. Here we propose an ab initio molecular dynamics (MD) method within the electric dipole approximation that allows a single machine-learning model to describe the coupling at diverse field strengths and with time dependence. Our requirement is that the system remains near the electronic ground state. We train an equivariant differentiable neural network to learn the dipole of isolated and periodic systems. Atomic tensor derivatives w.r.t. the nuclear coordinates are obtained through autodifferentiation. We present applications of this machine-learning-assisted MD protocol on water and LiNbO3. We show full-dimensional ab initio simulations of the excitation of vibrational modes at an ultrafast time scale and highlight the different nonlinearities of these dynamics.

O 2.4 Mon 11:15 HE 101
Anharmonic fingerprints from THz modes of naphtalene crystals enabled by machine-learning — •Paolo Lazzaroni, Shubham Sharma, and Mariana Rossi — Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany

Organic molecular crystals exhibit strong lattice anharmonicity, especially in the collective motions that are governed by intermolecular interactions and lie in the low-frequency THz range [1]. Inspired by recent observations that the polarization-orientation (PO) Raman spectra can give exquisite insight into the anharmonic couplings between modes [2], we devise a first-principles framework that aims at reproducing, explaining and give quantitative insight into the type and strengths of mode coupling. This framework is based on machine-learned potentials and polarizability tensors trained on ab-initio molecular dynamics trajectories [3]. We obtain results, even for large system supercells, through the time-correlation formalism for PO Raman signals, retaining the full anharmonic nature of the potential. In order to do this, a procedure has been developed that allows us to isolate the Γ-point Raman signal from our real-space molecular dynamics simulations. [1] M. Asher et al., Adv. Mater. 32, 1908028 (2020) [2] N. Benshalom et al., J. Phys. Chem. C 127, 36 (2023) [3] N. Raimbault et al., New J. Phys. 21 105001 (2019)

O 2.5 Mon 11:30 HE 101

Electron-phonon interaction in silicon and diamond using Gaussian orbitals

— •Gerrit Johannes Mann, Thorsten Deilmann, and Michael Rohlfing — Institute of Solid State Theory, University of Münster, Germany

Electron-phonon interaction is a crucial mechanism in solid state physics that is responsible for a multitude of phenomena. However, in electronic structure calculations it is often neglected. We developed an ab-initio implementation on top of density functional theory that combines finite differences calculations with the perturbative Allen-Heine-Cardona framework in order to calculate the temperature-dependent renormalization of the electronic bandstructure due to electron-phonon interaction using a basis set of localized Gaussian orbitals.

Our implementation circumvents the limiting problems of previous implementations while maintaining a good agreement with the literature. For the direct band gaps of silicon and diamond we find a zero-point renormalization of $-43\,\mathrm{meV}$ and $-411\,\mathrm{meV}$, respectively. In addition, the approach allows to evaluate Debye-Waller contributions beyond the rigid-ion approximation.

O 2.6 Mon 11:45 HE 101

Electron-phonon interactions in hybrid inorganic-organic systems from first-principles calculations

— •Ignacio Gonzalez Oliva[1], Sebastian Tillack[1], Fabio Caruso[1,2], Pasquale Pavone[1], and Claudia Draxl[1] — [1]Institut für Physik und IRIS Adlershof, Humboldt-Universität zu Berlin, 12489 Berlin, Germany — [2]Institut für Theoretische Physik und Astrophysik, Christian-Albrechts-Universität zu Kiel, 24118 Kiel, Germany

Hybrid materials composed of organic molecules and two dimensional (2D) transition metal dychalcogenides (TMDCs) exhibit interesting physical processes happening at the interface. By means of full-potential all-electron density-functional theory and many-body perturbation theory, we investigate the electronic properties and electron-phonon interactions of a prototypical system, comprising of a $\mathrm{MoS_2}$ monolayer and a physisorbed molecule, pyrene. G_0W_0 calculations show that the electronic levels of both components are renormalized due to dynamical screening effects. Temperature renormalization of the electronic bands are computed with the electron-phonon self-energy in the Fan-Migdal approximation. The calculated spectra reveal discernible features attributable to electron-phonon coupling within this system. Notably, distinctive signatures emerge, suggesting the manifestation of polaronic effects attributed to the presence of pyrene.

O 2.7 Mon 12:00 HE 101

Phonon mediated superconductivity obtained by combining `elphbolt` and `exciting`

— •Marten Pretorius, Sebastian Tillack, Ignacio Gonzalez Oliva, Pasquale Pavone, Nakib Haider Protik, and Claudia Draxl — Humboldt-Universität zu Berlin

We present an *ab initio* study of superconductors within the Migdal-Eliashberg formalism. Using the implementation of the full-potential all-electron density-functional theory package `exciting` [1], we construct compact real-space representations of the dynamical matrix, Hamiltonian, and the electron-phonon interaction matrix using density-functional perturbation theory and maximally-localized Wannier functions. These generated quantities are passed to the `superconda` app of the `elphbolt` [2] package, which solves the coupled Migdal-Eliashberg equations for the superconducting gap and the mass renormalization. We apply this workflow to selected materials.

[1] A. Gulans, *et al.*, J. Phys.: Condens. Matter **26**, 363202 (2014).

[2] N.H. Protik, *et al.*, npj Comput. Mater. **8**, 28 (2022).

O 2.8 Mon 12:15 HE 101

Phonons in FLEUR employing Density Functional Perturbation Theory: Towards 2D

— •Thomas Bornhake[1,2], Alexander Neukirchen[1,2], Gregor Michalicek[1], Daniel Wortmann[1], Gustav Bihlmayer[1], and Stefan Blügel[1,2] — [1]Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich GmbH and JARA, 52425 Jülich, Germany — [2]RWTH Aachen University, 52062 Aachen, Germany

Phonons are important excitations in solids. The investigation of phonon properties by density functional theory of magnetic systems is underrepresented in the literature. Recently, we completed successfully the implementation of the phonon dispersion using the density functional perturbation (DFPT) method in the all-electron full-potential linearized augmented plane-wave (FLAPW) method FLEUR [1,2]. In this talk, we present recent advancements for phonon calculations within FLEUR. We discuss calculations in magnetic bulk systems and compare these to phonon dispersions obtained with the finite displacement method for which the FLEUR code has been combined with the phonopy tool [3]. Motivated by the physics of 2D materials, we extended our methodology to 2D films in semi-infinite vacuum [4].

This work was supported by the Center of Excellence MaX (H2020-INFRAEDI-2018-1 767, Grant No. 824143)

[1] D. Wortmann *et al.*, 10.5281/ZENODO.7891361; www.flapw.de

[2] C.-R. Gerhorst *et al.*, arXiv:2309.14799v2 (2023).

[3] http://www.phonopy.github.io/phonopy/

[4] H. Krakauer *et al.*, Phys. Rev. B **19**, 1706 (1979)

O 2.9 Mon 12:30 HE 101

Light-matter dynamics in full minimal coupling Maxwell-TDDFT for beyond-dipole interactions

— •Franco P. Bonafe, Esra Ilke Albar, Heiko Appel, and Angel Rubio — MPI for Structure and Dynamics of Matter, Hamburg, Germany

The electric dipole approximation (EDA) has been by far the most extensively used treatment of light-matter interaction in molecular and nanoscopic systems, owing to the typically long wavelength of the incoming field compared to the size of these systems. However, for the deep understanding of novel spectroscopy techniques and phenomena where the interplay between electric and magnetic fields is relevant, a description beyond the EDA is sometimes mandatory.

In this talk, we show an efficient fully ab initio approach to couple electrons, nuclei and photons, by self-consistently coupling Maxwell's equations with time-dependent density functional theory (TDDFT) using the Pauli full minimal coupling Hamiltonian [1]. This method is based on a rigorous density-functional reformulation of the non-relativistic Pauli-Fierz Hamiltonian of quantum electrodynamics, and has been implemented in the Octopus package [2]. The effects of the non-uniform transverse fields in the dynamics will be exemplified with several systems, including the Cherenkov radiation from an electronic wavepacket, core-level photoabsorption, and chiroptical activity.

[1] R. Jestädt, M. Ruggenthaler, M.J.T. Oliveira, A. Rubio, and H. Appel. Adv. Phys. 68:4, 225 (2019)

[2] N. Tancogne-Dejean, M.J.T. Oliveira, et al. J. Chem. Phys. 152, 124119 (2020)

O 2.10 Mon 12:45 HE 101

Path-integral generalized Langevin dynamics: quantum nuclei with position-dependent friction

— •George Trenins[1], Paolo Lazzaroni[1], Yair Litman[2], and Mariana Rossi[1] — [1]MPI for the Structure and Dynamics of Matter, Hamburg, Germany — [2]Yusuf Hamied Department of Chemistry, University of Cambridge, UK

Dissipative dynamics model large-scale problems by treating a few degrees of freedom explicitly and describing the rest statistically. For example, in the study of nuclear dynamics at metal interfaces, nonadiabatic coupling to the electrons can be modelled as “electronic friction” [1]. Such systems with no clear separation of timescales are a computational challenge, due to the dependence of the dissipative forces on position and time. To address this, our group has developed an instanton rate theory that accounts both for the friction and for nuclear quantum effects [2]. However, since this theory cannot describe reactions taking place in liquid environments, we explore an alternative approach, explicitly introducing friction into ring-polymer molecular dynamics [3]. This framework uncovers the unexpected interplay between position-dependent friction and quantum dynamics in a model potential. We show practical approaches to propagating the underlying equations of motion and solutions to simulating multidimensional quantum dissipative systems. [1] W. Dou, J. E. Subotnik, JCP **148**, 230901 (2018). [2] Y. Litman et al. JCP **156**, 194106 (2022). [3] S. Habershon et al. Annu. Rev. Phys. Chem. **64**, 387–413 (2013).

O 3: Focus Session: Spins on Surfaces studied by Atomic Scale Spectroscopies I

Spins on surfaces represent a prospering field fueled by advancements in scanning probe microscopies and quantum impurity theory. This research spans three key areas: spins on superconductors revealing subgap states, spins on normal metals showcasing unexpected Kondo effects, and spins on thin insulating films enabling coherent spin control and qubit gate operations. These phenomena, united by fundamental principles, present intriguing challenges such as extending coherence times and establishing coherent coupling between different spin systems.

Organizers: Christian Ast (MPI Stuttgart), Susanne Baumann, Sebastian Loth (U Stuttgart)

Time: Monday 10:30–12:15 Location: MA 004

Topical Talk O 3.1 Mon 10:30 MA 004

Magnetic molecules on superconductors: from sensing experiments to novel molecular candidates for quantum platforms — •Giulia Serrano — Department of Industrial Engineering, University of Florence, florence, Italy

The interaction between magnetic molecules and superconductors (SC) has unveiled intriguing phenomena with implications for spintronics and quantum technologies. A monolayer of Single Molecule Magnets, characterized by magnetic hysteresis, has demonstrated a remarkable sensitivity to the SC transition. This sensitivity influences their hysteresis and triggers the activation of the resonant magnetization regime [1],[2].

At the single molecule level, the emergence of localized magnetic states, resulting from the interplay between the spin and the SC surface, holds the potential to pave the way for more sophisticated quantum platforms, such as those involving topologically protected qubits. In this context, innovative molecular systems are proposed, wherein ligands mediate spin-spin interactions. Their preliminary exploration on surfaces is presented [3],[4].

[1] Serrano et al., Nat. Mater., 19, 546, 2020. [2] Serrano et al., Nat. Commun., 13, 3838, 2022. [3] Santanni et al., JACS Au, 3, 1250, 2023. [4] Ranieri et al., Chem. Sci., 14, 61, 2023.

O 3.2 Mon 11:00 MA 004

Magnetic field dependence of Yu-Shiba-Rusinov (YSR) excitations — •Niels P.E. van Mullekom[1], Benjamin Verlhac[1], Werner M.J. van Weerdenburg[1], Hermann Osterhage[1], Manuel Steinbrecher[1], Katharina J. Franke[2], and Alexander A. Khajetoorians[1] — [1]Institute for Molecules and Materials, Radboud University Nijmegen, the Netherlands — [2]Fachbereich Physik, Freie Universität Berlin, Germany.

Yu-Shiba-Rusinov (YSR) states arise from the exchange coupling between a local spin and a superconductor and can be characterized by resonances inside the gap superconducting gap. From the in-gap excitations alone, the interplay of this exchange interaction with other energy scales, as well as the role of higher spin degrees of freedom is not clear. Magnetic field dependent characterization could clarify this, but most studies of YSR states to date have been limited to bulk superconductors, which easily quench in the presence of modest magnetic fields.

Utilizing the enhanced critical fields of a thin superconducting film we characterize the magnetic phase diagram of a molecule on the surface using high resolution milliKelvin scanning tunneling microscopy and spectroscopy. We observe nontrivial changes in the YSR excitations, that go beyond the trends that are expected in a spin 1/2 picture. We relate these changes to the various properties of the molecule, such as the role of multiple channels and magnetic anisotropy. These results provide an in-depth and detailed approach to understand the role of high spin states in the YSR excitations.

O 3.3 Mon 11:15 MA 004

Dynamics of individual Yu-Shiba-Rusinov states measured with stochastic resonance spectroscopy — •Nicolaj Betz[1], Vivek Krishnakumar Rajathilakam[1], Susan Coppersmith[2], Susanne Baumann[1], and Sebastian Loth[1] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]School of Physics, University of New South Wales, Sydney, Australia.

Coupling a spin to non-ohmic environments such as a superconductor can lead to new behavior with interesting dynamics. Thus far, the dynamics of emerging states like Yu-Shiba-Rusinov (YSR) states at individual atoms or molecules have mostly been investigated through tunneling rates [1,2]. Here, instead, we show a measurement that uses the synchronization of the state occupation with a frequency-dependent drive voltage to measure the dynamics of the tunneling through YSR states themselves. This technique relies on stochastic resonance which was previously used to measure spin dynamics [3]. We use a scanning tunneling microscope (STM) to measure the dynamics of YSR states of Fe atoms adsorbed on the oxygen-reconstructed surface of V(100) and find YSR state lifetimes in the picosecond range. This work provides a new level of insights into the dynamics of YSR states and can lead to a better understanding of relaxation mechanisms in spin-superconductor structures.

[1] M. Ruby et al., PRL 115, 087001 (2015).

[2] H. Huang et al., Nat. Phys. 16, 1227-1231 (2020).

[3] M. Hänze et al. Sci. Adv. 7, eabg2616 (2021).

O 3.4 Mon 11:30 MA 004

Electron transport in magnetic atomic and molecular junctions between superconducting leads — •Nicolas Lorente[1,2,3], Divya Jyoti[2,3], Cristina Mier[2,3], and Deung-Jang Choi[2,3] — [1]Centro de Física de Materiales (CSIC-EHU), San Sebastian, Spain — [2]Materials Physics Center, San Sebastian, Spain — [3]Donostia International Physics Center, San Sebastian, Spain

We present experiments and theory using self-consistent Bogoliubov-de Gennes equations implemented with Green's function for wide-band s-wave superconductors. We show the effect of magnetic junctions and multiple Andreev reflection when there are localized magnetic moments in the junction. The theoretical results agree with experiments performed on Nickelocene molecules trapped between a superconducting tip and a superconducting surface. Adding Fe atoms to the junctions enhances the magnetic scattering which can be easily modeled with the theory. Experimentally, the Fe atoms can be manipulated to be between molecule and interface or to be subsurface, with very different signature in the differential conductance. Self-consistency does not alter the results except when studying the evolution of the order parameter, which has bearings in the prediction of normal and topological quantum phase transitions.

O 3.5 Mon 11:45 MA 004

Magnetic bound states of iron clusters on a superconductor — •Ankur Das[1], Silas Amann[1], Nóra Kucska[2], András Lászlóffy[2], Nicolas Néel[1], Balázs Újfalussy[2], Levente Rózsa[2,3], Krisztián Palotás[2,3,4], and Jörg Kröger[1] — [1]Institut für Physik, Technische Universität Ilmenau, D-98693 Ilmenau, Germany — [2]Department of Theoretical Soild-State Physics, HUN-REN Wigner Research Centre for Physics, Institute for Solid State Physics and Optics, H-1525 Budapest, Hungary — [3]Department of Theoretical Physics, Institute of Physics, Budapest University of Technology and Economics, H-1111 Budapest, Hungary — [4]HUN-REN-SZTE Reaction Kinetics and Surface Chemistry Research Group, University of Szeged, H-6720 Szeged, Hungary

The magnetic exchange interaction of Fe$_n$ (n=1-3) clusters with superconducting Pb(111) is probed by scanning tunneling spectroscopy of Yu-Shiba-Rusinov states. The Yu-Shiba-Rusinov resonances are shifted from the coherence peaks in the Fe monomer spectrum towards the Fermi energy in the Fe dimer spectrum. Unexpectedly, the linear Fe trimer does not follow this trend, as it exhibits an almost identical spectrum to the single Fe atom. Kinked Fe trimers in contrast show strong Yu-Shiba-Rusinov resonances well within the Bardeen-CooperSchrieffer energy gap of the substrate. First-principles simulations of the Yu-Shiba-Rusinov states reveal which adsorption geometries and magnetic structures of the clusters can reproduce the experimental spectra most accurately.Funding by the Deutsche Forschungsgemeinschaft (KR 2912/18-1) is acknowledged.

O 3.6 Mon 12:00 MA 004

Ligand-mediated Yu-Shiba-Rusinov states of a S=1/2 molecule — •Lukas Arnhold[1], Matteo Briganti[2], Andrea Sorrentino[3], Nicolaj Betz[1], Lorenzo Poggini[4], Jaisa Fernandez Soarez[5], Luana C. de Camargo[5], Giulia Serrano[3], Matteo Mannini[2], Federico Totti[2], Roberta Sessoli[2], Susanne Baumann[1], and Sebastian Loth[1] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]Department of Chemistry Ugo Schiff, University of Florence, Italy — [3]Department of Industrial Engineering, University of Florence, Italy — [4]Institute of Chemistry of OrganoMetallic Compounds - CNR, Florence, Italy — [5]Department of Chemistry, Federal University of Paraná, Brazil

We couple ((*8-cyclooctatetraene)(*5-cyclopentadienyl)titanium) (CpTicot) molecules to superconducting Pb nanoislands on Si(111) in a scanning tunneling microscope (STM). This molecular magnet features a well isolated S=1 /2 ground state and is a prime candidate for molecular qubit applications due to exceptional spin coherence times in frozen solution [1]. On Pb nanoislands it exhibits a single YSR state stemming from a singly occupied molecular orbital (SOMO). We find that the molecule's spin interacts with the Pb surface via the cyclooctatetraene ligand making it possible to link the electronic properties of the molecule quantitatively to the in-gap YSR states both in theory and experiment. This experiment gives insights into chemical design priciples for targeted applications of YSR states. [1] de Camargo, L., et al. Angewandte Chemie. 60, Issue 5, 2588-2593

O 4: New Methods: Experiments

Time: Monday 10:30–12:30 Location: MA 005

O 4.1 Mon 10:30 MA 005

Record breaking resolution in cold antimatter detection using inexpensive hardware. — •Francesco Guatieri and Michael Berghold — Heinz Maier-Leibnitz Zentrum (MLZ), Technical University of Munich, Lichtenbergstr. 1, 85748 Garching, Germany

Modern beam facilities can provide two species of low-energy antiparticle beams: slow positron beams are available in many laboratories that perform surface studies and defect spectroscopy, while slow antiprotons are produced only by the AD/ELENA facility at CERN. Applications of both beams benefit greatly from the availability of realtime detectors of cold antimatter with a resolution in the scale of microns. Last year we demonstrated how modifying an inexpensive commercial camera sensor allows for the detection of positrons with a resolution that was unprecedented for real time applications. This year we broke another record by applying the same technology to cold antiprotons.

O 4.2 Mon 10:45 MA 005

how to stop motion of microparticles by using local light-driven diffusio-osmosis (l-LDDO) under external flow — •Daniela Vasquez, Fabian Rohne, Svetlana Santer, and Marek Bekir — University of Potsdam, Institute for Physic and Astronomy, Karl-Liebneckt str. 25, Haus 28, Potsdam, Germany

Liquid flow can be generated by external pressure (pumping) or by using different types of surface bound phenomena as diffusio-osmosis. For example, the latter requires solute concentration gradients with precise local control in a reversible way and at arbitrary surface. In this study, we use a local light-driven diffusion-osmosis (l-LDDO), where it is possible to manipulate the motion of micro particles reversibly and at any surface. The motor of the mechanism is a photosensitive azobenzene surfactant that present photo-isomerization between more hydrophobic trans and hydrophilic cis isomer. When porous microparticles are immersed in aqueous azobenzene surfactant solution, particle motion can be manipulated during light stimulation of appropriated wavelength.

Under external pumped fluid flow the local-LDDO motion is so strong that it provides a light induced velocity gain of porous particle along a streamline or it can be completely reversed by simply changing the wavelength into full motion stop of such particles against an external pumped fluid flow. Thus, with one stimulus we are able to manipulate the particle motion in both directions. Here we investigate stopping of the microparticles with studying the strength of local-light-driven diffusio-osmosis (l-LDDO) flow.

O 4.3 Mon 11:00 MA 005

Phase-Resolved, Wide-Field Sum-Frequency Generation Microscopy of Molecular Monolayers — •Ben John, Alexander Fellows, Tuhin Khan, Martin Wolf, and Martin Thämer — Fritz Haber Institut der MPG, Berlin, Germany

Inhomogeneous molecular assemblies at interfaces play an outstanding role in nature and industry e.g., in biological membranes, or lab-on-a-chip devices. Gaining experimental access to molecular compositions, arrangements, and packing is therefore of great scientific interest, however this has proven difficult. In particular, the elucidation of molecular conformations and the distribution of molecular orientations typically remains infeasible. Phase-Sensitive Vibrational Sum-Frequency-Generation (vSFG) microscopy can in principle yield such insight into molecular structures owing to its sensitivity to molecular directionality. However, vSFG microscopy still faces substantial technical obstacles, particularly its low signal-to-noise, which highly limits its application to either very thick films or those on metal substrates.

In this contribution, we introduce a novel vSFG microscope design and imaging system that overcomes these limitations. These technical advancements yield highly improved signal-to-noise ratios and a simplified experimental procedure, allowing for the first measurements of molecular monolayers on dielectric substrates. With measurements on phospholipid assemblies, we demonstrate that we can not only detect the molecular species in such monolayers but also characterize molecular compositions and fully determine the spatial distribution of their orientations.

O 4.4 Mon 11:15 MA 005

Energy Distributions of Material Sputtered under Swift Heavy Ion Bombardment — Tobias Heckhoff, •Lars Breuer, Marika Schleberger, and Andreas Wucher — Universität Duisburg-Essen, Fakultät für Physik and CENIDE, Duisburg, Germany

Ion-surface interactions represent a well-established field of research, serving as the foundation for numerous analytical and modification techniques. Despite decades of investigation, certain fundamental questions remain unanswered. These unresolved inquiries include phenomena like the secondary ion formation and the velocity distributions of particles sputtered during swift heavy ion bombardment.

While conventional laboratory-based ion sources can be used to determine the energy distributions of sputtered material by using short ion pulses, this approach is inadequate for prolonged pulses, as delivered by particle accelerators like the UNILAC at GSI.

We have developed a new measurement protocol that has been successfully applied in experiments involving various target materials, including metals, insulators, and organic molecules. We found that the energy distributions show a behavior where in all cases the maximum of the distribution is shifted to smaller energies and follows a different shape compared to the Thompson distribution of material sputtered under nuclear sputtering conditions.

O 4.5 Mon 11:30 MA 005

Quantifying monolayer coverage of endofullerenes by x-ray absorption spectroscopy from equivalent implanted ion doses — •W.C Lee[1], L. Yu[1], J. Oscarsson[2], M. W. Ochapski[3], R. Sagehashi[1], Y. Zhang[4], A. A. Popov[4], Z. M. Gebeyehu[3], L. Martini[3], S. Forti[3], C. Coletti[3], B. Delley[5], M. Muntwiler[5], D. Primetzhofer[2], and T. Greber[1] — [1]Physik-Institut, Universität Zürich, Switzerland — [2]Tandem Laboratory, Uppsala University, Sweden — [3]Istituto Italiano di Technologia, Pisa, Italy — [4]Leibniz-Institute, Dresden, Germany — [5]Paul Scherrer Institute, Switzerland

With x-ray absorption spectroscopy (XAS), the structure of magnetic atoms in molecules may be accessed directly [1]. For $Ho_3N@C_{80}$ endofullerenes on graphene/SiO_2, we calibrated the surface coverage by comparing the molecular XAS signal with implanted Ho ion doses of 3×10^{14} cm^{-2} that are directed to SiO_2 with energies between 2 and 115 keV. The corresponding Ho M_{45} intensity as a function of implantation depth d can be extrapolated to the zero-depth intensity of absorbed molecules. The Ho signal attenuation scales with exp(-d/Λ), with $\Lambda=10.3\pm0.7$ nm. The Si K-edge signal attenuation reveals an influence of the implantation process on the Si density. The XA spectra indicate trivalent Ho for both molecules and implants, while spectral differences and comparison to multiplet theory [2] suggest an isotropic $J=8$ Ho 4f electron distribution for implants and $J_z=\pm8$ ground states in the molecules due to the defined intramolecular ligand field.

[1] R. Westerström, et al., Phys. Rev. Lett., 087201 (2015)[2] A. Uldry, et al., Phys. Rev. B, 85, 125133 (2012)

O 4.6 Mon 11:45 MA 005

Autonomous nanoARPES Experiments — •Steinn Ýmir Ágústsson[1], Alfred J. H. Jones[1], Davide Curcio[1], Søren Ulstrup[1], Jill Miwa[1], Davide Mottin[2], Panagiotis Karras[2], and Philip Hofmann[1] — [1]Dpt. of Physics and Astronomy, Aarhus University — [2]Dpt. of Computer Science, Aarhus University

Angle-resolved photoemission spectroscopy (ARPES) is a technique used to map the occupied electronic structure of solids. Recent progress in X-ray focusing optics has lead to the development of ARPES into a microscopic tool, permitting the electronic structure to be mapped across the surface of a sample. This comes at the expense of a time-consuming scanning process to cover the whole surface.

We implemented a protocol which leverages Gaussian Processes to autonomously search the surface area in order to find positions of particular interest, based exclusively on the observed spectra. The protocol promises significant efficiency gains by avoiding redundant measurements and maximizing information gain from the data already measured. Furthermore, it can easily be expanded to explore a larger parameter space, including temperature or external perturbations.

The autonomous experimental control is implemented on the SGM4 micro-focus beamline of the synchrotron radiation source ASTRID2, where pilot experiments were used to quickly identify regions of interest to study in further detail. The successful implementation of the protocol shows the value of machine learning in the context of controlling complex experiments, and the potential for further development of autonomous experiments.

Topical Talk O 4.7 Mon 12:00 MA 005

Focused and coherent X-rays for the study of thin films and surfaces — •Ivan Zaluzhnyy — Institute of Applied Physics, University of Tübingen, Germany

X-ray diffraction techniques have been extensively used to study the atomic structure of bulk materials and thin films. The rise of new bright synchrotron X-ray sources opens up a possibility to use coherent X-ray diffraction to investigate the structure and morphology of surfaces and thin films. X-ray photon correlation spectroscopy (XPCS) is one of such techniques, which utilizes coherent X-ray beams to study the equilibrium as well as non-equilibrium dynamics of the film surface.

The grazing incidence geometry, which is typically employed to be surface-sensitive, puts certain requirements on the experimental parameters, such as X-ray beam size and sample alignment. This talk will give a few examples of XPCS studies of the thin film surfaces and show recent results on the kinetics and dynamics during in situ growth of organic films [1].

We thank colleagues from University of Tübingen, DESY and ESRF for their contributions.

[1] I. Dax et al., *New J. Phys.* **23** (2023) 103033

O 5: Ultrafast Electron Dynamics at Surfaces and Interfaces I

Time: Monday 10:30–12:30 Location: MA 041

O 5.1 Mon 10:30 MA 041

Electronic Structure and Carrier Dynamics of the Bulk Semiconductor Magnet CrSBr — •Lawson Lloyd[1], Tommaso Pincelli[1,2], Túlio de Castro[1], Ferdinand Menzel[3], Andreas Stier[3], Nathan Wilson[3], Martin Wolf[1], Laurenz Rettig[1], and Ralph Ernstorfer[1,2] — [1]Fritz Haber Institute, Berlin, Germany — [2]TU Berlin, Berlin, Germany — [3]TU Munich, Garching, Germany

Van der Waals (vdW) layered magnets have the potential to enable novel optoelectronic and spintronic applications. Among these, CrSBr is a direct band gap semiconductor that hosts interlayer antiferromagnetic order, a highly anisotropic, quasi-1D electronic structure, and strongly bound excitons. However, understanding how the carrier and exciton dynamics couple to the underlying spin order is currently lacking. Here, we employ time- and angle-resolved photoemission spectroscopy to map the temperature-dependent band structure and carrier dynamics in bulk CrSBr. We observe an electronic band splitting that emerges below the magnetic transition temperature, which we interpret as arising from a super-exchange mechanism giving rise to intralayer ferromagnetism. Time-resolved measurements reveal a rapid band renormalization of the lower conduction band that occurs during photoexcitation, pointing towards complex many-body effects governing the excited state dynamics and optical properties. These results provide important experimental observations of the low-temperature electronic band structure and shed further light on the microscopic interactions driving carrier dynamics and spin order in this vdW magnet.

O 5.2 Mon 10:45 MA 041

The wandering spins of Fe_3GeTe_2: itinerant vs localized behaviour in the electronic structure — •Tommaso Pincelli[1,2], Tania Mukherjee[1,2], Lawson Lloyd[2], Jyoti Krishna[3], Túlio de Castro[2], Shuo Dong[4], Victoria Taylor[2], Yoav William Windsor[1,2], Martin Wolf[2], Laurenz Rettig[2], and Ralph Ernstorfer[1,2] — [1]Institute of Optics and Atomic Physics, Technische Universität Berlin, Berlin, Germany — [2]Fritz Haber Institute of the Max Planck Society, Berlin, Germany — [3]Max-Born-Institut (MBI), Berlin — [4]Institute of Physics, Beijing National Laboratory for Condensed Matter Physics, Beijing, China

Iron Germanium Telluride (FGT) is a van der Waals metallic ferromagnet that has shown a wealth of exotic phenomena, ranging from 2D ferromagnetism with gate-tunable Curie temperature to heavy fermion behaviour. Despite its interest, its electronic structure and the microscopic interactions giving rise to magnetic order are still poorly understood: evidence exists of both itinerant and localized magnetic behaviour, an anomaly in a purely 3d electronic system. By employing time-resolved angle-resolved photoemission spectroscopy (trARPES), we observe clear evidence of the closure of the Stoner exchange gap, a clear indication of an itinerant character of the electronic excitations. This is in contrast with previous results of quasi-equilibrium studies, which ascribed FGT demagnetization to localized excitations. We also observe, by employing frequency domain ARPES, the impact of phononic excitations, resulting in an A_{1g} phonon strongly coupled with the bandstructure at the Brillouin zone centre.

O 5.3 Mon 11:00 MA 041

Electron Dynamics in the Conduction Band of the van-der-Waals Magnet $Cr_2Ge_2Te_6$ — •Stephan Schmutzler, Jan Böhnke, Martin Weinelt, and Cornelius Gahl — Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, 14195 Berlin, Germany

The electron dynamics in the central region of the Brillouin zone of $Cr_2Ge_2Te_2$ has been investigated by time-resolved ARPES. Optical excitation with a photon energy of 1.55 eV leads to a transient broadening of the valence band and simultaneously excites an electron population located at 0.7-1.3 eV above the Fermi level. Within 1 ps, this population continuously relaxes towards states at 0.5 eV. Considering their long lifetime of >3 ps, we attribute these states to the conduction band minimum. Our findings are in contrast to theoretical predictions that locate the minimum close to the $\overline{\mathrm{K}}$-point of the surface Brillouin zone [1,2]. The broad momentum distribution in the conduction band suggests enhanced scattering in the excited material.

[1] D.-Y. Wang et al. Phys. Rev. B 107, 125148 (2023)

[2] Y. F. Li et al., Phys. Rev. B 98, 125127 (2018)

O 5.4 Mon 11:15 MA 041

Ultrafast charge and spin response to linear and circular polarized laser excitations in naturally layered delafossites $PdCoO_2$ and $PtCoO_2$ from time-dependent DFT

— •Mike Bruckhoff, Markus E. Gruner, and Rossitza Pentcheva — Faculty of Physics and Center of Nanointegration, CENIDE, University of Duisburg-Essen

In the framework of real-time time-dependent density functional theory, we investigate the layer-resolved, ultrafast electronic dynamics after laser excitation of the non-magnetic, metallic delafossites $A\mathrm{CoO}_2$ A = Pd, Pt. We simulate the responses to linear and circular polarized laser pulses with multiple laser frequencies and laser fluences. We observe a marked difference in the redistribution of atomic charges regarding in-plane vs. out-of-plane linear polarized laser pulses, which results in a charge transfer from the A-layer into the CoO_2-layer and vice versa depending on the laser frequency and polarization direction. In addition we encounter for $PdCoO_2$ low-frequency charge oscillations within the CoO_2-layer which occur after the decay of the laser pulse. These oscillations are accompanied by significant transient spin magnetic moments reaching values up to $0.08\mu_\mathrm{B}$. Circular polarized laser pulses excite these low-frequency charge and spin oscillations in both delafossites using less intense laser pulses.

Funding by DFG within CRC1242 is gratefully acknowledged.

O 5.5 Mon 11:30 MA 041

Ultrafast lattice dynamics of $MoSe_2$ monolayers

— •Victoria C. A. Taylor[1], Yoav W. Windsor[1,2], Elizaveta Patutkina[2], Hyein Jung[1,2], Jannik Malter[1], Mustafa Hemaid[3], Tobias Korn[3], and Ralph Ernstorfer[1,2] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — [2]Technische Universität Berlin, Berlin, Germany — [3]Institut für Physik, Universität Rostock, Rostock, Germany

Transition metal dichalcogenides (TMDCs) are a promising family of 2D materials, which exhibit rich electronic phenomena. In particular, several notable TMDCs, such as $MoSe_2$, exhibit an indirect to direct band gap transition when reduced to monolayer thicknesses. Although extensive studies have investigated ultrafast electronic properties of TMDC monolayers, comparatively few have directly addressed the lattice response to photoexcitation.

Femtosecond electron diffraction (FED) is an ideal tool to study photoexcited lattice dynamics, as it provides quantitative information on coherent and incoherent atomic vibrations. Such information enables a direct understanding of energy flow from the electronic subsystem to the lattice via electron-phonon coupling, and within the lattice via phonon-phonon relaxation. We present FED measurements of $MoSe_2$ monolayers under different excitation conditions, in particular at the A-exciton resonance. As such, the results enable direct observation of the behaviour of phonons under ultrafast "nonthermal" conditions, i.e. those in which Bose Einstein statistics are not maintained.

O 5.6 Mon 11:45 MA 041

GHz manipulation of low-energy electron pulses

— •Dennis Epp[1,2], Benjamin Schröder[1,2], Marcel Möller[1,2], and Claus Ropers[1,2] — [1]Department of Ultrafast Dynamics, Max Planck Institute for Multidisciplinary Sciences, 37077 Göttingen, Germany — [2]4th Physical Institute - Solids and Nanostructures, University of Göttingen, 37077 Göttingen, Germany

Electron pulses are a powerful probe in time-resolved diffraction and microscopy. One of the main challenges in such experiments is the dispersion-induced pulse broadening, both due to Coulomb interactions and the initial photoelectron energy distribution [1]. One technique to overcome this challenge is radio-frequency (RF) pulse compression, which is routinely used to control high-energy electron pulses [2]. However, corresponding schemes have not been developed for energies below a few kilo-electronvolts.

In this contribution, we demonstrate longitudinal phase-space manipulation of low-energy electron pulses using RF fields. Specifically, we combine a millimetre-sized photoelectron gun [3] with a synchronized compression cavity driven at a frequency of 2.5GHz. The change in pulse duration induced by the cavity is measured by the deflection of transient space-charge cloud at a metal grid, resulting in a two- to four-fold compression of 80-,100- and 120-eV electron pulses [4].

[1] Dwyer, et al., Phil. Trans. R. Soc. 364,741-778 (2006). [2] Kassier, et al., Applied Physics B 109, 249-257 (2012). [3] Vogelgesang, et al., Nature Physics 14,184-190 (2018). [4] Epp et al., under review.

O 5.7 Mon 12:00 MA 041

Ultrafast Low-Energy Photoelectron Diffraction for the Study of Adsorbate-Surface Interactions with 100 fs Temporal Resolution

— •Hermann Erk, Carl Jensen, Stephan Jauernik, Petra Hein, and Michael Bauer — Christian-Albrechts-Universität Kiel, Germany

Ultrafast Low-Energy Electron Diffraction (ULEED) is a promising tool for studying the structural dynamics of ordered adsorbate layers following photoexcitation with femtosecond light pulses. However, the temporal spread of the probing electron pulse due to Coulomb interaction while it propagates toward the sample surface substantially limits the time resolution of this technique so far [1].

In this contribution, we present an alternative experimental scheme based on time- and angle-resolved ARPES that uses the photoelectrons generated in the substrate and diffracted by the adsorbate overlayer on the way to the electron detector to monitor ultrafast adsorbate structural dynamics. For tin-phthalocyanine adsorbed on single-crystalline graphite, we show that the intrinsic time resolution of the photoemission experiment of 100 fs is preserved in the diffraction process by the adsorbate overlayer. On a few ps timescale, we identify a transient and distinct decrease in the photoelectron diffraction intensity that we attribute to an increase in the adsorbate superlattice temperature due to phononic coupling with the substrate. The interpretation is substantiated by a comparison with phonon-temperature transients for graphite simulated based on a three-temperature model.

[1] S. Vogelgesang, et al. Nature Physics 14, 184 (2018)

O 5.8 Mon 12:15 MA 041

Development and characterization of an ultrafast scanning electron microscope

— •Paul H. Bittorf[1] and Nahid Talebi[1,2] — [1]Institute for Experimental and Applied Physics, Kiel University, Leibnizstraße 19, D-24118 Kiel, Germany — [2]Kiel Nano, Surface and Interface Science KiNSIS, Kiel University, Christian-Albrechts-Platz 4, D-24118 Kiel, Germany

A variety of processes are involved within the interaction of free-electron wave packets with light or matter, such as coherent and incoherent excitation of the investigated material. Understanding these interaction processes of pulsed electron beams with shaped light fields or nanostructured matter is crucial to achieve a favored shaping of electron wave packets, as well as exploring the dynamics of material excitations. Therefore, we developed an ultrafast scanning electron microscope (USEM) for exploring the dynamics at the nanoscale and at the femtosecond time regime. Our setup is based on the combination of a commercial SEM with an ultrafast laser system, where a laser-driven pulsed electron source is achieved via the photoemission process. In addition to the electron beam excitation, a time-delayed laser pulse is focused onto the sample to induce an optical near-field and implement a time-resolved pump-probe measurement. The interaction properties of electrons with matter are analyzed through the emitted light, either cathodoluminescence (CL) or photoluminescence. Here, we report on the technical aspects of the development and characterization of our USEM setup and present measurements of the photoemitted pulsed electron beams and a novel fiber-based CL detector.

O 6: Organic Molecules on Inorganic Substrates I: Adsorption & Growth

Time: Monday 10:30–13:00 Location: MA 042

O 6.1 Mon 10:30 MA 042

Unraveling Glycan Structures: ESIBD+STM Exploration of Stapled Conformations — •Dhaneesh Kumar[1], Kelvin Anggara[1], Manuel G. Ricardo[2], Peter H. Seeberger[2], and Klaus Kern[1,3] — [1]Max-Planck-Institute for Solid State Research, Stuttgart, Germany — [2]Max-Planck-Institute of Colloids and Interfaces, Potsdam, Germany — [3]École Polytechnique Fédérale de Lausanne, Switzerland

”Function follows form” holds a particular significance in the realm of biomolecules, where structural modifications can profoundly influence functionality. Recently, introduction of linkages or ’staples’ at specific sites within a linear oligosaccharide (a.k.a. glycan) has been demonstrated to alter the biochemical properties of the glycan (1). Despite these advancements, the structural effect of these staples on glycan conformation has yet to be probed directly, leaving the bioactive conformations unknown.

Using Electrospray Ion Beam Deposition (ESIBD), we successfully transferred these stapled glycans onto a surface in vacuum, which are subsequently imaged individually by Scanning Tunnelling Microscopy (STM). By systematically varying factors such as stapling length and sites, we can visualize the possible conformational states of these stapled glycans. This integrated ESIBD+STM approach provides a complementary tool for studying biomolecular conformational states at the single-molecule level, overcoming limitations of conventional techniques.

(1) Ricardo M. G., et al.,J. A. Chem. Soc. (2022)

O 6.2 Mon 10:45 MA 042

Unlocking non-volatile molecular building blocks for atomically clean vacuum depositions — •Andreas Walz[1,2], Annette Huettig[1,2], Michael Walz[1,2], Hartmut Schlichting[1,2], and Johannes V. Barth[1] — [1]Technical University of Munich, Physics E20, Garching, Germany — [2]pureions GmbH, Gilching, Germany

Novel nanostructures of organic and anorganic compounds require precise growth of clean films on well-defined surfaces. Their fabrication is technically limited: Thermal evaporation in vacuum (MBE, OMBE) is constrained to volatile species. Wet/ solution-based techniques (drop casting, spin coating, inkjet printing) are broadly applicable, but often lack purity and quality.

Controlled Ion Beam Deposition (CIBD), realized in a NanoPrinter device from pureions, combines the best of both: a broad range of fragile species under ultra-clean conditions enabled by Electrospray Ionization (ESI) e.g., functionalized small molecules (e.g. linkers for MOFs), graphene nanoribbons, up to large biomolecules like proteins or DNA. Other ion sources such as MALDI, laser vaporization or magnetron sputtering cluster sources are also considered. Transfer of the ions to ultra-high vacuum (UHV) and mass filtering via a digital quadrupole mass filter (dQMF) removes all unwanted neutral or ionic impurities. Soft-landing or if wished, reactive-landing for on-surface chemistry, is both ensured by control of energy and dose. CIBD is a new tool providing functional nanostructures for all kinds of applications, e.g. in organic electronics / photovoltaics / LEDs / sensors / transistors, nanocatalysis, energy storage, molecular packaging, biotech, pharma.

O 6.3 Mon 11:00 MA 042

Growth of ordered cobalt phthalocyanine monolayers on a terbium modified silicon surface — •Milan Kubicki, Martin Franz, and Mario Dähne — Technische Universität Berlin, Institut für Festkörperphysik, Berlin, Germany

The formation of self-assembled layers of organic molecules on solid surfaces is an important subject because of their possible application in advanced optical and electronic devices. While the formation of self-assembled layers is well established on metal surfaces, the growth on silicon surfaces is much less studied.

On pure silicon surfaces the number of dangling bonds leads to a highly reactive surface and often to rather disordered organic films. The modification of silicon surfaces with thin atomic layers is an interesting alternative. Here, a metallic rare earth silicide monolayer is used as a substrate for the molecular growth.

Using scanning tunneling microscopy and spectroscopy the molecular arrangement and the electronic properties of cobalt phthalocyanine (CoPc) molecules on the $\mathrm{TbSi_2/Si(111)}$ surface are studied. CoPc belongs to the transition metal phthalocyanines being a class of organic semiconductors, which has been already employed e.g. in organic light emitting diodes, photovoltaic cells, and field-effect transistors.

Our data demonstrate that single CoPc molecules stay mobile at the $\mathrm{TbSi_2/Si(111)}$ surface at room temperature. By reaching a coverage of a full monolayer a highly ordered film of flat-lying CoPc molecules in different orientations is formed, demonstrating the great potential of rare earth silicides on Si(111) as template for organic film growth.

O 6.4 Mon 11:15 MA 042

Identifying the Surface Arrangement of a Mixed Zinc Tetraphenylporhyrin and Monocarboxyphenyl Triphenylporphyrin Layer on Rutile TiO2(110) Using XPS — •Maximilian Muth, Alexander Wolfram, Hans-Peter Steinrück, and Ole Lytken — Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany

Gaining deeper knowledge about the adsorption behavior of organic molecules on substrate surfaces is crucial for possible applications. One approach to learn about the (relative) adsorption strength of different molecules is to subsequently adsorb different molecules on top of each other at low temperature, and study the preferential displacement of the weaker bound species from the surface upon heating. Using this method, we have identified the relative adsorptions strengths of three tetraphenylporphyrins (ZnTPP, CoTPP, MgTPP) on rutile TiO2(110).[1] Moreover, we have recently addressed layers of these non-functionalized molecules and the carboxylic-acid-functionalized tetraphenylporphyrin (2HMCTPP), which is expected to bind more strongly to the surface. Interestingly, instead of completely displacing the non-functionalized molecules, we found 2HMCTPP to adsorb upright standing on free adsorption sites in between the flat-lying non-functionalized molecules.

[1] Muth et al., Accurate Determination of Adsorption-Energy Differences of Metalloporphyrins on Rutile TiO2(110) 1x1, Langmuir 2022, 38, 28, 8643-8650

O 6.5 Mon 11:30 MA 042

On-surface synthesis of well-aligned pentacene arrays and graphdiyne nanowires on 1D-patterned VSe2 — •Zhongliu Liu[1], Ignacio Piquero-Zulaica[1], Harald Brune[2], and Johannes Barth[1] — [1]Physics Department E20, Technical University of Munich, Germany — [2]Institute of Physics, EPF Lausanne, Switzerland

On-surface synthesis of one-dimensional (1D) polymers is a promising method for creating well-ordered nanostructures, such as graphene nanoribbons and graphdiyne nanowires. Vanadium diselenide (VSe2), as a versatile member from the transition-metal-dichalcogenide family, not only exhibits unique characteristics like ferromagnetism and excellent electrocatalysis, but also presents a unique 1D-patterned structure upon annealing. The pertaining 1D patterns of Se-deficient furrows could influence the positioning of adsorbed molecules and modify their electronic structure, guiding the on-surface synthesis of new organic entities. Here, we present the formation of oriented arrays of pentacene and diethynyl-terphenyl (DETP) 1D polymers on 1D-patterned VSe2. Pentacene arraying along the substrate pattern exhibits a reduced bandgap of approximately 1.60 eV. And for DETP arrangements on the 1D pattern, upon annealing, homocoupling reactions of terminal alkyne tectons entail the formation of oriented nanowires, some of which extend up to 30 molecular units in length. The resulting graphdiyne nanowires show a small energy shift in their HOMO. These findings highlight 1D-patterned VSe2 as a template for modulating the structural and electronic properties of organic molecules, providing a new approach for on-surface synthesis of organic polymers.

O 6.6 Mon 11:45 MA 042

Selectivity in photo- and thermal-induced trans-cis isomerization in the ultra-thin film of azobenzene derivatives on graphite — •Hariom Birla[1], Showkat H Mir[2], Thomas Halbritter[3], Alexander Heckel[4], Jayant K. Singh[5], and Thiruvancheril G. Gopakumar[6] — [1]Department of Chemistry, Indian Institute of Technology Kanpur, Kanpur, UP-208016, India — [2]Department of Chemical Engineering, Indian Institute of Technology Kanpur, Kanpur, UP-208016, India — [3]Eurofins Genomics Germany GmbH, Anzinger Str. 7a, 85560, Ebersberg, Germany — [4]Institute for Organic Chemistry and Chemical Biology, Goethe-University Frankfurt, Max-von-Laue-Str.9, 60438 Frankfurt, Germany — [5]Department of Chemical Engineering, Indian Institute of Technology Kanpur, Kanpur, UP-208016, India — [6]Department of Chemistry, Indian Institute of Technology Kanpur, Kanpur, UP-208016, India

Azobenzene (AB) and its derivatives are known for isomerization between its geometrical isomers using light, temperature, electrons/holes, and electrical field on surfaces. The derivatives of AB could be used as an electronic switch as there is a significant change in the electronic properties upon isomerization (trans-cis). In this work, we show the trans-cis isomerization in the ultra-thin films of azobenzene derivatives on a highly oriented pyrolytic graphite (HOPG)-air interface. The microscopic structure and the on-surface isomerization are investigated using atomic force microscopy (AFM). The trans-to-cis isomerization is induced by a photon of wavelength 360 nm whereas the cis-to-trans isomerization is induced by thermal excitation.

O 6.7 Mon 12:00 MA 042

Density functional theory investigation of Porphine adsorption on the BC monolayer — •Pinar Kaya[1], Çağil Kaderoğlu[1], Ethem Aktürk[2], and Handan Arkin[1] — [1]Department of Physics Engineering, Faculty of Engineering, Ankara University, 06100 Ankara, Turkey — [2]Department of Physics, Adnan Menderes University, 09100, Aydin, Turkey

In recent years, developments in technology have increased the importance of multifunctional, nano-sized, cheap and accessible materials. The first and foremost of these materials is the 2-dimensional (2D) graphene structure [1]. The unique physical, chemical and mechanical properties of 2D graphene have led researchers to investigate new surfaces [2]. In this context, numerous boron-doped graphene (BC_x) structures, whether uniform or non-uniform, have been examined experimentally and theoretically in the literatüre [3,4]. There are very few studies in the literature on BC monolayer, which has not yet been synthesized. In this aim, our study presents first-principle density functional theory calculations for the structural, electronic and magnetic properties of Porphine molecule on the bare doped BC monolayer surface. It is thought that the new functional structures created as a result of the findings will pioneer different application areas. [1] Novoselov, K. S., Geim, A. K., Morozov, S. V., ... & Firsov, A. A. (2004), 306(5696), 666-669. [2] Zhang, H. (2018), 118(13), 6089-6090. [3] Durajski, A. P., & Kasprzak, G. T. (2023), Physica B: Condensed Matter, 660, 414902. [4] Bute, A., Jena, S., Kedia, S., Udupa, D. V., ... & Sinha, S. (2021), Materials Chemistry and Physics, 258, 123860.

O 6.8 Mon 12:15 MA 042

Combining experimentally-measured and DFT-predicted XPS spectra to identify the bond of phenylphosphonic acid to rutile TiO2(110) — •Alexander Wolfram[1], Maximilian Muth[1], Julia Köbl[1], Sascha Mehl[2], Nataliya Tsud[3], Hans-Peter Steinrück[1], Bernd Meyer[1], and Ole Lytken[1] — [1]Uni Erlangen, Germany — [2]Elettra, Italy — [3]Charles Uni, Czech Republic

Anchoring groups, such as carboxylic or phosphonic acids, are common tools to immobilize organic molecules on oxide surfaces and understanding these interfaces is crucial for the optimization of devices like dye-sensitized solar cells. Because the bond to the surface occurs through the oxygen atoms, the O 1s core level region in XPS contains important information about the nature of the bond. However, it is extremely challenging to disentangle the O 1s contribution of the adsorbate from that of the oxide support, as the contribution of the support can significantly change upon adsorption of molecules. We will show how DFT calculations of phenylphosphonic acid on rutile TiO2(110) can be used to predict the full experimentally-measured XPS spectra, based on calculated O 1s binding energies of the oxygen atoms of the adsorbed phenylphosphonic acid molecules as well as the oxygen atoms within the first three trilayers of the surface. The calculated spectra accurately account for both the O 1s contributions of the adsorbed molecules, as well as the change to the O 1s contribution of the rutile TiO2(110) substrate upon adsorption of the molecules.

O 6.9 Mon 12:30 MA 042

“Organic” ab initio thermodynamics for interfaces: Which corners can we (not) cut? — •Christoph Wachter and Oliver T. Hofmann — Institute of Solid State Physics, Graz University of Technology, Graz, 8010, Austria

Ab initio thermodynamics is a powerful tool to computationally predict the most stable structure under given environmental conditions, i.e. at finite temperature and pressure. In principle, this requires evaluating the free energy for a large number of structures, which is a costly endeavor. To keep the computational cost tractable, the current paradigm is to approximate the free energy and neglect several expensive terms, such as the vibrational zero-point energy, the thermal occupation of vibrations and the configuration entropy.

However, these approximations have been developed with simple inorganic adsorbates in mind. It is a priori unclear whether they also hold true for organic/inorganic interfaces, which feature a rich chemistry. Moreover, several effects can occur in these systems (such as cis-trans isomery) that are hardly present for inorganic adsorbates. In this work, we therefore re-evaluate the most common approximations for three conceptually very different systems: a clearly physisorbed system, a covalently bonded self-assembled monolayer, and an interfaces that undergoes charge-transfer reactions. In an attempt to generalize across the different types of interfaces, we discuss which calculations can generally be skipped in order to allow efficient evaluations of the relative stabilities of different phases.

O 6.10 Mon 12:45 MA 042

Understanding the role of adsorbed CTAB in anisotropic nanostructure growth — •Esmée Berger[1], Narjes Khosravian[1], Joakim Löfgren[2], and Paul Erhart[1] — [1]Department of Physics, Chalmers University of Technology, Gothenburg, Sweden — [2]Department of Applied Physics, Aalto University, Espoo, Finland

Surfactants have immense technological relevance. Cetyltrimethylammonium bromide (CTAB) is, for example, commonly used for structural tuning and increased stability during wet-chemical synthesis of nanostructures. The functionality of surfactants is dictated by their phase diagrams, which are often very complex in aqueous solution. The complexity of these systems is further increased during nanostructure growth, due to the presence of surfaces. To improve control over the shapes and sizes of wet-chemically synthesized nanostructures, we must understand the functionality, and thus the structure, of the adsorbed surfactants as these fundamentally alter the nature of the interface, affecting the growth. Phase diagrams are, however, very difficult to probe. To get some insight into the role of surfactants during nanostructure growth we consider CTAB, whose micellar phase has been pointed out as the primary mechanism for anisotropic growth of inorganic nanoparticles. Those results were, however, based on single values of CTAB surface density. To elucidate the role of CTAB in this context, a surface phase diagram must be established. In this work, we use atomistic simulations to develop an understanding of this surface phase diagram from a modeling perspective.

O 7: Plasmonics and Nanooptics I

Time: Monday 10:30–13:00 Location: MA 043

O 7.1 Mon 10:30 MA 043

Plasmonic Twistronics: Discovery of Plasmonic Skyrmion Bags — •Julian Schwab[1], Alexander Neuhaus[2], Pascal Dreher[2], Shai Tsesses[3], Anant Mantha[1], Florian Mangold[1], Bettina Frank[1], Guy Bartal[3], Frank-J. Meyer zu Heringdorf[2], Timothy J. Davis[1,2,4], and Harald Giessen[1] — [1]4th Physics Institute, Research Center SCoPE, and Integrated Quantum Science and Technology Center, University of Stuttgart, Germany — [2]Faculty of Physics and Center for Nanointegration, University of Duisburg-Essen, Germany — [3]Andrew and Erna Viterbi Department of Electrical Engineering, Technion-Israel Institute of Technology, Israel — [4]School of Physics, University of Melbourne, Australia

We explore the application of twistronics in plasmonic systems by superimposing two plasmonic skyrmion lattices to create a moiré skyrmion superlattice. We combine this concept with plasmonic topological excitations and demonstrate that the topology of moiré skyrmion lattices contains skyrmion bags as complex topological quasiparticles that so far have been demonstrated only in liquid crystals and chiral ferromagnets. The size of plasmonic skyrmion bags can be controlled by the twist angle and its center of rotation. The resulting electric field distribution of a skyrmion bag is derived numerically and verified experimentally using time-resolved two-photon photoemission electron vector microscopy (PEEM).

O 7.2 Mon 10:45 MA 043

Photoelectron imaging of topological edge states in one-dimensional plasmonic Su–Schrieffer–Heeger chains — •Luisa Brenneis[1], Benedikt Schurr[2,3], Matthias Hensen[1], Philipp Kessler[1], Bert Hecht[2,3], and Tobias Brixner[1] — [1]Institut für Physikalische und Theoretische Chemie — [2]NanoOptics Biophotonics Group, Experimental Physics 5 — [3]Würzburg-Dresden Cluster of Excellence ct.qmat, [1-3]: Universität Würzburg, Am Hubland, 97074 Würzburg, Germany

Plasmonic nanostructures exhibiting different topological phases are proposed to offer unique properties like energy flow along the phase separating interfaces [1] or localized edge states [2]. Here, we use photoemission electron microscopy (PEEM) to directly image the mode pattern of one-dimensional nanochains with trivial and nontrivial topology, representing a plasmonic analogue of a Su–Schrieffer–Heeger chain [3]. To ensure sufficient electron delivery for the photoemission process we exploit Babinet's principle by investigating nanoslit chains written in monocrystalline gold flakes. The precision of the helium ion beam milling used for this purpose enables us to produce the individual chain elements with a distance down to 8 nm. Therefore, we can precisely control the dipole–dipole coupling between the individual chain elements. By comparing the experimental results with finite-difference time-domain simulations we identify the presence of edge states.

[1] M. Proctor, Appl. Phys. Lett. 118, 091105 (2021).

[2] S. R. Pocock, ACS Photonics 5, 22712279 (2018).

[3] W. P. Su, Phys. Rev. B 22, 2099 (1980).

O 7.3 Mon 11:00 MA 043

Attosecond electron microscopy by free-electron homodyne detection — John H. Gaida[1,2], Hugo Lourenço-Martins[1,2], •Murat Sivis[1,2], Thomas Rittmann[1,2], Armin Feist[1,2], F. Javier García de Abajo[3,4], and Claus Ropers[1,2] — [1]MPI for Multidisciplinary Sciences, Göttingen, Germany — [2]4th Physical Institute, University of Göttingen, Germany — [3]ICFO-Institut de Ciencies Fotoniques, Castelldefels (Barcelona), Spain — [4]ICREA-Institució Catalana de Recerca i Estudis Avançats, Barcelona, Spain

Photon-induced near-field electron microscopy (PINEM) enables the imaging of near-field intensities [1,2]. However, access to the evolution of nanoscale fields and structures within the light cycle requires a sensitivity to the optical phase [3,4].

Here, we introduce Free-Electron Homodyne Detection (FREHD) for phase-resolved electron microscopy of optical responses [5], which we demonstrate for the plasmonic fields of a gold nanoprism with few-nanometer spatial and sub-cycle temporal resolutions.

Our approach generalizes the high-resolution measurement of attosecond materials responses in electron microscopy, without a need for electron density bunching, and offers fascinating new possibilities to image local attosecond and phase-resolved responses on the nanometer scale.

[1] B. Barwick, et al. Nature 462, 902 (2009). [2] L. Piazza et al. Nat Commun 6, 6407 (2015). [3] D. Nabben, et al. Nature 619, 63 (2023). [4] T. Bucher et al., arXiv:2305.04877 (2023). [5] J. H. Gaida, et al. arXiv:2305.03005 (2023).

O 7.4 Mon 11:15 MA 043

Ultrafast near-field scanning optical oscilloscopy — •Juanmei Duan, Tom Jehle, Sam Nochowitz, and Christoph Lienau — Universität Oldenburg, D-26129, Germany

Metallic, dielectric and hybrid nanoparticles offer exciting opportunities to localize, manipulate and switch light on the nanoscale. A direct measurement of the local electric field at the surface of the nanostructures is challenging however, since these fields are often localized on exceedingly short length and time scales. While experiments such as attosecond photoelectron emission microscopy or phase-resolved photon-induced near-field electron microscopy have been proposed, direct time-resolved measurements are still lacking. Here, we describe and demonstrate a new experimental technique, ultrafast near-field oscilloscopy, to probe coherent optical near-fields in the time with nanometer spatial resolution. For this, amplitude and phase of the local near-field scattered by a sharp metal taper are recorded in a broad spectral range and on a time scale that is faster than the tip modulation period. This allows us to record spectra as a function of tip-sample distance, the key to probe tip-sample coupling experimentally. Direct Fourier transform of the scattering spectra gives the local near-field dyanamics with sub-cycle temporal and nanometer spatial resolution. We demonstrate the versatility of this new approach by probing near-fields of dielectric and semiconducting nanoparticles, as well as different localized and propagating plasmon mode of metal nanostructures.

O 7.5 Mon 11:30 MA 043

Determination of the transient absorbance during ultrafast laser-matter interaction by applying ultrafast imaging ellipsometry — •Markus Olbrich, Pflug Theo, Lungwitz Philipp, and Alexander Horn — Laserinstitut Hochschule Mittweida, Hochschule Mittweida, Technikumplatz 17, 09648 Mittweida

Applying ultrafast imaging metrology combined with modeling the ablation represents a promising way to comprehend the interaction of ultrafast laser radiation with matter fundamentally. Accurately simulating the absorption of the laser radiation is challenging due to the complexity of the needed models and the temperature- and density-dependencies of all included parameters. To solve this, determining the absorbed energy by ultrafast imaging ellipsometry neglects the necessity of using an optical model.

In this study, a 150 nm thick gold film, including an adhesive layer of 25 nm chromium on a glass substrate is irradiated by single-pulsed ultrafast laser radiation (pump radiation: pulse duration $\tau_\mathrm{H} = 40\,\mathrm{fs}$, wavelength $\lambda = 800\,\mathrm{nm}$, peak fluence $H_0 = 5.0\,\mathrm{J/cm^2}$). Ultrafast imaging ellipsometry reveals the complex refractive index at the probe wavelengths 700 nm and 900 nm within 10 ps post-pump irradiation. The complex refractive index for the pump radiation at $\lambda = 800\,\mathrm{nm}$ is calculated by linear interpolation afterward. Subsequently, the absorbed energy of the pump radiation is derived and included in the model. The simulations incorporating the experimentally determined absorbed energy yield a much better fit to the experimental results than uncorrected simulations.

O 7.6 Mon 11:45 MA 043

Photon-Induced Near-Field Interaction in Ultrafast Point-Projection Electron Microscopy — •Germann Hergert[1], Andreas Wöste[1], Martin Silies[1], Dong Wang[2], Petra Gross[1], and Christoph Lienau[1] — [1]Institut für Physik, Carl-von-Ossietzky Universität, 26129 Oldenburg, Germany — [2]Institut für Werkstofftechnik, TU Ilmenau, 98693, Germany

Photon-induced electron microscopy (PINEM) utilizes the coherent interaction of swift electrons (10-100keV) with optical near-fields for high-resolution imaging of nano-sized systems. Transferring PINEM to significantly lower electron energies (<100eV) increases the interaction time between electrons and near-fields and may enhance coupling to nano-confined optical modes. However, this interaction has not been demonstrated due to high momentum mismatch at such low energies.

This study presents the first PINEM-like interactions in ultrafast point-projection electron microscopy (UPEM) with 100eV electrons [1]. Plasmonic nanofocusing generates 30fs electron pulses from a sharp gold taper that are accelerated by a -100V bias voltage towards an optically pumped Yagi-Uda antenna milled inside a 13nm thick free-standing gold film. The 3D momentum of the transmitted electrons is detected with a time-of-flight delay-line detector. A spread in the electron's momentum distribution is observed, originating from their coupling to both - longitudinal and transverse - electric field components of the antenna. This paves the way for spatial and temporal characterization of vectorial near fields in UPEM.

[1] Wöste, A. et al., Nano Lett. 23, 5528-5534 (2023)

O 7.7 Mon 12:00 MA 043

Spectroscopic and Interferometric Sum-Frequency Imaging of Strongly Coupled Phonon Polaritons in SiC Metasurfaces

— •Niclas S. Mueller[1], Richarda Niemann[1], Sören Wasserroth[1], Guanyu Lu[2], Martin Wolf[1], Joshua D. Caldwell[2], and Alexander Paarmann[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — [2]Department of Mechanical Engineering, Vanderbilt University, Nashville, USA

Phonon polaritons enable waveguiding and localization of infrared light with extreme confinement and low losses. Here, we introduce sum-frequency spectro-microscopy as a tool to image phonon polaritons in infrared metasurfaces with sub-wavelength spatial resolution and highly-resolved spectral resonance information.[1] This is implemented by resonantly exciting polaritons using a tunable infrared free-electron laser and wide-field microscopic detection of the upconverted light. We employ this technique to image hybridization and strong coupling of localized and propagating surface phonon polaritons in metasurfaces of SiC micropillars. Spectro-microscopy allows us to measure the polariton dispersion simultaneously in momentum space by angle-dependent resonance imaging, and in real space by polariton interferometry. Notably, we directly visualize how strong coupling affects the spatial localization of polaritons, inaccessible with conventional spectroscopic techniques. We further observe the formation of edge states at excitation frequencies where strong coupling prevents polariton propagation into the metasurface.

[1] R. Niemann, N.S. Mueller et al. arXiv 2311.13284 (2023)

O 7.8 Mon 12:15 MA 043

Highly polarization dependent formation of polaritons due to molecular alignment

— •Roland Schäfer, Philipp Weitkamp, Dirk Hertel, Klaus Meerholz, and Klas Lindfors — Department für Chemie, Universität zu Köln, 50939 Köln, Germany

Spin cast films of HB238 [1] aggregate, forming both a J- and H-like transition that have a high polarization dependence, because the J-transition is located in the substrate plane, while the H-transition is located in the surface normal. In our previous work we have shown that both transitions can be simultaneously strongly coupled to planar microcavites, forming four polaritons. The two H-polaritons are highly polarization dependent.[2]

By aligning the aggregates along one direction, we achieve strong polarization dependence for both the J- and H-transition, enabling us to control the strong coupling behavior of both transitions by polarization.

[1] Bürckstümmer, et al., *Angew. Chem. Int. Ed.* **2011**, 50: 11628-11632.

[2] Schäfer, et al., *ACS Photonics*, in press.

O 7.9 Mon 12:30 MA 043

On-Chip Strong Coupling Device based on a slit nanoresonator

— •Benedikt Schurr[1], Patrick Pertsch[1], Jin Qin[1], Saeid Asgarnezhadzorgabad[2], Alagesan Subramaniango[3], Monika Emmerling[1], Artur P. Podhorodecki[3], Ortwin Hess[2], and Bert Hecht[1] — [1]NanoOptics & Biophotonics Group, Experimental Physics 5, University of Würzburg, Am Hubland, 97074 Würzburg, Germany — [2]School of Physics and CRANN Institute, Trinity College Dublin, Dublin 2, Ireland — [3]Wroclaw University of Science and Technology, Department of Experimental Physics, wyb. Stanislawa Wyspianskiego 27, 50-370 Wroclaw, Poland

Strong coupling (SC) of light and matter states at ambient conditions is of interest in quantum technologies. In previous work, we demonstrated ambient condition SC between single quantum dots and a plasmonic slit resonator at a scanning probe tip [1,2]. To gain stability and scalability we have now developed an on-chip platform. We fabricate plasmonic slit resonators on a glass substrate located at the tip of a first electrode structure opposed by a second electrode. We then use dielectrophoresis to attract & position single quantum dots at the nanoresonator. This combination of techniques provides additional degrees of freedom as well as the opportunity to apply electric fields to tune the system.

[1] Groß, H. et al., Sci. Adv.4, eaar4906 (2018)

[2] Friedrich, D. et al., arXiv:2305.06909 (2023)

O 7.10 Mon 12:45 MA 043

Plasmon-mediated coherent population oscillations in molecular aggregates

— •Daniel Timmer[1], Moritz Gittinger[1], Thomas Quenzel[1], Sven Stephan[1], Yu Zhang[2], Marvin Schumacher[3], Arne Lützen[3], Martin Silies[1], Sergei Tretiak[2], Jin-Hui Zhong[1], Antonietta De Sio[1], and Christoph Lienau[1] — [1]University of Oldenburg, Germany — [2]Los Alamos National Laboratory, USA — [3]University of Bonn, Germany

Strong coupling between plasmonic nanostructures and excitonic quantum emitters is a powerful way to alter material properties on the nanoscale. Dipolar coupling of excitons to vacuum field fluctuations of surface plasmon polaritons (SPPs) leads to a periodic energy transfer (Rabi oscillations) and the creation of hybridized polariton states. Often, experiments are limited to linear spectroscopy and time-domain studies of the energy transfer are lacking. We investigate the coherent dynamics of J-aggregate excitons that strongly couple to structured SPP fields of a periodic nanoslit array using two-dimensional electronic spectroscopy (2DES) [1]. Strong coupling predicts that "oscillating cross-peaks" should appear between diagonal polariton peaks. Our experiment indeed resolves these Rabi oscillations. Their analysis reveals that they reflect a coherent transfer between different, spatially separated excitons. The structured SPP field creates two classes of excitons that either couple strongly or weakly. Our study shows that 2DES allows to directly access the quantum dynamics of the strongly couples system, revealing a new and unexpected plasmon-mediated energy transport. [1]: D. Timmer, et al. arXiv:2307.14708 (2023).

O 8: Surface Reactions

Time: Monday 10:30–13:00 Location: MA 141

O 8.1 Mon 10:30 MA 141

Reactivity of single metal atoms in different coordination environments — •Nan Cao, Longfeng Huang, Johannes V. Barth, and Alexander Riss — Physics Department E20, Technical University of Munich, 85748 Garching, Germany

Metal centers within metalorganic networks hold promise for applications in single-atom catalysis, gas storage and sensing. Their activity is not only determined by the chemical nature of the metal, but is also significantly influenced by the bonding configuration. It is of fundamental scientific and technological relevance to understand how the reactivity of these sites can be controlled via chemical design.

In this work we present direct measurements of the interaction forces of single metal atoms[1] in different metalorganic coordination environments via atomic-scale atomic force microscopy (AFM) experiments. In particular, we examine how the interaction of metal sites with different types of probe atoms (directly attached to the AFM tip) changes as a function of their bonding configuration. Our measurements show that while chemically relatively inert CO-functionalization yields little discrimination between metal centers in different environments, Cl-functionalization gives rise to significant differences: low-coordinated metal atoms exhibit notably stronger interactions with the probe atom.

Such atomic-scale studies provide means to characterize active sites within complex metalorganic architectures, thus aiding in a rational design strategy toward materials with tailored catalytic properties.

[1]*Science* 2019, **366**, 235-238; *J. Phys. Rev. Lett.* 2020, **124**, 096001.

O 8.2 Mon 10:45 MA 141

On-Surface Isomerization of Indigo within 1D Coordination Polymers — •Hongxiang Xu[1], Ritam Chakraborty[2], Abhishek Kumar Adak[2], Arpan Das[2], Biao Yang[1], Dennis Meier[1], Alexander Riss[1], Joachim Reichert[1], Shobhana Narasimhan[2], Johannes V. Barth[1], and Anthoula C. Papageorgiou[1,3] — [1]Technische Universität München, Germany — [2]Jawaharlal Nehru Centre for Advanced Scientific Research, India — [3]National and Kapodistrian University of Athens, Greece

We present surface-confined metallosupramolecular engineering of coordination polymers using natural dyes as molecular building blocks: indigo and the related Tyrian purple. Both building blocks yield identical, well-defined coordination polymers composed of (1 dehydroindigo : 1 Fe) repeat units on two different silver single crystal surfaces. These polymers are characterized atomically by submolecular resolution scanning tunneling microscopy, bond-resolving atomic force microscopy and X-ray photoelectron spectroscopy. On Ag(100) and on Ag(111), the *trans* configuration of dehydroindigo results in N,O-chelation in the polymer chains. On the more inert Ag(111) surface, the molecules undergo thermally induced isomerization from the *trans* to the *cis* configuration and afford N,N- plus O,O-chelation. Density functional theory calculations confirm that the coordination polymers of the *cis*-isomers on Ag(111) and of the *trans*-isomers on Ag(100) are energetically favored. Our results demonstrate post-synthetic linker isomerization in interfacial metal-organic nanosystems.

O 8.3 Mon 11:00 MA 141

Thioetherification of Br-Mercaptobiphenyl Molecules on Au(111) — Ana Barragan[1], Roberto Robles[2], Nicolas Lorente[2,4], and •Lucia Vitali[1,2,3,4] — [1]Advanced Polymers and Materials: Physics, Chemistry and Technology, Chemistry Faculty (UPV/EHU) — [2]Centro de Física de Materiales CFM/MPC(CSIC-UPV/EHU), — [3]Ikerbasque Research Foundation for Science, Bilbao 48009, Spain; — [4]Donostia International Physics Center (DIPC), 20018 San Sebastián,

Thioether polymers are fundamental for a variety of applications. Their synthesis is, however, more challenging than that of other metal-catalyzed reactions due to the reported detachment of the S atom during thermal activation. In this study, it has been demonstrated unambiguously that thermal annealing results in the thioetherification of the 4-bromo-4-mercaptobiphenyl molecule (Br-MBP) adsorbed on the surface of Au(111). Through complementary techniques, such as scanning tunneling microscopy, spectroscopy, and first principle calculations, we have identified four reaction steps, involving sulfhydryl or bromine molecular functional groups and leading to the formation of intermolecular C-S bonds. To form the thioether polymer and to overcome the competitive formation of C-C bonds, two reaction steps, the dehalogenation, and dissociation of the S-Au bond, must occur simultaneously. We detail the electronic properties of the phenyl-sulfur bond and the polymer as a function of the ligand length. This result suggests a wider perspective of this chemical synthesis.

Nano Letters 23, 1350-1354 (2023)

O 8.4 Mon 11:15 MA 141

Real-Space Imaging of Surface Reactions: Deprotonation and Metalation of Phthalocyanine — •Andreas Christ, Matthias Bode, and Markus Leisegang — Physikalisches Institut, Experimentelle Physik II, Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany

On-surface reactions of phthalocyanines allow to drastically change the molecular properties as well as the interaction with a substrate. Several studies have demonstrated metalation process with both co-deposited atoms and by substrate atoms [1]. However, insights into the process related with metalation by means of direct observation on a single-molecule level have rarely been reported.

In our study, we investigate the temperature-driven self-metalation from H_2Pc to CuPc on a Cu(111) surface by means of STM. We find that the metalation process proceeds at a significantly higher temperature threshold than previous reported [2] and that it competes with a deprotonation process. All three molecule species, H_2Pc, H_0Pc, and CuPc, can unambiguously be identified by their distinct STM-induced rotational behavior. This allows us to determine the species of each individual adsorbed molecule and thus gain a detailed insight into the temperature dependence of both surface-induced reactions [3].

[1] J. M. Gottfried, Surf. Sci. Rep. **70**, 259 (2015)
[2] M. Chen *et al.*, J. Phys. Chem. C **118**, 8501 (2014)
[3] A. Christ *et al.*, Phys. Chem. Chem. Phys. **25**, 7681 (2023)

O 8.5 Mon 11:30 MA 141

Surface Chemistry of Porphyrins: Studying Molecular Transformations with XPS and STM — •Eleanor Frampton[1], Matthew Edmondson[2], Michael Clarke[2], Ailish Gray[2], and Alex Saywell[2] — [1]MAX IV Laboratory, Lund, Sweden — [2]School of Physics, University of Nottingham, UK

Porphyrin molecules may participate in a number of on-surface reactions [1], the dynamics of which may be influenced by several factors, including temperature, chemistry of the porphyrin species, choice of substrate, etc. Porphyrin molecules themselves are extremely versatile and act as building blocks in the self-assembly of 2D molecular arrays [2], which can be tailored to have specific electronic, optical or catalytic properties. By employing a combination of spectroscopy and imaging techniques, these reactions can be studied in detail. [1-3]

The presentation will focus in particular on the on-surface transformations of trans-Br2TPP on Cu(111). This molecule is known to undergo Ullman coupling, an intramolecular ring closing reaction, and self-metalation on this surface. The stepwise progression of the reaction will be discussed, particularly how these processes can be probed using techniques such as scanning tunnelling microscopy (STM) and x-ray photoelectron spectroscopy (XPS). I detail how these techniques can be complementary when applied to on-surface chemistry. [1] Gottfried, J. M. (2015), Surf. Sci. Rep., 70(3), 259-379.[2] L. Grill, S. Hecht, Nat. Chem. 12, 115*130 (2020) [3] Edmondson, M., Frampton, E.S. * & Saywell, A. (2022), Chem. Commun. 58(42), 6247-6250.

O 8.6 Mon 11:45 MA 141

Plasmon-driven/enhanced N-N and C-C coupling reactions — •Radwan M. Sarhan — Helmholtz Zentrum Berlin fur Materialien und Energie, Berlin, Germany

Plasmon-induced catalysis, driven by light and noble metals (e.g., Au, Cu, and Ag), offers a promising, ecofriendly and efficient alternative strategy to conventional thermochemistry, thereby driving chemical reactions via resonant excitation of the plasmonic nanoparticles. After excitation of the surface plasmon of the particles, energetic charge carriers (hot electrons and holes) and a high local temperature are generated, which would initiate and/or enhance chemical transformations of the adsorbed molecules. However, the exact contribution of the hot electrons and heat in the overall reaction performance and mechanism is still a hot topic. In this talk, I will present various plasmonic particles with tunable plasmon resonances (e.g., AuNRs and AuPd NRs), which have been used to achieve efficient solar-to-chemical/thermal energy conversion. In addition, I will show our recent results on different chemical reactions ranging from a reduction of small nitroaromatic molecules to more complex N-N and C-C coupling reactions (e.g., Heck reactions) that have been driven via the hot electrons or enhanced by the high local heat. Moreover, different experimental analysis will be shown to unfold the main driving force of the reaction mechanism.

O 8.7 Mon 12:00 MA 141

Unfolding Chemical Diversity in ORR Catalysts: A DFT Study on Experimentally-Inspired Bimetallic-Organic Monolayers — •Juan Manuel Lombardi[1,2], Doris Grumelli[3], Rico Gutzler[4], Fabio Heriberto Busnengo[1], and Paula Abufager[1] — [1]IFIR, Rosario, Argentina — [2]Fritz-Haber-Institut der MPG, Berlin — [3]INIFTA, La Plata, Argentina — [4]MPI for Solid State Research, Stuttgart

In the quest for more efficient catalysts for the oxygen reduction reaction (ORR), the synergy between metallic and organic components presents a promising avenue for catalytic innovation. We focus on self-assembled monolayers (SAM) of FeTPyP on Au(111) and their bimetallic-organic counterpart, the metal-organic coordination network (MOCN) FeTPyP+Co complex on Au(111), which is experimentally prepared through Co atom sublimation. Using Density Functional Theory (DFT), this study elucidates the intricate electronic behavior of the active metal sites in these systems and their mechanistic roles in ORR. We find that the FeTPyP+Co complex on Au(111) achieves a more balanced adsorption of ORR intermediates, owing to a more diverse electronic environment induced by the bimetallic interaction within the metal-organic framework. This study not only widens the spectrum of chemical landscapes for ORR catalysts but also establishes a theoretical basis that could inspire the proposal and conceptual development of novel catalysts.

O 8.8 Mon 12:15 MA 141

Free Energy Barrier Calculation for Catalytic Hydrogen Assisted CO Dissociation using Machine-Learning Surrogate Models — •Hyunwook Jung, Karsten Reuter, and Johannes T. Margraf — Fritz-Haber-Institut der MPG, Berlin

The calculation of accurate reaction rate constants of elementary processes is a central element of computational surface catalysis research. At present, this proceeds prevalently via transition state theory, with the process free energy barrier as primary input. Ideally, such barriers are obtained through extensive sampling of configurations along the transition path, using techniques such as umbrella sampling. However, the associated biased molecular dynamics simulations are prohibitively expensive at the density functional theory level, making the more efficient but less accurate harmonic approximation a common practice in most studies. In order to overcome this limitation, we develop a machine learning surrogate model using an iterative training workflow. We apply this computationally undemanding surrogate model to hydrogen assisted CO dissociation as the key rate limiting step in synthesis gas conversion on Rh, Cu, and Ru surfaces. Obtaining significant thermal effects not captured in the harmonic approximation, we investigate the consequences on product selectivities.

O 8.9 Mon 12:30 MA 141

Dynamics of enantioselective surface chemistry on chiral intermetallic substrates — •Raymond Christopher Amador[1,2], Hannah Bertschi[1,2], and Daniele Passerone[1,2] — [1]Empa, Dübendorf, Switzerland — [2]ETH Zürich, Zürich, Switzerland

In this work, we explore the enantioselective surface adsorption and reaction of two representative organic molecules (DBBA and 9AP) on intermetallic palladium gallium (PdGa) chiral substrates. By briefly analysing dispersive forces in conjunction with density-functional theory, we elucidate the enantioselective interactions between the (pro)chiral molecules and the PdGa surface, focusing on the dynamic processes that govern adsorption/desorption and surface diffusion. We study kinetics and thermodynamics of the processes by means of tailored molecular dynamics simulation methods (metadynamics) that are able to sample the free energy landscape of the process at realistic temperatures. Adsorption configurations and free energy barriers are obtained and enlighten the dynamics of chirality sensitive reactions. The presented findings contribute to a deeper understanding of the molecular mechanisms governing chiral selectivity in catalytic systems, providing insights that can guide the design and optimization of enantioselective catalysts, with possible applications to the pharmaceutical and chemical industry.

O 8.10 Mon 12:45 MA 141

Propionic Acid at Air/water Interface: An Ab initio Molecular Dynamic Simulation Study — •Ali Ghiami-Shomami[1], Dominika Lesnicki[2], and Marialore Sulpizi[1] — [1]Faculty of Physics and Astronomy, Department of Physics, Ruhr Universität Bochum, NB6, 44780, Bochum, Germany — [2]LPCT UMR 7019 CNRS, Université de Lorraine 54506 Vandoeuvre-lès-Nancy Cedex, France

Organic materials are abundant in Earth's atmosphere and are present in aerosols. Particularly, organic acids can catalyze reactions on aerosols and also contribute to atmospheric acidity and regulate precipitation pH [1-2]. Therefore, understanding the structural, dynamical, and acidic properties of such acids at the air/water interface is important. Extending our previous work [3], we employ here ab initio molecular dynamics (AIMD) simulations to investigate propionic acid's behavior in the bulk and at the air/water interface. In particular we use DFT-based simulations at the BLYP level to investigate cis and trans conformers. We find that in bulk the cis conformer is more stable and this observation is in line with findings for formic acid as parent of propionic acid. Moreover, structural analysis indicate that there is no isomerization during simulation time. Additionally the solvation structure for the acid in bulk and at the interface is discussed and compared to other small organic acids. Finally the relative acidity between bulk and interface molecules is also presented.

References [1] P. G. Blower et al., J. Phys. Chem. A, 2013, 117, 2529. [2] B. Franco et al., Nature, 2021, 593, 233. [3] D. Lesnicki et al., Phys. Chem. Chem. Phys., 2022, 24, 13510.

O 9: Semiconductor Substrates I: Adsorption of Small Molecules, Metallic Nanowires, Overlayers

Time: Monday 10:30–12:15 Location: MA 144

O 9.1 Mon 10:30 MA 144

Bayesian and Multi-Objective Optimization for Sensor Technologies — •Ransell D'Souza[1], Shuja Malik[2], Fatima Annanouch[2], Eduard Valero[2], and Milica Todorović[1] — [1]University of Turku, Turku, Finland — [2]Universitat Rovira i Virgili

Sensors, a device that can detect toxic and combustible gasses, play a crucial role in identifying potentially hazardous gas leaks. Sensors typically function by employing inorganic substrates that respond to the adsorption of target molecules. To design selective and sensitive sensors, density functional theory studies should explore the interplay between adsorption energies, charge transfer, recovery time, and sensor response during interactions between toxic targets and the substrate.

While conventional computational approaches have focused on individual material properties, we need to balance different considerations of these properties for a truly predictive sensor design. This presents a multi-objective optimization problem, which can be effectively addressed using Bayesian Optimization (BO)-based machine learning techniques.

In this study, we employ multi-objective BO techniques to investigate the stable adsorbate structures of NH_3 on WS_2. We identify optimal tradeoffs between energetically stable structures and optimal response functions (Pareto optimal solutions) for the rational design of new sensors. Our resistivity and sensor response functions agree with experimental data, validating active learning's success in gas sensor material optimization.

O 9.2 Mon 10:45 MA 144

N-Heterocyclic Olefins on a Silicon Surface — •Martin Franz[1], Mowpriya Das[2], Conor Hogan[3,4], Robert Zielinski[1], Milan Kubicki[1], Maximilian Koy[2], Canan Kosbab[1], Simone Brozzesi[4], Ankita Das[2], Mike Thomas Nehring[1], Viktoria Balfanz[1], Juls Brühne[1], Mario Dähne[1], Norbert Esser[1,5], and Frank Glorius[2] — [1]Technische Universität Berlin, Institut für Festkörperphysik, Berlin, Germany — [2]Westfälische Wilhelms-Universität Münster, Organisch-Chemisches Institut, Münster, Germany — [3]Istituto di Struttura della Materia-CNR (ISM-CNR), Rome, Italy — [4]Università di Roma "Tor Vergata", Dipartimento di Fisica, Rome, Italy — [5]Leibniz-Institut für Analytische Wissenschaften "ISAS e.V.", Berlin, Germany

N-Heterocyclic carbenes are known to be excellent ligands for surface modification, and recently also the formation of ordered monolayers on silicon has been reported [1]. In contrast, surface adsorption of their close relatives, the N-heterocyclic olefins (NHOs), has not been studied yet. Here, a combined scanning tunneling microscopy, X-ray photoelectron spectroscopy, and density functional theory study of NHOs on silicon is presented. We find that the two studied NHOs bind covalently with ylidic character to the silicon adatoms of the substrate and exhibit good thermal stability. The adsorption geometry is found to depend on the N-substituents and strongly influences the quality and properties of the obtained monolayers.

[1] M. Franz et al., Nat. Chem. **13**, 828-835 (2021).

[2] M. Das et al., Angew. Chem. Int. Ed. **2023**, e202314663.

O 9.3 Mon 11:00 MA 144

Adsorption of cysteine on the surface of TiO2 — •Miguel Blanco Garcia[1], Mona Kohantorabi[1], Andreas Stierle[1,2], and Heshmat Noei[1,2] — [1]Centre for X-ray and Nanoscience (CXNS), Deutsches Elektronen-Synchrotron (DESY), Notkestr. 85, 22607 Ham-burg, Germany — [2]Fachbereich Physik Universität Hamburg, Jungiusstr. 9-11, D-20355, Hamburg, Germany

Titanium oxide stand as a promising material for the inactivation of the viruses under light irradiation [1]. Our investigation centers on the most abundant amino acid within the spike proteins of SARS-CoV-2, cysteine [2], exploring its adsorption behavior on the surface of TiO2 under ultra-high vacuum (UHV) and aqueous conditions. Employing X-ray photoelectron spectroscopy, we observe UV light induces desorption of the cysteine molecule as well as its mineralization. Fourier-Transform Infrared Reflection Absorption Spectroscopy (FT-IRRAS) sheds light on the preferred binding modes and molecular arrangement of the cysteine during adsorption on TiO2. In addition, FT-IRRAS elucidates which bonds are influenced by UV treatment and unveils the molecular breakdown process. Furthermore, we monitored the adsorption of cysteine with TiO2 in an aqueous solution using Grazing-Incidence Small-Angle X-ray Scattering (GISAXS) at DESY P03 beam line which provides further information on the adsorption geometry and self-assembly of the cysteine amino acid TiO2 surfaces. [2] Kohantorabi, et al., ACS Appl. Mater. Interface., 15 (2023) 8770-8782. [3] Wang D, et al., Nano today 40 (2021) 101243.

O 9.4 Mon 11:15 MA 144

The impact of adatom adsorption on the interfacial structure of InP(001) — •Vibhav Yadav[1,2], Margot Guidat[1,2], Mario Löw[2], Jognmin Kim[1,2], Holger Euchner[1], and Matthias May[1,2] — [1]Institute of Physical and Theoretical Chemistry, Universität Tübingen, — [2]Institute of Theoretical Chemistry, Universität Ulm, Germany

Controlling the electrochemical interface of III-V semiconductors such as InP is of great relevance for hydrogen production in Photoelectrochemical water-splitting devices [1]. Here, reducing corrosion, while at the same time maintaining efficient charge transfer, remains challenging and requires an atomistic understanding of the InP(001) surface in an electrochemical environment. While InP(001) is known to exhibit different surface reconstructions, depending on the exact synthesis conditions, the presence of oxygen strongly affects the surface structure.

In this work, we apply Density Functional Theory to investigate the thermodynamic stability of the different reconstructions pertaining to adatom exposure such as oxygen, finally resulting in an extended surface phase diagram. Furthermore, we show that combining computational and experimental Reflection Anisotropy Spectroscopy has the potential to provide a better understanding of InP surfaces/interfaces and their (electronic) structure under operation conditions[2].

[1] : May MM; Lewerenz HJ; Lackner D; Dimroth F; Hannappel T. In: Nat Commun 6.1 (Sept. 15, 2015), p. 8286

[2] :Löw, M.; Guidat, M.; Kim, J.; May, M. M. RSC Adv. 2022, 12 (50), 32756*32764

O 9.5 Mon 11:30 MA 144

Realizing strictly one dimensional behavior in arrays of atomic wires on semiconducting substrate emerging from dispersive two dimensional system — Keshab Sony and •Anas Abdelwahab — Leibniz Universität Hannover, Hanover, Germany

We studied single and two Su-Shreiffer-Heeger wires on simple cubic semiconducting substrate. The wire-wire coupling is either perpendicular or diagonal hopping respecting the particle-hole and time-reversal symmetries. The hybridization to substrate renormalizes the model parameters of the wires towards the hopping parameter of the substrate without changing the basic nature of perpendicular or diagonal coupling and it can mediate effective perpendicular hopping but not diagonal hopping in the absence of direct wire-wire coupling. This justifies the investigation of multi uniform tight binding wires with perpendicular or diagonal hopping parameters while neglecting the substrate. Perpendicularly coupled uniform wires reveal anisotropic two dimensional band dispersion. Diagonally coupled uniform wires reveal strictly one dimensional bands parallel to the wires direction if the intra-wire hopping parameter is larger than twice the diagonal hopping parameter despite strong dispersion perpendicular to the wires. Otherwise, they reveal strictly one dimensional bands parallel and perpendicular to the wires direction simultaneously. We established the possibility of realizing strictly one dimensional properties emerging from dispersive two dimensional system if time-reversal and particle-hole symmetries are respected. This can facilitate the debate on one dimensional behavior of the Au/Ge(001) surface reconstruction.

O 9.6 Mon 11:45 MA 144

Diffusive mass transport of Pb on Si(111) (7x7) — •Felix Hartmann[1], Paul P. Schmidt[1], Regina Hoffmann-Vogel[1], Ralf Metzler[1,3], and Janet Anders[1,2] — [1]Institute of Physics and Astronomy, University of Potsdam, 14467 Potsdam, Germany — [2]Department of Physics and Astronomy, University of Exeter, Stocker Road, Exeter EX4 4QL, UK — [3]Asia Pacific Center for Theoretical Physics, Pohang 37673, Republic of Korea

Pb on Si(111)-(7x7) has been subject of numerous studies due to its strong quantum size effect, which explains the height selection and the characteristic shape of coarsened islands [1]. Previous experiments have shown the fast nucleation and “explosive” growth of the Pb islands, which happens on a timescales much faster than explained by classical nucleation theory [2,3]. In this talk we focus on numerically modelling the mass transport onto a growing island, as a first step to find an explanation for the “explosive” island growth. Our simulations are motivated by a recent experiment, which locally triggers stable Pb islands to grow in height. We show that certain surface processes in the Pb/Si(111)-(7x7) are well-described and explained by classical diffusion models.

[1] Z. Kuntová et. al. Phys. Rev. B, **78** (2008)
[2] H. Hattab et. al. Surf. Sci. **646** (2016)
[3] M. Hershberger et. al. Phys. Rev. Lett., **113** (2014)

O 9.7 Mon 12:00 MA 144

An in-situ microGISAXS growth study of metallic thin films on self organized nanopatterned templates — •Sarathlal Koyiloth Vayalil[1,2], Praveen Kumar Dubey[2], Benedikt Sochor[1], Matthias Schwartzkopf[1], Stephan V. Roth[1], and Ajay Gupta[2] — [1]Deutsches Elektronen Synchrotron DESY, Notkestr. 85, 22607, Germany — [2]UPES, Bidholi Campus, Dehradun, 248007, Uttarakhand, India

In this work, a detailed insitu μGISAXS investigation of the growth of metallic thin films on self-organized nanopatterned templates prepared by low energy ion beam erosion has been done. GISAXS experiments were performed at the P03/MiNaXS beamline of the PETRA III storage ring at DESY (Hamburg, Germany).Two different kind of nanopatterned templates, (i) GaSb nanocones and (ii) nanorippled Si prepared by low energy ion beam erosion under two different experimental geometry have been used as templates to grow thin films of Py and Ag separately. Different growth regimes have been analyzed with increasing thickness. Obtained structural information have been correlated with observed variation in the functional properties of the thin films with increasing thickness values.

O 10: Focus Session: Frontiers of Electronic-Structure Theory – Advances in Time-Dependent and Nonequilibrium Ab Initio Methods II

Time: Monday 15:00–17:15 Location: HE 101

O 10.1 Mon 15:00 HE 101

Thermoelectric efficiency in multiterminal quantum thermal machines from steady-state density functional theory — Nahual Sobrino[1], •Roberto D'Agosta[1,2], and Stefan Kurth[1,2,3] — [1]Nano-Bio Spectroscopy Group and European Theoretical Spectroscopy Facility (ETSF), Departamento de Polímeros y Materiales Avanzados: Física, Química y Tecnología, Basque Country University UPV/EHU, San Sebastián, Spain — [2]IKERBASQUE, Basque Foundation for Science, Bilbao, Spain — [3]Donostia International Physics Center, San Sebastián, Spain

Steady-state density Functional Theory for electronic and thermal transport (iq-DFT) is a formalism to describe open quantum systems in nonequilibrium steady states. iq-DFT relies on the one-to-one map between the set of ”density” variables (particle density, electrical, and heat current) and the set of ”potentials” (local potential, applied voltage, and thermal gradient). The resulting Kohn-Sham system features three exchange-correlation (xc) potentials: a local xc potential, an xc contribution to the voltage, and an xc contribution to the thermal gradient. In the linear-response regime, we arrive at exact expressions for the many-body transport coefficients and thermoelectric efficiency purely in terms of quantities accessible to iq-DFT. We generalize iq-DFT to the multitermal situation and apply the formalism to a multiterminal interacting quantum dot in the Coulomb blockade regime for which accurate parametrizations of the xc kernel matrix are given, allowing us to study the thermoelectric efficiency and output power of the multiterminal system.

O 10.2 Mon 15:15 HE 101

Anisotropic Charge, Thermal, and Thermoelectric Transport in β-Ga$_2$O$_3$ from First Principles — •Sebastian Tillack, Nakib Protik, and Claudia Draxl — Humboldt-Universität zu Berlin and IRIS Adlershof, 12489 Berlin, Germany

Gallium oxide gained interest for high-power applications due to its large band gap of ~ 5 eV and a high breakdown voltage. On the other hand it shows only modest electron mobility and relatively low thermal conductivity. While the latter is a clear limitation for power electronics it might render the material useful for thermoelectric applications.

We present an *ab initio* study of transport in β-Ga$_2$O$_3$ including electron–phonon and phonon–phonon interactions. Electrons and phonons are described within a full-potential all-electron framework [1] using many-body perturbation theory and density-functional perturbation theory. Maximally localized Wannier functions and Wannier-Fourier interpolation facilitate ultra fine samplings of the Brillouin zone. We solve the coupled electron–phonon Boltzmann transport equations [2] to compute the anisotropic electron mobility, Seebeck coefficient and thermal conductivity for different temperatures and electron doping levels, and the effect of the mutual electron–phonon drag on these transport coefficients. Our work provides insight into the fundamental scattering processes limiting transport in this material and explores its usefulness for various applications.

[1] A. Gulans, *et al.*, J. Phys.: Condens. Matter **26**, 363202 (2014).
[2] N.H. Protik, *et al.*, npj Comput. Mater. **8**, 28 (2022).

O 10.3 Mon 15:30 HE 101

Defect assisted charge transfer across silicon-related heterojunctions — •Marvin Krenz, Wolf Gero Schmidt, and Uwe Gerstmann — Paderborn University, Warburger Str. 100, 33098 Paderborn

Charge transfer is highly relevant for many physical, chemical, and biological processes. The transfer of triplet excitons across the tetracene (Tc)-silicon interface in sensitized solar cells is a recent example. Another one is the active a-Si:H/c-Si interface of silicon-based solar cells. In both cases, the presence of dangling bond (db) defects are often considered detrimental to material performance. While the atomistic details of the interfaces are decisive for maximizing the charge transfer, the transfer mechanism and related dynamics are essentially unknown.

Here, we present ab initio molecular dynamics calculations that provide a microscopic understanding of the charge and exciton transfers across the above mentioned interfaces. The related electron hopping at level crossings in the band structure of the 2D periodic structures is described by c-DFT based surface hopping algorithms [1]. The transfer characteristics are found to depend in fact strongly on the presence of Si dangling bonds at the interfaces. At the Tc-Si(111):H interface they are even required to enable the excitation transfer. Defect induced states close to the valence band directly assist the hole transfer into Si bulk, and the exciton's electron is found to follow the hole within a few femtoseconds.

[1] M. Krenz, U. Gerstmann, W.G. Schmidt, ACS Omega 5, 24057 (2020).

O 10.4 Mon 15:45 HE 101

Deciphering the Mechanisms of Oxygen Reduction Reaction and Reactive Sites in Polymeric Carbon Nitride Structures for Photocatalysis — •Changbin Im and Timo Jacob — Ulm University, Institute of Electrochemistry, Ulm, Germany

Solar-to-Fuel technology holds immense promise for sustainable energy solutions, with polymeric carbon nitrides (PCN) emerging as pivotal catalysts in harnessing solar energy for hydrogen peroxide production through water reduction. Our ab initio calculations meticulously examine the oxygen reduction reaction (ORR) on diverse monolayers of PCNs, unraveling the energetics and mechanisms underlying ORR. Notably, structural factors, including condensation and corrugation degrees, exert profound influences on these processes. Departing from conventional 4-electron transfer mechanisms, our research introduces an innovative perspective by highlighting the significant contribution of electron shooting to the outer Helmholtz plane. Furthermore, we underscore the imperative nature of a pre-protonation process preceding ORR initiation. Employing calculated absorption patterns from TDHF@Casida, we uncover potential reactive sites within the structural features of polymeric carbon nitrides during photo-induced ORR.

This investigation enhances our comprehension of the ORR mechanism on PCN structures, shedding light on photo-induced ORR mechanisms and identifying promising reactive sites. Our comprehensive exploration not only deepens understanding but also provides insights crucial for optimizing reactive sites in PCN structures, thereby amplifying their efficacy in sustainable energy applications.

O 10.5 Mon 16:00 HE 101

Multi-channel Dyson equation: coupling many-body Green's functions — •Arjan Berger — Paul Sabatier University, Toulouse, France

We present the multichannel Dyson equation that combines two or more many-body Green's functions to describe the electronic structure of materials. In this work we use it to model photoemission spectra by coupling the one-body Green's function with the three-body Green's function. We demonstrate that, unlike methods using only the one-body Green's function, our approach puts the description of quasi-particles and satellites on an equal footing. We propose a multichannel self-energy that is static and only contains the bare Coulomb interaction, making frequency convolutions and self-consistency unnecessary. Despite its simplicity, we demonstrate with a diagrammatic analysis that the physics it describes is extremely rich. Finally, we present a framework based on an effective Hamiltonian that can be solved for any many-body system using standard numerical tools.

References:
G. Riva, P. Romaniello, and J. A. Berger, Phys. Rev. Lett. 131, 216401 (2023)
G. Riva, T. Audinet, M. Vladaj, P. Romaniello, and J. A. Berger, SciPost Phys. 12, 093 (2022)

O 10.6 Mon 16:15 HE 101

Third-order susceptibility of crystalline materials from time-dependent density functional theory — •Xiao Chen — ZGH,RUB,Bochum,Germany

We use real-space real-time time-dependent density functional theory to investigate the non-linear properties of centrosymmetric crystals. We perform calculations varying laser intensities and extract non-linear coefficients, such as the Kerr coefficient, the two-photon absorption and the third-order susceptibility. In semiconductors such as diamond, we investigate nonlinear properties at different laser frequencies. In metals such as gold, we investigate the effect of different laser pulse durations. The results compare well with experimental reference data.

O 10.7 Mon 16:30 HE 101

Linear scaling approach for optical excitations using maximally localized Wannier functions — •Konrad Merkel and Frank Ortmann — TUM School of Natural Sciences, Technische Universität München

We present a theoretical method for calculating optical absorption spectra based on maximally localized Wannier functions, which is suitable for large periodic systems. For this purpose, we calculate the exciton Hamiltonian, which determines the Bethe-Salpeter equation for the macroscopic polarization function and optical absorption characteristics. The Wannier functions are specific to each material and provide a minimal and therefore computationally convenient basis. Furthermore, their strong localization greatly improves the computational performance in two ways: first, the resulting Hamiltonian becomes very sparse and, second, the electron-hole interaction terms can be evaluated efficiently in real space, where large electron-hole distances are handled by a multipole expansion. For the calculation of optical spectra we employ the sparse exciton Hamiltonian in a time-domain approach, which scales linearly with system size. Further information can be found in J. Phys. Mater. 7 015001 (2024)

O 10.8 Mon 16:45 HE 101

Exploiting Crystal Symmetry in the LAPW Method — •Mara Voiculescu, Sven Lubeck, and Claudia Draxl — Humboldt-Universität zu Berlin, Berlin, Germany

In the framework of density functional theory (DFT) calculations, the linearized augmented planewave (LAPW) method uses a dual representation for physical quantities such as the charge density and potential. This approach consists of a spherical harmonic expansion inside the so-called muffin-tin spheres around the nuclear positions and a planewave expansion in between them. The direct implementation results in a large number of expansion coefficients which need to be computed. Exploiting symmetry by employing group-theoretical methods simplifies the mathematical description and therefore reduces computational expenses. Consequently, symmetry can be used to speed up the setup of the Hamiltonian matrix, while at the same time resulting in better numerical precision. We implement this approach in the all-electron full-potential computer package exciting [1], by using symmetrized spherical harmonics (lattice harmonics) and planewaves (stars), yielding a considerable computational speedup for a variety of different materials.

[1] A. Gulans et al., J. Phys. Condens. Matter 26, 363202 (2014).

O 10.9 Mon 17:00 HE 101

Brillouin zone sampling in ONETEP — •Chengcheng Xiao, Peter Haynes, and Arash Mostofi — Departments of Materials and Physics, and the Thomas Young Centre for Theory and Simulation of Materials, Imperial College London, London SW7 2AZ, United Kingdom

ONETEP is a linear-scaling density functional theory (DFT) program with large basis set (plane-wave) accuracy on parallel computers. ONETEP was originally designed to study large-scale systems, including macromolecules and nanocrystals. Because of this, ONETEP is restricted to Gamma-point only for self-consistent calculations. However, many systems of interest are periodic at short-scales in one or two dimensions, e.g., nanowires and surfaces. In such cases, significant computational efficiency can be achieved via dense sampling of the Brillouin zone (rather than creating large supercells).

Here, we introduce two types of Brillouin zone sampling methods in ONETEP. The first is targeted at small periodic systems and exploits the fact that the localization constraints of the non-orthogonal Wannier functions (NGWFs) can be lifted. The second keeps the NGWFs localized and the k-dependence is introduced as phase factors to the Hamiltonian matrix elements (similar to the tight-binding method). This method keeps the code linear scaling with k-points and is aimed at intermediate-sized systems.

These developments extend the operability of ONETEP to short- and intermediate-scale periodic systems, including 2D materials, nanowires, nanotubes, surfaces, and interfaces.

O 11: Focus Session: Spins on Surfaces studied by Atomic Scale Spectroscopies II

Time: Monday 15:00–17:45 Location: MA 004

Topical Talk O 11.1 Mon 15:00 MA 004

Unusual magnetism of closed-shell molecules on metal substrates — •Alexander Weismann[1], Arnab Banerjee[1], Manuel Gruber[2], Jan Homberg[1], Niklas Ide[1], and Richard Berndt[1] — [1]Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, D-24098 Kiel, Germany — [2]Faculty of Physics and CENIDE, University of Duisburg-Essen, 47057 Duisburg, Germany

Molecular magnetism is studied using low temperature scanning tunneling spectroscopy by resolving spectroscopic fingerprints close to the Fermi energy like Kondo resonances, inelastic spin excitations and Yu-Shiba-Rusinov (YSR) states. We find a variety of closed-shell molecules that can become paramagnetic in suitable environments or by molecular switching. Furthermore, an unexpectedly large orbital moment related to molecular ring currents is observed. Using YSR states we sensitively detect a large number of molecular vibrations via inelastic tunneling. Magnetic and vibrational excitations are spatially mapped and energy shifts caused by intermolecular interactions are resolved on a 100 μeV scale. Taken together our results highlight the importance of electrostatic stray fields due to polar bonds in molecules.

O 11.2 Mon 15:30 MA 004

Theory of a Single Magnetic Impurity on a Thin Metal Film in Proximity to a Superconductor — •Jon Ortuzar Andres[1], Stefano Trivini[1], Katerina Vaxevani[1], Jingchen Li[2], F. Sebastian Bergeret[3,4], Miguel A. Cazalilla[4,5], and Jose Ignacio Pascual[1,5] — [1]CIC nanoGUNE, Donostia-San Sebastián, Spain — [2]School of Physics, Sun Yat-sen University, Guangzhou 510275, China — [3]Centro de Física de Materiales (CFM-MPC) Centro Mixto CSIC-UPV/EHU, Donostia-San Sebastián, Spain — [4]Donostia International Physics Center (DIPC), Donostia-San Sebastián, Spain — [5]Ikerbasque, Basque Foundation for Science, Bilbao, Spain

Magnetic impurities on proximitized thin films can host Yu-Shiba-Rusinov states, entangled excitation between the spin of the impurity and the electrons of the substrate. We argue that the formation of YSR excitations in proximitized thin films is largely mediated by a type of Andreev-bound state named after de Gennes and Saint-James. This is shown by studying an experimentally motivated model and computing the overlap of the wave functions of these two sub-gap states. We find the overlap stays close to unity even as the system moves away from weak coupling across the parity-changing quantum phase transition. Based on this observation, we introduce a single-site model of the bound state coupled to a quantum spin. This model can qualitatively explain the complex behaviour of magnetic impurities interacting with the proximitized metal. The adequacy of this description is assessed by reintroducing the coupling to the continuum as a weak perturbation within the framework of Anderson's poor-man's scaling.

O 11.3 Mon 15:45 MA 004

Microwave excitation of atomic scale superconducting bound states — Janis Siebrecht[1], Haonan Huang[1], Piotr Kot[1], Robert Drost[1], Ciprian Padurariu[2], Björn Kubala[2], Jochim Ankerhold[2], Juan Carlos Cuevas[3], and •Christian R. Ast[1] — [1]MPI für Festkörperforschung, Stuttgart — [2]Institut für Komplexe Quantensysteme, Universität Ulm, Ulm — [3]Departamento de Física Teórica de la Materia Condensada, Universidad Autónoma de Madrid, Madrid, Spain

Magnetic impurities on superconductors lead to bound states within the superconducting gap, so called Yu-Shiba-Rusinov (YSR) states. They are parity protected, which enhances their lifetime, but makes it more difficult to excite them. Here, we realize the excitation of YSR states by microwaves facilitated by the tunnel coupling to another superconducting electrode in a scanning tunneling microscope (STM). We identify the excitation process through a family of anomalous microwave-assisted tunneling peaks originating from a second order resonant Andreev process, in which the microwave excites the YSR state triggering a tunneling event transferring a total of two charges. We vary the amplitude and the frequency of the microwave to identify the energy threshold and the evolution of this excitation process. Our work sets an experimental basis and proof-of-principle for the manipulation of YSR states using microwaves with an outlook towards YSR qubits.

O 11.4 Mon 16:00 MA 004

YSR-STM thermometer for superconducting quasiparticle temperatures — •Ciprian Padurariu[1], Sujoy Karan[2], Haonan Huang[2], Björn Kubala[1,3], Christian R. Ast[2], and Joachim Ankerhold[1] — [1]Institute for Complex Quantum Systems and IQST, Ulm University, Ulm, Germany — [2]Max-Planck-Institut für Festkörperforschung, Stuttgart, Germany — [3]Institute of Quantum Technologies, German Aerospace Center (DLR), Ulm, Germany

The occupation of the quasiparticle continuum in a superconductor is of critical importance for the functionality of Josephson junction-based quantum devices. While significant progress has been made recently, the detection and removal (trapping) of quasiparticles remains a significant challenge. This talk will describe the design of a thermometer that detects the presence and effective temperature of superconducting quasiparticles. The device consists of a mK-STM functionalized with a single Yu-Shiba-Rusinov (YSR) state inside the gap. Transport between the tip and a clean superconducting substrate shows Zeeman-split peaks in the differential conductance associated to the YSR state [1]. The width of the peaks is sensitive to the effective temperature of quasiparticles above the gap. We show that this width manifests not only in the voltage-dependence, but also in the magnetic field- and exchange coupling-dependence of the current. This allows determining the effective temperature below the limit of the STM energy resolution. [1] W.-V. van Gerven Oei, *et al.*, "Magnetic impurities in spin-split superconductors", Phys. Rev. B **95**, 085115 (2017).

O 11.5 Mon 16:15 MA 004

Non-local detection of Yu-Shiba-Rusinov states in quantum corrals — Khai Ton That[1], Chang Xu[2], Ioannis Ioannidis[3], Lucas Schneider[1], Thore Posske[3], Dirk K. Morr[2], Roland Wiesendanger[1], and •Jens Wiebe[1] — [1]Department of Physics, University of Hamburg, Hamburg, Germany — [2]University of Illinois at Chicago, Chicago, USA — [3]Institute for Theoretical Physics, University of Hamburg, Hamburg, Germany

Quantum mirages are non-local projections of local quantum states induced by an atomic-scale defect in a bath of conduction electrons that can be tailored by cages of nonmagnetic atoms, so-called quantum corrals [1,2]. Here, we observe, by scanning tunnel spectroscopy, the mirage of the Yu-Shiba-Rusinov (YSR) state induced by a magnetic atom in the gap of a superconductor and a characteristic corral-eigenenergy dependent oscillation in its particle-hole asymmetry. Our modelling shows that the mirage is dominated by an indirect coupling of the YSR state induced in the bulk superconductor with the corral eigenmodes. [1] H. Manoharan et al., Nature 403, 512 (2000); [2] L. Schneider et al., Nature 621, 60 (2023).

O 11.6 Mon 16:30 MA 004

Yu-Shiba-Rusinov bands in a self-assembled kagome lattice of magnetic molecules — •Laëtitia Farinacci[1], Gaël Reecht[1], Felix von Oppen[2], and Katharina J. Franke[1] — [1]Fachbereich Physik, Freie Universität Berlin, Germany — [2]Dahlem Center for Complex Quantum Systems and Fachbereich Physik, Freie Universität Berlin, Germany

The kagome lattice features both a flat-band and a Dirac cone, which makes it of particular interest for electronic correlations and relativistic effects. Moreover, if anti-ferromagnetic nearest-neighbour coupling takes place, its geometry ensures magnetic frustration. This further broadens the range of physical phenomena that a kagome lattice can host.

However, it is challenging to realize an ideal two-dimensional lattice as interlayer coupling or interactions with a solid-state support may suppress the correlations. Here, we initiate a study a kagome lattice hosting unpaired electron spins on a superconductor. Using molecular self-assembly we show that the deposition of Fe-porphine-chloride molecules on a Pb(111) surface can lead to the formation of a kagome lattice in 2D molecular islands, and also of smaller units that correspond to kagome precursors. Each FeP molecule induces a magnetic bound state, called Yu-Shiba-Rusinov (YSR) state and we show that these hybridize in the kagome precursors and lead to YSR bands in large islands.

O 11.7 Mon 16:45 MA 004

Spin-flip excitations in organometallic chains — •Jung-Ching Liu[1], Chao Li[1], Outhmane Chahib[1], Xing Wang[2], Ping Zhou[2], Robert Häner[2], Silvio Decurtins[2], Ulrich Aschauer[3], Shi-Xia Liu[2], Ernst Meyer[1], and Rémy Pawlak[1] — [1]Department of Physics, University of Basel, Klingelbergstrasse 82, CH-4056 Basel — [2]Department of Chemistry and Biochemistry, University of Bern, Freiestrasse 3, CH-3012 Bern — [3]Department of Chemistry and Physics of Materials, University of Salzburg, Jakob-Haringer-Strasse 2A, A-5020 Salzburg

Controlling the spin of metal adatoms in well-ordered organometallic (OM) frameworks is a key step towards the realization of molecular electronics and spintronics. Here, we present the study of magnetic signatures at metal centers in the complex structure coordinated by PTO and Fe adatoms[1]. STM and AFM with CO-terminated tips[2] reveal the structure of the OM chains. STS at 1K indicates three spin-flip excitations and collective spin-state excitations of Fe on Ag(111). On Pb(111), PTO-Fe chains only show two spin-flip excitations outside the superconducting gap. By comparing these two systems, we conclude that the spin characteristics of the PTO-Fe chain do not change drastically between these substrates, while spin-state excitation is expected to have a longer lifetime on superconductors[3,4]. We believe our study provides a route for fundamental studies in spin-spin and spin-substrate interactions with different lattice structures.[1]Chem. Eur. J., 2016, 22, 8105-8112. [2]PRL, 2011, 107, 086101. [3]Nat. Phys., 2013, 9, 765-768. [4]Nano Lett. 2022, 22, 6075-6082.

O 11.8 Mon 17:00 MA 004

Tunable ferromagnetic order in 2D layers of transition metal dichlorides — •Andrea Aguirre[1,2], Andrés Pinar[3], Jon Ortuzar[2], Oleksandr Stesovych[3], Nacho Pascual[2], Celia Rogero[1], Pavel Jelínek[3], Maxim Ilyn[1], and Martina Corso[1] — [1]Materials Physics Center (CSIC-UPV/EHU), San Sebastian, Spain — [2]CIC nanoGUNE-BRTA, San Sebastian, Spain — [3]Institute of Physics, Czech Academy of Sciences, Prague, Czech Republic

Metal dihalides TMX2, where TM is a transition metal and X a halogen, are a class of 2D layered materials bonded through van der Waals interactions. These binary compounds exhibit magnetic texture with semiconducting electronic properties. Since single layers can grow epitaxially on metal substrates, there is a strong interest in determining whether these properties persist at the 2D limit.

Here we study the epitaxial growth of FeCl2 and NiCl2 on Au (111). Their chemical and electronic properties were explored high-resolution X-ray photoelectron spectroscopy (XPS) and low-temperature scanning tunneling microscopy and spectroscopy (STM). These 2D materials grow in large and flat monolayers electronically decoupled from the substrate. Our studies on the magnetic properties by X-ray absorption spectroscopy (XAS) revealed a ferromagnetic order related to the 3d electrons of the metal in the compound. The magnetic alignment depends on the TM used, so that it can be switched from out-of-plane to in-plane by substituting the metal ion from iron to nickel. Additionally, we probed the same magnetic behavior at the atomic scale by using nickelocene functionalized STM tips.

O 11.9 Mon 17:15 MA 004

Distinct Quantum States in Topological Insulator Surfaces of Nanowires and Nanoribbons of Bismuth Selenide — •Christian Nweze[1], Tomke Eva Glier[1], Mika Rerrer[1], Malte van Heek[1], Sarah Scheitz[1], Lewis Akinsinde[1], Niklas Kohlmann[2], Lorenz Kienle[2], Yalan Huang[1], Wolfgang Parak[1], Nils Huse[1], and Michael Rübhausen[1] — [1]Institute of Nanostructure and Solid-State Physics, University of Hamburg, Hamburg, 22761, Germany. — [2]Institute for Materials Science, Faculty of Engineering, Kiel University, Kaiserstrasse 2, 24143, Kiel, Germany

Topological insulators (TIs) exhibit unconventional quantum phases that can be manipulated with external quantum confinements. The geometry of the surface of 3D TIs plays a crucial role in tuning the quantum confinement in TIs. For instance, morphing a 3D TI into a 1D cylinder results into a curved surface in which the electron spin is locked perpendicular to the momentum and, as well as, tangential to the perimeter of the TI nanowire. This leads to a new state with a Spin-Berry Phase (SBP). We utilized Surface-Enhanced Raman Scattering (SERS) as sensor to study the confinement of quasi-relativistic electrons in TI nanowires. Our result shows diameter-dependent SERS in nanowires which is attributed to the self-interference effect of the electronic wavefunction along the curved surface of the nanowires. Rectangular shaped Nanoribbons do not show this effect. Plasmonic gold nanoparticles attached at the distinct topological surface states enables us to discriminate different subband splitting between nanowires and nanoribbons.

O 11.10 Mon 17:30 MA 004

Quantum simulator to emulate lower-dimensional molecular structure — •Emil Sierda, Xiaochun Huang, Danis Badrtdinov, Brian Kiraly, Elze Knol, Anna M.H. Krieg, Gerrit C. Groenenboom, Mikhail I. Katsnelson, Malte Rösner, Daniel Wegner, and Alexander A. Khajetoorians — Institute for Molecules and Materials, Radboud University, Nijmegen, The Netherlands

Bottom-up quantum simulators have been developed to quantify the role of various interactions, dimensionality, and structure in creating electronic states of matter. Here, we demonstrated a solid-state quantum simulator emulating molecular orbitals, based solely on positioning individual cesium atoms on an indium antimonide surface. Using scanning tunneling microscopy and spectroscopy, combined with ab initio calculations, we showed that artificial atoms could be made from localized states created from patterned cesium rings. These artificial atoms served as building blocks to realize artificial molecular structures with different orbital symmetries. These corresponding molecular orbitals allowed us to simulate two-dimensional structures reminiscent of well-known organic molecules. The platform could further be used to monitor the interplay between atomic structures and the resulting molecular orbital landscape with submolecular precision.

O 12: 2D Materials I: Electronic Structure (joint session O/TT)

Time: Monday 15:00–17:30 Location: MA 005

O 12.1 Mon 15:00 MA 005

Electronic and magnetic properties of single chalcogen vacancies in MoS_2/Au(111) — Sergey Trishin[1], •Christian Lotze[1], Nils Krane[2], and Katharina J. Franke[1] — [1]Fachbereich Physik, Freie Universität Berlin, Germany — [2]nanotech@surfaces Laboratory, Empa - Swiss Federal Laboratories for Materials Science and Technology, Dübendorf, Switzerland

Two-dimensional (2D) transition-metal dichalcogenides (TMDCs) are considered highly promising platforms for next-generation optoelectronic devices. Because of their 2D structure, the performance of potential devices is strongly impacted by defects. Nowadays a lot of research aims to optimize growth methods towards defect-free TMDCs. However, defect engineering has also gained a lot of attention, as it may allow for control and design of new properties of 2D materials.

Here, we create single S vacancies in a monolayer of MoS_2 that was grown on Au(111). Using combined scanning tunneling and atomic force microscopy, we show that these defects are negatively charged and give rise to a Kondo resonance. The latter reveals the presence of an unpaired electron spin that is exchange-coupled to the underlying metal substrate. The strength of the exchange coupling depends on the density of states at the Fermi level, which is modulated by the moiré structure of the MoS_2 lattice and the Au(111) substrate. We also show that in the absence of direct hybridization of MoS_2 with the metal substrate, the S vacancy remains charge-neutral. Our results suggest that defect engineering may be used to induce and tune magnetic properties of otherwise nonmagnetic materials.

O 12.2 Mon 15:15 MA 005

Non-ambient Raman spectroscopy combined with ion bombardment — •André Maas, Joel Verlande, Leon Daniel, Lucia Skopinski, Lars Breuer, and Marika Schleberger — Universität Duisburg-Essen, Fakultät für Physik and CENIDE, Germany

Characterizing materials in non-ambient conditions poses a persistent challenge, particularly in understanding irradiation-induced defects and their effects on crystal structure and electronic/optoelectronic properties. While ambient conditions often saturate defects with adsorbates, investigating the influence of unsaturated defects remains crucial for a comprehensive understanding.

In the first part of this study, we focused on analyzing defect formation in CVD-grown WS_2 by irradiating it with low-energy Ar^+ ions ($E_{\mathrm{kin}} \leq 500$ eV) to create sulfur vacancies. Subsequent analysis via Raman and photoluminescence spectroscopy revealed insights into the nature of these defects. A custom-built cell was used enabling us to characterize the electrical and optical properties at a pressure of $p = 10^{-6}$ mbar, to study the effects of temperature (77 K to 600 K) and the presence of sulfur hexafluoride on the irradiated samples.

In a novel experiment, defects were induced in graphene using highly charged ions (Xe^{17+} - Xe^{40+} at around $E_{kin} = 200$ keV). In-situ optical measurements at ultra-high vacuum ($p = 10^{-9}$ mbar) allowed us to detect the influence of saturated defects on the properties of this important 2D material.

O 12.3 Mon 15:30 MA 005

Local creation and manipulation of sulfur vacancies in two-dimensional MoS_2 — •Daniel Jansen[1], Tfyeche Tounsi[1], Jeison Fischer[1], Arkady Krasheninnikov[2], Thomas Michely[1], Hannu-Pekka Komsa[3], and Wouter Jolie[1] — [1]II. Physikalisches Institut, Universität zu Köln, Köln, Germany — [2]Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany — [3]Faculty of Information Technology and Electrical Engineering, University of Oulu, Oulu, Finland

Point defects in two-dimensional semiconductors can exhibit spatially confined and electronically isolated quantum states in the band gap of their host material. A prerequisite for the use of such point defects in quantum applications is to gain control over defect creation and manipulation.

Here, we report on a new technique for the local creation of individual sulfur vacancies in two-dimensional MoS_2 involving the tip of a scanning tunneling microscope and single Fe adatoms that we utilize as chemical markers. We exemplify how this technique can be employed to tailor the in-gap states by the creation of a vacancy dimer, giving rise to hybrid orbitals. Additionally we show that the tip can also be used to manipulate the sulfur vacancy charge state through local gating. When negatively charged, two distinct Jahn-Teller distortions are observed and characterized using scanning tunneling spectroscopy and density functional theory.

O 12.4 Mon 15:45 MA 005

Charge State-Dependent Symmetry Breaking of Atomic Defects in Transition Metal Dichalogenides — •Lysander Huberich[1], Feifei Xiang[1], Jonas Allerbeck[1], Preston A. Vargas[2], Riccardo Torsi[3], Anne Marie Tan[2], Pascal Ruffieux[1], Roman Fasel[1], Oliver Gröning[1], Yu-Chuan Lin[3], Richard Henning[2], Joshua Robinson[3], and Bruno Schuler[1] — [1]Empa - nanotech@surfaces Laboratory, 8600 Dübendorf, Switzerland — [2]University of Florida, Gainesville, FL, 32611, USA — [3]The Pennsylvania State University, University Park, PA, 16802, USA

The electronic properties of atomically thin 2D materials are strongly influenced by occuring atomic defects and their interaction with the host lattice. Here we report on the direct imaging of charge state-dependent symmetry breaking of single rhenium dopants (Re_{Mo}) and negatively charged sulfur vacancies (${Vac_S}^-$) in mono- and bilayer MoS_2 by atomically-resolved STM and nc-AFM. While ${Vac_S}^-$ occur in both the symmetric and the symmetry-broken state, Re_{Mo} exhibit charge-dependent symmetry breaking stabilized by the difference in substrate workfunction. The local lattice distortions and symmetry-broken defect orbitals of ${Vac_S}^-$ as well as ${Re_{Mo}}^0$ and ${Re_{Mo}}^{-1}$ are attributed to the (pseudo-)Jahn-Teller effect. By mapping of electronic orbitals and geometric structures, we can disentangle effects of spatial averaging, charge multistability, configurational dynamics, and external perturbations that often mask the presence of local symmetry breaking. [Xiang, Huberich, et al., arXiv:2308.02201]

O 12.5 Mon 16:00 MA 005

Tuning Intrinsic Transition Probabilities in CVD-Grown WS_2 through Introduction of Sulfur Vacancies — •Leon Daniel, Charleen Lintz, Osamah Kharsah, Andre Maas, Stephan Sleziona, and Marika Schleberger — Universität Duisburg-Essen

Monolayer transition metal dichalcogenides (TMDCs) like tungsten disulfide (WS_2) are highly interesting materials for optoelectronic and valleytronic applications. Although WS_2 has a significantly higher quantum efficiency compared to its MoS_2 counterparts, its intrinsic properties are much less studied. This study investigates the controlled creation of sulfur vacancies in chemical vapor deposition (CVD)-grown WS_2 by high-temperature annealing in vacuum conditions up to 627 K. Employing in-situ photoluminescence (PL) spectroscopy, we observe a selective reduction in the intensity of the A exciton, while the B exciton remains unaffected upon introduction of vacancies. This distinctive behavior provides valuable insights into intrinsic transition probabilities associated with deliberately induced defect levels in WS_2. Additionally, an increased trion emission was detected, indicating increased doping by selectively removing sulfur. Intriguingly, despite the vacancy introduction, no observable localized states are detected. Similar results are achieved through 100 eV argon ion irradiation, if the sample is annealed with high laser powers before PL measurements are conducted; we therefore attribute this to the desorption of adsorbates. Our findings suggest that low energy ion bombardment is a suitable option for selectively tailoring the material's optical properties.

O 12.6 Mon 16:15 MA 005

Formation of complex CDW patterns in single-layer TiSe2 — •Wen Wan, Paul Dreher, Maria Gastiasoro, Fernando de Juan, and Miguel Ugeda — Donostia International Physics Center, San Sebastián, Spain

While the CDW in TiSe2 emerges naturally with a commensurate 2x2x2 symmetry below ~200 K, superconductivity only develops induced by pressure, doping, and intercalation. Interestingly, these external stimuli triggering superconductivity also induce a CDW phase transition from a commensurate (CCDW) to an incommensurate state (ICCDW). Therefore, it has been speculated that superconductivity in TiSe2 is triggered by the CCDW to ICCDW transition in an unprecedented, exotic mutual interplay.

In this work, we explore the CDW state of TiSe2 in the single-layer limit by means of low-temperature STM/STS measurements. nm-scale

STM imaging reveals the presence of a CDW with a local 2x2 atomic periodicity as in bulk TiSe2 which, in contrast, is markedly inhomogeneous in space. On a larger scale, we find that such inhomogeneity is due to incommensuration of the CDW, which leads to the formation of complex 4-fold 2D matrix of CDW domains. Our high-resolution large-scale STM images allow us to spatially map the full complex CDW order parameter (amplitude and phase) with sub-nm precision. This analysis reveals that the CDW develops π-phase shifts at the domain walls. Lastly, while the matrix of CDW domains is unaffected by point defects in the Se layer, artificially introduced defects in the Ti plane pin the domain walls.

O 12.7 Mon 16:30 MA 005

Unconventional charge-density-wave gap in monolayer NbS$_2$ — Timo Knispel[1], Jan Berges[2], Arne Schobert[3], Erik van Loon[4], Wouter Jolie[1], Tim Wehling[3], Thomas Michely[1], and •Jeison Fischer[1] — [1]II. Physikalisches Institut, Universität zu Köln, Köln — [2]Universität Bremen, Bremen — [3]I. Institut für Theoretische Physik, Universität Hamburg, Hamburg — [4]Lund University, Lund, Sweden

Here, we report scanning tunneling microscopy and spectroscopy measurements for a monolayer of H-NbS$_2$ grown by molecular beam epitaxy on graphene/Ir(111). We find that monolayer NbS$_2$ displays a 3×3 modulation superstructure due to a charge density wave (CDW), which is not present in bulk NbS$_2$. Evidence for the CDW is given by bias voltage contrast inversion and temperature suppression of the CDW signal. Our high-resolution differential conductance spectra display a pronounced gap of the order of 20 meV at the Fermi level. Within the gap low energy features are present. The gap structure with its low energy features is at variance with the expectation for a gap opening in the electronic band structure due to a static CDW distortion. Instead, comparison with ab initio calculations indicates that the observed gap structure is due to combined electron-phonon quasiparticles. The phonons in question are the elusive amplitude (Higgs) and phase (Goldstone) collective modes of the CDW transition [1].

[1] Knispel et al., arXiv:2307.13791.

O 12.8 Mon 16:45 MA 005

Orbital character and ground-state electronic properties in van der Waals semiconductors VI3 and CrI3 — •Alessandro De Vita[1,2], Thao Nguyen[3], Roberto Sant[4], Gian Marco Pierantozzi[1], Danila Amoroso[5], Chiara Bigi[1,6], Vincent Polewczyk[1], Giovanni Vinai[1], Loi Nguyen[7], Tai Kong[7], Jun Fujii[1], Ivana Vobornik[1], Nicholas Brookes[4], Giorgio Rossi[1,2], Robert Cava[7], Federico Mazzola[1], Kunihiko Yamauchi[3], Silvia Picozzi[5], and Giancarlo Panaccione[1] — [1]IOM-CNR, Laboratorio TASC, Trieste, Italy — [2]Dipartimento di Fisica, Università di Milano, Italy — [3]ISIR, Osaka University, Japan — [4]ESRF, Grenoble, France — [5]CNR-SPIN c/o Università G. D'Annunzio, Chieti, Italy — [6]University of St Andrews, United Kingdom — [7]Department of Chemistry, Princeton University, NJ USA

Layered van der Waals magnetic semiconductors CrI3 and VI3 hold promise for novel electronic and spintronic 'few-layers' functionalities; however, detailed experimental information on the electronic structure, the interplay with relevant interactions (e.g. spin-orbit coupling), and the crossover of 3D vs 2D properties, is lacking. By combining X-ray electron spectroscopies and density functional theory calculations, we report a complete characterization of CrI3 and VI3 electronic ground states. We show that the transition metal-induced orbital filling drives the stabilization of distinct phases. X-ray absorption measurements on VI3 reveal that its electronic properties are strongly influenced by dimensionality effects. Our results have direct implications in band engineering and layer-dependent properties of two-dimensional systems.

O 12.9 Mon 17:00 MA 005

Exchange splitting in the electronic structure of quasi-2D antiferromagnet CrSBr — •Matthew D. Watson[1], James Nunn[1,2], Swagata Acharya[3], Laxman Naga-Reddy[2], Dimitar Pashov[4], Malte Rösner[5], Mark van Schilfgaarde[3], Neil R. Wilson[2], and Cephise Cacho[1] — [1]Diamond Light Source Ltd, Harwell Science and Innovation Campus, Didcot, OX11 0DE, UK — [2]Department of Physics, University of Warwick, Coventry, CV4 7AL, UK — [3]National Renewable Energy Laboratory, Golden 80401 CO, USA — [4]Theory and Simulation of Condensed Matter, King's College London, The Strand, London WC2R2LS, UK — [5]Institute for Molecules and Materials, Radboud University, Heijendaalseweg 135, 6525AJ Nijmegen, The Netherlands

We present the evolution of the electronic structure of CrSBr from it's antiferromagnetic ground state to the paramagnetic phase above $T_N =$ 132 K, in both experiment and theory. The ground state angle-resolved photoemission spectroscopy (ARPES) results, obtained using a novel method to overcome sample charging issues, are very well reproduced by our QSGW calculations including Bethe-Salpeter Equations (BSE) self-consistently. By tracing band positions as a function of temperature, we identify certain bands at the X points to be exchange-split pairs of states with mainly Br and S character, with the splitting disappearing above T_N. Our results lay firm foundations for the interpretation of the many other intriguing physical and optical properties of CrSBr.

O 12.10 Mon 17:15 MA 005

Ultrafast momentum microscopy of hybrid exciton dynamics in homobilayer 2H-MoS$_2$ — •Paul Werner[1], Jan Philipp Bange[1], Wiebke Bennecke[1], David Schmitt[1], Giuseppe Meneghini[2], Anna Seiler[1], AbdulAziz AlMutairi[3], Marco Merboldt[1], Sabine Steil[1], Daniel Steil[1], Stephan Hofmann[3], G. S. Matthijs Jansen[1], Samuel Brem[2], R. Thomas Weitz[1], Ermin Malic[2], Marcel Reutzel[1], and Stefan Mathias[1] — [1]I. Physikalisches Institut, Georg-August Universität Göttingen, Germany — [2]Fachbereich Physik, Philipps-Universität Marburg, Germany — [3]Department of Engineering, University of Cambridge, United Kingdom

Transition-metal dichalcogenides (TMDs) monolayers host a rich landscape of excitonic states. If, in addition, these monolayers are stacked on top of each other, novel interlayer and hybrid excitonic states can form. Hybrid excitons, where either the excitons' hole or electron is layer-delocalized as a result of interlayer hopping, are responsible for efficient charge transfer between the layers [1, 2]. In homobilayer MoS$_2$, hybrid excitons are predicted to be the energetically most favorable excitonic state, making it an ideal system to study their properties. We employ time-resolved momentum microscopy to study the ultrafast exciton dynamics in H-stacked homobilayer MoS$_2$. By directly imaging the electron and hole contributions of the hybrid excitons, we are able to track their ultrafast dynamics.

[1] Schmitt *et al.*, Nature **608**, 499-503 (2022)

[2] Bange, Werner *et al.*, 2D Mater. *10* 035039 (2023)

O 13: Ultrafast Electron Dynamics at Surfaces and Interfaces II

Time: Monday 15:00–17:30 Location: MA 041

Topical Talk O 13.1 Mon 15:00 MA 041

Photocatalysis at the surface the TiO_2 on its real time — Michael Wagstaffe[1], Lukas Wenthaus[2], Adrian Dominguez-Castro[3], Dmytro Kutnyakhov[2], Siarhei Dziarzhytski[2], Simon Chung[1], Guilherme Dalla Lana Semione[1], Steffen Palutke[2], Federico Pressacco[2], Michael Heber[2], Giuseppe Mercurio[2], Harald Redlin[2], Helena Gleissner[1], Verena Kristin Gupta[3], Nicolai Klemke[4], Yudong Yang[4], Thomas Frauenheim[5], Adriel Dominguez[6], Franz Kärtner[4], Angel Rubio[7], Wilfried Wurth[2], Andreas Stierle[1], and •Heshmat Noei[1] — [1]Centre for X-ray and Nanoscience (CXNS), Deutsches Elektronen-Synchrotron (DESY), Hamburg, Germany — [2]Deutsches Elektronen-Synchrotron (DESY), Notkestr. 85 D-22607, Hamburg, Germany — [3]Bremen Center for Computational Material Science (BCCMS), Uni- versity of Bremen, Bremen, Germany — [4]Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany — [5]Computational Science and Applied Research Institute (CSAR), 518110, Shenzhen, China — [6]Nano-Bio Spectros- copy Group, Departamento de Fisica de Materiales, San Sebastián, Spain; — [7]Max Planck In- stitute for the Structure and Dynamics of Matter, Hamburg, Germany

The femtosecond resolution soft X-ray photoemission spectroscopy results are combined with theoretical calculations to provide crucial insight concerning reaction mechanisms and dynamics of the interaction of water, oxygen and carbon monoxide with the surface of TiO_2 and the interfacial charge transfer during the initial steps of the reaction.

O 13.2 Mon 15:30 MA 041

Photo-induced charge-transfer renormalization in NiO — •T. Lojewski[1], D. Golez[2,3], K. Ollefs[1], L. Le Guyader[4], L. Kämmerer[1], N. Rothenbach[1], R. Y. Engel[5], P. S. Miedema[5], M. Beye[5], G. S. Chiuzbaian[6], R. Carley[4], R. Gort[4], B. E. Van Kuiken[4], G. Mercurio[4], J. Schlappa[4], A. Yaroslavtsev[4,7], A. Scherz[4], F. Döring[8], C. David[8], H. Wende[1], U. Bovensiepen[1,9], M. Eckstein[10], P. Werner[11], and A. Eschenlohr[1] — [1]Univ. Duisburg-Essen and CENIDE — [2]Jozef Stefan Inst. — [3]Univ. of Ljubljana — [4]European XFEL — [5]DESY — [6]Sorbonne Univ. — [7]Uppsala Univ. — [8]PSI — [9]Univ. of Tokyo — [10]Univ. of Hamburg — [11]Univ. of Fribourg

Strongly correlated materials, such as the charge transfer insulator NiO, exhibit fascinating properties due to the interaction-induced localisation of electrons, which competes with their itinerant nature. Here, optical excitation of charge carriers, i.e. photodoping, results in complex dynamics involving both *d*-*p* and *d*-*d* excitations. Through a resonant optical pump, femtosecond time-resolved X-ray absorption probe experiment in combination with dynamical mean-field theory, we analyse these dynamics in photo-doped NiO. We find a redshift of the Ni L_3 and O K edges (persisting for > 10 ps), relating to a simultaneous occurrence of Hartree shifts and a renormalisation of local interactions. In addition, we observe a feature below the Ni L_3 edge (at < 1 ps), which we relate to a transient nonthermal population of local many-body multiplets [1]. Financial support by DFG through SFB 1242 is acknowledged. - [1] T. Lojewski et al., arXiv:2305.10145

O 13.3 Mon 15:45 MA 041

Spatio-Temporal Electron Propagation Dynamics in Au/Fe/MgO(001) in Nonequilibrium Analyzed by Femtosecond Two-Photon Photoemission — •Florian Kühne[1], Markus Heckschen[1], Yasin Beyazit[1], Elaheh Shomali[1], Jesumony Jayabalan[1], Ping Zhou[1], Detlef Diesing[2], Markus Gruner[1], Rossitza Pentcheva[1], Axel Lorke[1], Björn Sothmann[1], and Uwe Bovensiepen[1] — [1]Fakultät für Physik, Universität Duisburg-Essen and CENIDE, Duisburg D-47048, Germany — [2]Fakultät für Chemie, Universität Duisburg-Essen and CENIDE

Optically excited electrons in metals travel ballistically on a femtosecond timescale until they scatter due to electron-electron or electron-phonon interactions. To gain a deeper understanding of the ballistic and few times scattered electrons under non-equilibrium conditions we combine time-resolved two-photon photoelectron emission spectroscopy with real-time time-dependent density functional theory(RT-TDDFT) as well as a random-walk-like transport simulation, see Heckschen et al., PRX Energy **2**, 043009(2023). Here, we discuss the experimental details using a back-side pumped geometry on a MgO/Fe/Au epitaxial heterostructure probing the Au surface. The time-delayed response of the photoelectrons at lower energies is used to investigate transport and scattering pathways. Analyzing this time delay of 10 to 100fs shows an apparent acceleration with increasing film thickness. We find the electron trajectories angular dependence is of key importance to explain this effect. Funding by the DFG through Project No. 278162697 - SFB1242 is gratefully acknowledged.

O 13.4 Mon 16:00 MA 041

Spatio-Temporal Electron Propagation Dynamics in Au/Fe/MgO(001) in Nonequilibrium: A Random Walk Simulation — •Markus Heckschen[1], Yasin Beyazit[1], Elaheh Shomali[1], Florian Kühne[1], Jesumony Jayabalan[1], Ping Zhou[1], Detlef Diesing[2], Markus E. Gruner[1], Rossitza Pentcheva[1], Axel Lorke[1], Björn Sothmann[1], and Uwe Bovensiepen[1] — [1]Fakultät für Physik, Universität Duisburg-Essen and CENIDE, Duisburg D-47048, Germany — [2]Fakultät für Chemie, Universität Duisburg-Essen and CENIDE, Essen D-45711, Germany

Electron transport in metals can be either ballistic, superdiffusive or diffusive. We analyze time resolved two-photon photoelectron emission (2PPE) spectroscopy performed in a back-pump front-probe geometry on MgO/Fe/Au which provides information about the nonequilibrium electron distribution as a function of time, energy and Au layer thickness [1].

Here, we present a trajectory-based Monte-Carlo simulation of classical, quasiballistic electrons that undergo inelastic electron-electron scattering. Our simulation reproduces well the experimentally observed 2PPE spectra and allows us to identify high-energy electrons passing ballistically through the sample, as well as low-energy electrons which are transported in a superdiffusive manner.

[1] M. Heckschen et al., PRX Energy **2**, 043009 (2023).

O 13.5 Mon 16:15 MA 041

Thermal boundary conductance of ultrathin epitaxial Pb films on Si(111) — •Christian Brand[1], Tobias Witte[1], Mohammad Tajik[1], Jonas D. Fortmann[1], Michael Horn-von Hoegen[1], Laurenz Rettig[1,2], and Uwe Bovensiepen[1] — [1]University of Duisburg-Essen, Duisburg, Germany — [2]Fritz Haber Institute, Berlin, Germany

The non-equilibrium dynamics of electrons in a metal subsequent to excitation with a fs-laser pulse couple to other degrees of freedom during the relaxation phase of the system. Here we have studied the electron and lattice dynamics in ultrathin Pb films grown on Si(111) by means of time-resolved photoemission spectroscopy and reflection high-energy electron diffraction. After thermalization of the electron and lattice system, the transient cooling of the film is determined by heat transport across the interface into the substrate, i.e., by the thermal boundary conductance (TBC) on time scales of a few 100 ps. We experimentally find for crystalline Pb films with low defect density at a base temperature of 19.3 K a TBC of less than 2 MW/m^2K. We discuss the results in terms of diffuse and acoustic mismatch models.

O 13.6 Mon 16:30 MA 041

Electron-phonon coupling in ultrathin Pb films on Si(111): Where the heck is the energy? — •Mohammad Tajik[1], Tobias Witte[1], Christian Brand[1], Laurenz Rettig[2], Björn Sothmann[1], Uwe Bovensiepen[1], and Michael Horn-von Hoegen[1] — [1]Department of Physics, Universität Duisburg-Essen, Lotharstraße 1, 47057 Duisburg, Germany — [2]Department of Physical Chemistry Fritz Haber Institute Faradayweg 4-6 14195 Berlin Germany

In this work, we studied the heat transfer from electron to phonon subsystem within a five monolayer thin epitaxial Pb film on Si(111) upon fs-laser excitation. The response of the electron subsystem was determined using time-resolved photoelectron spectroscopy (tr-PES) while the lattice excitation was measured by means of the Debye-Waller effect in time-resolved reflection high-energy electron diffraction (tr-RHEED). The electrons lose their heat in less than 0.7 ps while the lattice temperature rises slowly in 3.5 to 8 ps. This raises the question where is the energy hidden for 3-7 ps? Within a three-temperature model we used three heat baths, namely electrons, high-frequency and low-frequency phonon modes to simulate the observations. We propose that the hidden energy is transiently stored in high-frequency phonon modes at the zone boundary for which the tr-RHEED is insensitive and which are excited in less than 0.7 ps. The excitation of low-frequency acoustic phonons, i.e., thermalization of the lattice is facilitated through anharmonic phonon-phonon interaction.

O 13.7 Mon 16:45 MA 041

Non-Equilibrium Pathways for Excitation of Bulk and Surface Phonons through Anharmonic Coupling — C. Brand[1], V. Tinnemann[1], A. Hanisch-Blicharski1[1,3], M. Tajik[1], •J. D. Fortmann[1], A. Kassen[1], F. Thiemann[1], and M. Horn-von Hoegen[1,2] — [1]Department of Physics, University of Duisburg-Essen, 47048 Duisburg, Germany — [2]Center for Nanointegration (CENIDE), 47048 Duisburg, Germany — [3]present address: Institut für IT-Management & Digitalisierung, FOM Hochschule, Germany

Upon impulsive optical excitation of solid-state materials, the non-equilibrium flow of energy from the excited electronic system to the lattice degrees of freedom typically happens in a few picoseconds. Here we identified the surface of thin Bi films grown on Si(001) as an additional subsystem which is excited much slower on a 100 ps timescale that is caused by decoupling due to mismatched phonon dispersions relations of bulk and surface. Anharmonic coupling among the phonon systems provides pathways for excitations which exhibits a 1/T-dependence causing a speed-up of surface excitation at higher temperatures. A quantitative justification is provided by phonon Umklapp processes from lattice thermal conductivity of the Bi bulk. Three-temperature model simulations reveal a pronounced non-equilibrium situation up to nanoseconds: initially, the surface is colder than the bulk, that situation is then inverted during cooling and the surface feeds energy back into the bulk phonon system.

O 13.8 Mon 17:00 MA 041

Temporal evolution of energy-resolved non-thermal electron densities — •Christopher Seibel, Markus Uehlein, Tobias Held, Sebastian T. Weber, and Baerbel Rethfeld — Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau

When a metal is irradiated with an ultrashort laser pulse, the initially Fermi-distributed electrons are excited to a state far from equilibrium. These hot electrons can be exploited for numerous processes and applications, such as photodetection, solar energy conversion, and photocatalysis. However, the non-thermal electrons rapidly thermalize by collisions with each other, thereby limiting their availability for specific applications. Thus, it is crucial to understand the microscopic processes that determine the timescales of hot electron thermalization.

Here, we use a kinetic model based on full Boltzmann collision integrals to trace the non-equilibrium dynamics of the electronic distribution function during excitation and thermalization. We evaluate the time-dependent electron densities in various energy intervals and show the dependence of the dynamics of these spectral electron densities on the excitation conditions. We find energy regions where the interplay between primary and secondary electron generation leads to a behavior revealing a long-lasting non-equilibrium that cannot be explained with a single relaxation time [1].

[1] C. Seibel *et al.*, J. Phys. Chem. C (2023), DOI: 10.1021/acs.jpcc.3c04581

O 13.9 Mon 17:15 MA 041

Band-resolved relaxation of laser-excited gold — •Tobias Held, Stephanie Roden, Pascal D. Ndione, Sebastian T. Weber, and Baerbel Rethfeld — Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau

When a noble metal is irradiated with a short-pulsed laser in the visible spectrum, sp- and d-electrons are excited into energetically higher free states while the phonons are not directly affected. This process increases the energy content of the electron system, alters the partial electron densities and induces non-equilibrium electron distributions. We investigate how the non-equilibrium evolves in the individual bands towards Fermi distributions using the Boltzmann equation with full Boltzmann collision integrals for the excitation, electron-electron scattering and electron-phonon scattering, respectively.

Following the fully kinetic stage, when a temperature description is valid, an occupational non-equilibrium can still persist between sp- and d-electrons [1]. We investigate how this occupational non-equilibrium affects the electron-phonon coupling parameter, finding a strong dependence of the coupling strength on the band occupation [2]. We also observe signatures of features of the partial densities of states in the coupling parameter.

[1] P. D. Ndione, S. T. Weber, D. O. Gericke and B. Rethfeld, Scientific Reports 12, 1 (2022)

[2] T. Held, S. T. Weber and B. Rethfeld, arXiv:2308.01067 (2023)

O 14: Plasmonics and Nanooptics II: Light-Matter Interaction and Spectroscopy

Time: Monday 15:00–17:30 Location: MA 042

O 14.1 Mon 15:00 MA 042

Towards Isolated Attosecond Electron Trios — Germann Hergert, •Rasmus Lampe, Andreas Wöste, and Christoph Lienau — Institut für Physik, Carl-von-Ossietzky Universität, 26129 Oldenburg, Germany

Very recently, the formation of multi-electron number states in the laser-triggered photoemission from metal tips has created much attention [1,2]. These number-states show intriguing properties, like highly correlated electron energies and increasing nonlinearities in their power scaling. Here, we use, for the first time, few-cycle near-infrared driving pulse around 2000 nm for the creation of such number states. The low photon energy, 1/7 of the work function of gold, drastically increases the nonlinear order of the emission process, up to the 21st order for multiphoton-emission of a three-electron state.

Such extreme nonlinearities make the emission process exceptionally sensitive to minor changes in the driving field. We demonstrate this by monitoring the electron yield while varying the delay of a phase-locked pulse pair. We observe autocorrelations that are fully governed by an isolated central peak with 0.8 fs width. Only two side peaks with amplitudes $< 25\%$ are seen. This may enable the generation of isolated attosecond electron trios. We are currently studying the CEP dependence of the emission to gain further insight into the emission regime.

[1] S. Meier et al., Nat. Phys. 19, 1402 (2023)

[2] R. Haindl et al., Nat. Phys. 19, 1410 (2023)

O 14.2 Mon 15:15 MA 042

Soliton Formation Dynamics in a Microresonator Probed by Electron Energy Gain Spectroscopy — •F. Jasmin Kappert[1], Yujia Yang[2], Jan-Wilke Henke[1], Arslan S. Raja[2], Germaine Arend[1], Guanhao Huang[2], Armin Feist[1], Zheru Qiu[2], Rui Ning Wang[2], Aleksandr Tusnin[2], Alexey Tikan[2], Tobias J. Kippenberg[2], and Claus Ropers[1] — [1]Max Planck Institute for Multidisciplinary Sciences, Göttingen,Germany — [2]Institute of Physics, Swiss Federal Institute of TechnologyLausanne (EPFL), Switzerland

Ultrafast electron microscopes provide a powerful platform for investigating confined optical modes with high temporal and spatial resolution. Recently, integrated photonics has boosted the interaction strength of light with free electrons, enabling nanoscale spectroscopy of optical states with a continuous electron beam.

Here, we use continuous-beam electron energy gain spectroscopy (EEGS) to probe nonlinear optical excitations inside a microresonator [1]. Starting from single-color excitation, increasing the optical input power leads to the formation of dissipative Kerr solitons via various nonlinear comb states, evident in characteristic optical and EEG spectra. This scheme enables non-invasive probing of nonlinear optical dynamics with direct access to the intracavity field. Moreover, the interaction with solitons facilitates high-frequency spatiotemporal electron beam modulation.

[1] Y. Yang, et al., arXiv:2307.12142 (2023)

Topical Talk O 14.3 Mon 15:30 MA 042

Time-resolved interaction of the electron system with strong surface plasmon polariton fields — •Frank Meyer zu Heringdorf — Faculty of Physics, University of Duisburg-Essen, 47048 Duisburg, Germany

Time-resolved experiments often rely on femtosecond laser pulses to create a non-equilibrium situation in the electron system of a surface. The specifics of the created nonequilibrium hereby depends on the detailed spatiotemporal properties of the excitation field. It has been demonstrated that by controlling surface plasmon polaritons (SPPs) it is possible to create unique (even topologically non-trivial) polarization fields at surfaces. Experimental knowledge of the SPP vector field on a few hundred nm length- and a fs time-scale can be obtained using vector microscopy and polarimetry, where time-resolved, probe-polarization-dependent two photon photoemission data is obtained in a spectroscopic low energy electron microscope (SPE-LEEM). Using Archimedean spirals for excitation, strong SPP foci can be created that exhibit highly-nonlinear electron emission, and the ponderomotive interaction of the electron with the SPP field during the photoemission process can be used to quantify the field strength. Time- and angle-resolved experiments at a SPP focus clarify the intermediate states involved in the emission process and demonstrate that SPPs can coherently couple to electronic states in above-threshold electron emission.

O 14.4 Mon 16:00 MA 042

Electron driven photon sources for engineered generation of light propagating in Free Space — •Masoud Taleb and Nahid Talebi — Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, Germany

Optical beams with angular momentum have been intensively investigated for offering an additional degree of freedom for information transfer or manipulating the chiral degrees of freedom in matter. Such beams are generally realized via metasurfaces, photon sieves, or structured lenses. However, most of them lack key properties in controlling the polarization, directionality and intensity of the light which is generated and directed from a localized radiation sources such as electron beam induced emission. Previously, we have realized electron-driven photon sources which are fabricated by specific orders of nanoholes in a thin layer of gold (Nano Lett. 2020, 20, 8, 5975*5981). Here, we further elucidate on a variety of degrees of freedom in our design principle to both control the polarization and the shape of the generated light. A moving electron at the kinetic energy of 30 kV interacts with the system acting as a broadband source of optical excitation. Coupled plasmon polaritons propagate on the lattice forming a chain plasmon polaritons, that further radiates to the far field and generates a vortex beam. This phenomenon is experimentally studied utilizing cathodoluminescence spectroscopy and angle-resolved mapping.

O 14.5 Mon 16:15 MA 042

Tuning Disorder-Driven Localization of Plasmons in Random Assemblies of Gold Nanoparticles — •Mohammed Fayis Kalady[1,2], Johannes Schultz[1], Kristina Weinel[1,3], Daniel Wolf[1], and Axel Lubk[1,4] — [1]Leibniz Institute for Solid State and Materials Research (IFW) Dresden, Helmholtzstraße 20, 01069 Dresden, Germany — [2]Indian Institute Of Technology (IIT) Delhi, Hauz Khas, New Delhi, Delhi 110016, India — [3]Federal Institute of Materials Research and Testing (BAM), Unter den Eichen 87, 12205 Berlin, Germany — [4]Institute of Solid State and Materials Physics, Dresden University of Technology, Haeckelstraße 3, 01069 Dresden, Germany

It is well-known that assemblies of plasmonic NPs support hybridized modes of localized surface plasmons, which delocalize in geometrically well-ordered arrangements. Here, we study the hybridization behaviour in geometrically completely disordered systems of Au NPs fabricated by a newly developed e-beam synthesis method that facilitates the production and arrangement of NPs of different sizes and distances. Employing state-of-the-art STEM Electron Energy Loss Spectroscopy in combination with numerical simulations, we reveal the spatial and spectral distribution of the coupled LSP modes. We show, amongst others, that their disorder-driven localization behaviour crucially depends on the thickness of NPs, exhibiting a transition in the energy dependence of the localization at a critical thickness of about 10 nm. Employing numerical simulations, we discuss this behaviour with respect to the size and distance distribution of the NPs.

O 14.6 Mon 16:30 MA 042

Exploring the limits of refractive index sensing using Mie voids — •Micha Kappel, Serkan Arslan, Michelle Pfahl, Mario Hentschel, and Harald Giessen — 4th Physics Institute and Research Center SCoPE, University of Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany

The research of resonant phenomena using dielectric nanostructures, described by Mie's scattering theory, has enable new technologies in the field of nanophotonics. Resonant structures such as Mie voids have recently been realized, but have not been characterized in terms of their sensing potential.

To investigate their resonant features, the reflectance spectra are recorded via microspectroscopy. By inducing known refractive index changes inside the Mie voids by temperature change or liquid mixtures, the spectral shift of the resonances can be tracked by comparing the spectra at each refractive index. Comparing the resonances positions to a linear fit yields us the smallest measurable refractive index change.

For a 10 by 10 array of Mie voids with an overall volume of 11 fL, the smallest measurable refractive index change Δn is 0.00029 RIU. For a single Mie void with a volume of 520 aL, it is possible to detect a refractive index change Δn of 0.0049 RIU.

Using dielectric Mie voids we have realized the refractive index sensor the smallest sensing volume in the range of hundreds of attoliters. By using this technology, new applications in the field of biology and medicine could become possible, such as detecting neurotransmitters from single vesicles.

O 14.7 Mon 16:45 MA 042

Emission from propagating Bloch surface wave polaritons — •Sebastian Henn, Andreas Müller, Marius Grundmann, and Chris Sturm — Felix-Bloch-Institut für Festkörperphysik, Fakultät für Physik und Erdsystemwissenschaften, Uni Leipzig, Leipzig, Deutschland

Bloch surface waves (BSW) correspond to photonic modes propagating along the interface between a dielectric multilayer and the ambient. Due to their evanescent field on both sides of the surface they cannot couple to the electromagnetic far-field of the ambient yielding high lateral propagation lengths in the surface plane. This can be exploited to obtain long-range polaritons by strongly coupling BSW to excitons. In this contribution we demonstrate emission from propagating exciton-polariton states in ZnO. They are detected by diffractive outcoupling using line gratings, produced by electron-beam lithography, in the sample surface. By means of Fourier-imaged photoluminescence we are able to investigate the influence of the grating geometry on the emission behaviour and show propagation lengths of up to 30 μm for states of the lower polariton branch. This is makes Bloch surface wave polaritons interesting for on-chip polaritonic applications.

O 14.8 Mon 17:00 MA 042

Spatially resolved nonlinear plasmonics — •Florian Mangold, Johannes Schust, Mario Hentschel, Bettina Frank, and Harald Giessen — 4th Physics Institute, Research Center SCoPE, and IQST, University of Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany

The nonlinear behavior of plasmonic nanoantenna arrays can be investigated using nonlinear spectroscopy. In our contribution we introduce nonlinear spatially resolved spectroscopy, which is capable of imaging the k-space behavior and the spectral behavior of nonlinear nanoantenna signals, as well as to spatially resolve the nonlinear signal of single nanoantennas.

These additional abilities give us the possibility to spatially resolve the THG signal of gold nanoantenna arrays and investigate the homogeneity of the antenna field. Furthermore, we are able to spatially resolve the THG emission centers of the third-order mode and observe their response to tuning the wavelength over the resonance.

In addition, we discovered that by increasing the laser intensity, certain antennas in our array became exceptionally bright. By correlating our spatially resolved nonlinear image with structural SEM data, we can prove that these bright antennas have deformed into a peanut shape. Thus our NSRS setup enables the investigation of the nonlinear self-enhancement process of nanoantennas under intense laser heating. We also simulated these peanut shaped antennas and could observe that these 3-dimensional structures lead to the enhancement of the nonlinear emission.

O 14.9 Mon 17:15 MA 042

Mie Void Metasurfaces — •Michelle Pfahl, Benjamin Reichel, Serkan Arslan, Mario Hentschel, and Harald Giessen — 4th Physics Institute and Research Center SCoPE, University of Stuttgart, Pfaffenwaldring 57, 70569, Stuttgart, Germany

The field of nanooptics has developed into a subject of large interest as it addresses the manipulation of light on this microscopically small scale. Metasurfaces, which are based on the interaction of optical resonators with light, offer novel possibilities for compact optics replacing conventional optical devices as well as offering novel functionalities. Dielectric Mie voids have emerged as a new building block for these metasurfaces. Spherical air inclusions in high index materials allow to resonantly confine light, circumventing material loss. Thus, these structures show pronounced resonances even in the UV spectral range. We take the first step to implement Mie void based metasurfaces by realizing a blazed grating. Using microspectroscopy in the real as in the spatial Fourier domain, as well as carrying out diffraction experiments with monochromatic light, we confirm the asymmetric diffraction behavior of our gratings. Furthermore, we implemented an analytical model which reproduces our results well. Mie voids promise further interesting applications for metasurfaces, such as reflective lenses, which could even work in the deep UV spectral range.

O 15: Organic Molecules on Inorganic Substrates II: Electronic, Optical and other Properties

Time: Monday 15:00–18:00 Location: MA 043

O 15.1 Mon 15:00 MA 043

Investigation of the electronic structure of solvent based multispin bearing metallacrown CuCu4 molecules on a Au(111) surface — •Benedikt Baumann[1], Robert Ranecki[1], Lukas Bolz[1], Ellen Brennfleck[1], Stefan Hornig[1], Yves Kurek[1], Frederik Pütz[1], Philip Schreyer[1], Paul Steuernagel[1], Dominic Stephan[1], Robert Wendels[1], Stefan Lach[1], Eva Rentschler[2], and Christiane Ziegler[1] — [1]RPTU Kaiserslautern - Landau and Research Center OPTIMAS, Erwin-Schrödinger-Str. 56, 67663 Kaiserslautern, Germany — [2]Inst. of Inorganic and Analytical Chemistry, Johannes Gutenberg University Mainz, Germany

Transition metal complexes with exchange-coupled open-shell metal ions have attracted attention in the last decade. In such systems, spin relaxation time enhancement can provide a route to nanoscale spintronic devices. Because of their mostly low thermal stability, such molecules must be prepared by solvent based methods to investigate the electronic structure on metallic substrates,. We will demonstrate different UHV compatible preparation setups to prepare ultra-thin layers of the solvent based metallacrown complex CuCu4 [1] on a Au(111) surface. Furthermore, we show how a multi-method approach of vibrational spectroscopy and complete photoelectron spectroscopy (X-ray, UV, and inverse photoemission) can be used to demonstrate the integrity of the molecules during the adsorption process as well as the evolution of the electronic structure of this molecule in comparison with theoretical single molecule calculations.

[1] P. Happ et al. Phys. Rev. B 93, 174404 (2016)

O 15.2 Mon 15:15 MA 043

Influence of Core-Substitution on the Electronic Structure of Thiadiazole Derivatives — •Mohsen Ajdari[1], Felix Landwehr[1], Ronja Pappenberger[1], Marvin Hoffmann[2], Hendrik Hoffmann[3], Lukas Ahrens[3], Uwe H. F. Bunz[3], Andreas Dreuw[2], and Petra Tegeder[1] — [1]Physikalisch-Chemisches Institut — [2]Interdisziplinäres Zentrum für Wissenschaftliches Rechnen — [3]Organisch-Chemisches Institut, Universität Heidelberg

Thiadiazole derivatives, such as naphtothiadiazole (NTD) and benzobisthiadiazole (BBT) are of great interest for the use as electron acceptor in (opto)electronic applications.

In this study, vibrational and electronic high-resolution electron energy loss spectroscopy (HREELS) in combination with quantum-chemical calculations are utilized to investigate the influence of core-substitution via halogens or aromatic groups on the adsorption geometry and electronic structure of NTD and its halogenated derivatives (NTD- Cl/Br) as well as phenyl (BBT-Ph) and thiophene (BBT-Th) substituted BBT on Au(111).

Our findings indicate that core-substitution influences the electronic structure of the corresponding molecules by narrowing of the optical gap from 2.6 eV for NTD to 2.4 eV for NTD-Cl/Br, as well as a reduction from 2.2 eV for BBT-Ph to 1.8 eV for BBT-Th. Additionally, the first triplet state energies of NTD at 1.7 eV is reduced to 1.6 eV for NTD-Cl/Br and BBT-Ph at 1.2 eV to 0.7 eV for BBT-Th.

O 15.3 Mon 15:30 MA 043

Interaction between polyatomic molecules on layered surfaces beyond the dipole approximation — •Hsiao-Han Chuang, Ulf Saalmann, and Alexander Eisfeld — Max Planck Institute for the Physics of Complex Systems, Nöthnitzer Str 38, Dresden

For aggregates of molecular monomers above layered surfaces, the dipole approximation breaks down when the distance between monomers and surfaces is less than several nanometers. To overcome this problem, we employ macroscopic quantum electrodynamics and use the complete transition current density of the individual monomers. Within this framework, the resulting Master Equation for the excitonic degrees of freedom of the aggregate is derived. For example, we discuss the case of PTCDA on a KCL surface.

O 15.4 Mon 15:45 MA 043

Non-conductive NaCl films impact the charge configuration in a single molecule — •Moritz Frankerl[1], Laerte Patera[2,3], Thomas Frederiksen[4], Jascha Repp[3], and Andrea Donarini[1] — [1]Institute of Theoretical Physics, University of Regensburg — [2]Institute of Physical Chemistry, University of Innsbruck — [3]Institute of Experimental and Applied Physics, University of Regensburg — [4]Donostia International Physics Center (DIPC), Spain

The utilization of non-conductive NaCl films allows the stabilization of multiple charge states on a single molecule [1]. Consequently, these films enable the observation of electronic transitions between distinct charge states by single-electron alternate-charging scanning tunneling microscopy (AC-STM) [2]. Specifically, when charging a copper-phthalocyanine (CuPc) molecule, the Jahn-Teller (JT) distortion induces a splitting of the degeneracy between the lowest unoccupied molecular orbitals (LUMOs). Notably, the AC-STM measurements reveal a double occupation of the same LUMO upon charging CuPc a second time. We show by DFT calculations complemented by a grouptheoretical analysis how the molecular electron-phonon coupling in gas phase is insufficient to explain this behavior. To address this, we propose a coupling between vibronic modes within the NaCl substrate and charges residing on the molecule. This proposition highlights the significant influence of the NaCl substrate on the electronic configuration of charged molecules, extending beyond charge stabilization. [1] Fatayer S. et al., Nature Nanotechnology 13, 376-380(2018) [2] Patera L.L. et al., Nature 566, 245-248(2019)

O 15.5 Mon 16:00 MA 043

Integer Charge Transfer at an Organic–2D Material Interface: PTCDA on h-BN/Ni(111) — •Maximilian Schaal[1], Anu Baby[2,3], Marco Gruenewald[1], Felix Otto[1], Roman Forker[1], Guido Fratesi[4], and Torsten Fritz[1] — [1]Institute of Solid State Physics, Friedrich Schiller University Jena, Helmholtzweg 5, 07743 Jena, Germany — [2]Department of Materials Science, University of Milano-Bicocca, Via R. Cozzi 55, 20125 Milano, Italy — [3]STMicroelectronics, Via Tolomeo 1, 20010 Cornaredo, Italy — [4]ETSF and Dipartimento di Fisica "Aldo Pontremoli", Università degli Studi di Milano, Via Celoria, 16, 20133 Milano, Italy

Weakly interacting systems such as organic molecules on monolayers of hexagonal boron nitride (h-BN) offer the possibility of integer charge transfer leading to the formation of organic ions. Such systems exhibit unique optical and electronic properties which differ from their neutral counterparts. In this study, we used a joint experimental and theoretical approach to investigate the charge transfer of perylene-3,4,9,10-tetracarboxylic dianhydride (PTCDA) molecules on h-BN/Ni(111) by using *in-situ* differential reflectance spectroscopy (DRS), scanning tunneling spectroscopy (STS) and photoelectron orbital tomography (POT) measurements in combination with density functional theory (DFT) calculations. Our results show that the PTCDA monolayer consists of highly-ordered organic radical anions and neutral molecules. In addition, the occurrence of the integer charge transfer is discussed based on the energy-level alignment.

O 15.6 Mon 16:15 MA 043

Dioxygen ligation and activation at a biomimetic 2D metal organic framework — •Stefania Baronio[1], Michela De Col[1], Davide Bidoggia[1], Danilo Comini[2], Mattia Scardamaglia[3], and Erik Vesselli[1,4] — [1]Physics Department, University of Trieste, Trieste, Italy — [2]II Physics Department, University of Cologne, Cologne, Germany — [3]MAX IV Laboratory, University of Lund, Lund, Sweden — [4]CNR-IOM, Istituto Officina dei Materiali, Trieste, Italy

The investigation of 2D metal organic frameworks at surfaces is a hot topic in surface science due to their potential applications also as single atom catalysts [1]. Our work focuses on the in situ spectroscopic characterization of a bimetallic MnTPyP-Co framework self-assembled on Gr/Ir(111) and structurally similar to previously investigated Fe- and Co-based systems active towards the oxygen evolution and reduction reactions [2,3]. By means of a combined approach exploiting Sum Frequency Generation Spectroscopy and Ambient Pressure XPS, we show how the tetra-coordination of Co ad-atoms by adjacent MnTPyPs affects the overall electronic and vibrational structure of the layer, including the oxidation state of the Mn centers. The bimetallic system is extremely reactive towards O_2: while dioxygen weakly ligates at room temperature and under UHV conditions, at close-to-ambient pressure the molecule undergoes activation and dissociation at the Co sites, and CO oxidation becomes then possible.

[1] W. Auwärter et al., Nat. Chem. 7, 105-120(2015) [2] B. Wurster et al., J. Am. Chem. Soc. 138, 3623-3626(2016) [3] F. Armillotta et al., ACS Catal. 12, 7950-7959(2022)

O 15.7 Mon 16:30 MA 043

Electronic structure of acenes on an MoS2 monolayer — •Jan-Phillip Topmöller and Michael Rohlfing — Institute of Solid State Theory, University of Münster, Germany

TMDCs show great potential in terms of their applicability in optical electronics. Due to their large surface, molecules can easily bind to the TMDC and affect its electronic properties. Here we investigate acenes as a prototypical class of molecule.

We use DFT (GGA) and DFT-D3 to determine the most stable adsorption position of the smallest acenes like benzene, naphthalene and anthracene on MoS2. Subsequently, we use many body perturbation theory to calculate the electronic structure of the adsorbate system and its individual components (MoS2 and the molecule) in order to investigate the effect of the molecule on the MoS2 electronic and optical spectrum.

O 15.8 Mon 16:45 MA 043

Energy level alignment at organic-transition metal dichalcogenide interfaces — •Hibiki Orio[1], Maximilian Ünzelmann[1], Kiana Baumgärtner[1], Christian Metzger[1], Markus Scholz[2], Kai Rossnagel[2,3], and Friedrich Reinert[1] — [1]Universität Würzburg, Experimentelle Physik VII and Würzburg-Dresden Cluster of Excellence, Würzburg, Germany — [2]Deutsches Elektronen Synchrotron DESY, Hamburg, Germany — [3]IEAP and KiNSIS, CAU Kiel, Kiel, Germany

The hybrid interface between organic molecules and transition metal dichalcogenides (TMDCs) is characterized by weakly binding van der Waals interactions. In such systems, the interfacial energy level alignment is not in equilibrium and can be modified by external perturbations [1]. Here, we investigate the electronic structure of copper phthalocyanine (CuPc) films grown on different layered materials (graphite, $TiSe_2$, and WSe_2). Using ultraviolet and x-ray photoelectron spectroscopy, we study in particular the influence of x-ray radiation on the electronic states. We find that the HOMO level is stable in the case of CuPc/graphite, but shifts by 140 meV and 70 meV for $TiSe_2$ and WSe_2 substrates, respectively. Changes in film morphology and beam damage are subtle and insufficient to explain these different energy shifts.

[1] D. A. Racke, and O. L. A. Monti, Surf. Sci. **630**, 136 (2014).

O 15.9 Mon 17:00 MA 043

Momentum-sensitive characterization of hybrid states in metal-organic coordination networks — •Simone Mearini[1], Daniel Baranowski[1], Dominik Brandstetter[2], Andreas Windischbacher[2], Iulia Cojocariu[3], Luca Schio[4], Luca Floreano[4], Pierluigi Gargiani[5], Manuel Valvidares[5], Peter Puschnig[2], Vitaliy Feyer[1,6], and Claus M. Schneider[1,6] — [1]Forschungszentrum Jülich, Germany — [2]University of Graz, Austria — [3]University of Trieste, Italy — [4]CNR-IOM, Trieste, Italy — [5]ALBA Synchrotron, Barcelona, Spain — [6]Universität Duisburg-Essen, Germany

The electronic and magnetic properties of metal-organic coordination networks (MOCN) are to a great extent defined by the nature of the metal core and the coordinating organic ligand. To probe the new electronic states emerging due to the metal-organic bond formation, we fabricate MOCNs via co-deposition of transition metals (TMs) and benzene derivatives onto metallic substrates and apply a multi-technique approach supported by theoretical calculations to comprehensively study the local electronic structure of MOCNs. Using ARPES, we observed the appearance of energy-dispersive electronic states, attributed to the interaction between the 3d states of the TM and the ligand π molecular orbitals. This interaction stabilizes specific electronic and spin configurations of the TM ion, as determined by XPS and XMCD experiments. Finally, experimental and theoretical evidences fully support the crucial role held by the choice of the TM in the control of MOCNs properties.

O 15.10 Mon 17:15 MA 043

Imaging single protonated molecules — •Jimin Ham[1], Xu Wu[1], Klaus Kern[1,2], and Kelvin Anggara[1] — [1]MPI for Solid State Research, Stuttgart — [2]Institute de Physique, École Polytechnique Fédérale de Lausanne

Protonation of molecules is one of the most fundamental events in chemistry. Direct study of protonated molecules, however, has remained challenging due to their transient existence, short lifetimes, low abundance, and high reactivities. These characteristics cause studies of protonated molecules to be intractable for ensemble-averaged analytical methods. Here we circumvented these challenges by direct imaging of protonated molecules at single molecule level. We accomplish this by soft landing protonated molecules on surfaces using the Electrospray Ion Beam Deposition (ESIBD) technique, followed by direct imaging using Scanning Tunneling Microscopy (STM). By imaging single protonated molecules, we are able to observe directly the effect of protonation on molecular motions and electronic structures, as well as the structure of proton-catalyzed reaction intermediates. These findings open new avenues to explore proton-mediated chemical effects at the ultimate single molecule level.

O 15.11 Mon 17:30 MA 043

Electronic properties of N-heterocyclic carbenes and olefins on Si(111)$\sqrt{3}\times\sqrt{3}$ R30°-B — •Eva Glomski[1], Milan Kubicki[1], Mowpriya Das[2], Ankita Das[2], Robert Zielinski[1], René Schöder[1], Susi Lindner Franz[1], Mike Thomas Nehring[1], Mario Dähne[1], Frank Glorius[2], and Martin Franz[1] — [1]Technische Universität Berlin, Institut für Festkörperphysik, Berlin, Germany — [2]Westfälische Wilhelms-Universität Münster, Organisch-Chemisches Institut, Münster, Germany

Silicon is the most widely used semiconductor material in electronic devices and a modification or functionalization by organic overlayers is a promising approach to enhance their functionality. Recently, N-heterocyclic carbenes (NHCs) and olefins (NHOs) were found to be promising candidates for a modification of silicon surfaces [1,2]. In this work, the adsorption of NHCs and NHOs on the Si(111)($\sqrt{3}\times\sqrt{3}$)R30°-B surface is investigated using synchrotron based high-resolution photoelectron spectroscopy allowing an in-depth study of the binding behavior and the electronic properties of the formed interfaces. Furthermore, the evolution of the band bending is investigated on the basis of Si 2p spectra revealing a different behavior for the studied NHCs and NHOs.

[1] M. Franz et al., Nat. Chem. 13, 828-835 (2021).

[2] M. Das et al., Angew. Chem. Int. Ed. 2023, e202314663.

O 15.12 Mon 17:45 MA 043

Simulating Smart Surfaces for Lithium Ion Batteries — •Fabian Dietrich — Departamento de Ciencias Físicas, Universidad de La Frontera, Temuco, Chile

Self-assembled monolayers (SAM) are a common tool for the functionalization of surfaces. Such surfaces can be used form controlable solid electrolyte interfaces (SEI) on cathode materials for lithium ion batteries. Using photo-switchable organic molecules for the formation of the SAM yields a controlabilty of the surface properties - a so-called smart surface. These smart surfaces are supposed to be used to avoid undesired charge (and with that energy) loss in a fully charged lithium ion battery.

For being used as smart surface, the organic molecules have to fulfill the following requirements: 1.) A photo-switchable moiety with both photoisomers separated by at least 5000 cm^{-1}; 2.) Having polar functional groups for the interaction with the metal oxide surface; 3.) Both photo-isomers can build a stable SAM and 4.) The structural rearrangement upon photo-switching is large enough to influence the Li ion insertion and small enough to keep the SAM stable. For this challenge, we applied density functional theory (DFT) to evaluate different molecules with respect to their aggregation behavior on the previously studied vanadium pentoxide surface and subsequently different diffusion pathways through a SAM of the selected molecules.

O 16: Surface Dynamics & Electron-Driven Processes

Time: Monday 15:00–17:30 Location: MA 141

O 16.1 Mon 15:00 MA 141

Role of hydration and electrostatics in salt precipitation — •Adyant Agrawal[1], Simon Gravelle[2], Christian Holm[1], and Alexander Schlaich[3] — [1]Institute for Computational Physics, University of Stuttgart — [2]Univ. Grenoble Alpes, CNRS, LIPhy, 38000 Grenoble, France — [3]SC SimTech, University of Stuttgart

The precipitation of salt leading to the formation of a salt crust is a crucial process in the environment, yet it is still not fully resolved at the molecular scale. We perform an extensive molecular dynamics simulation study of supersaturated solutions of simple salts in contact with their crystal surface to understand the precipitation mechanism. For NaCl, we found that the sodium ions have a larger preference to be adsorbed at a crystal surface than the chloride ions, which results in a buildup of positive charge on the crystal. An excess of sodium ions then drives the adsorption of chloride ions at the surface. This leads to a depletion in the charge distributions above the surface as the crystal grows. This mechanism is different from other common salts we tested. We also obtain the dependence of the free energy for adsorbed ions on the defect topology to capture the thermodynamics of crystal surface formation. We identify a complex competition between hydration and electrostatic interactions at the interface that controls the precipitation process and the interfacial topology. The grown surface can be anomalously patterned since some sites can be more favourable for water molecules than ions. We discuss these competing effects in the context of different common salts.

O 16.2 Mon 15:15 MA 141

Unraveling the Intricacies of Surface Salt Formation on Mg(0001): Implications for Chloride-Ion Batteries — •Kanchan Sarkar[1] and Axel Gross[1,2] — [1]Institute of Theoretical Chemistry, Ulm University, 89069 Ulm, Germany — [2]Helmholtz Institute Ulm (HIU), Electrochemical Energy Storage, 89081 Ulm, Germany

We present a density functional theory study of the initial steps of chlorine deposition on the Mg(0001) surface. Such processes occur in chloride-ion batteries in which lithium and magnesium are used as anode materials. In addition, it is also of fundamental interest, as halide adsorption on metal electrodes is an important process in interfacial electrochemistry. We discuss the adsorption properties and determine the stable adsorption structures, both with respect to the free chlorine molecule but also as a function of the electrode potential. We find indications of the immediate formation of the $MgCl_2$ surface salt structure upon exposure of Cl to a Mg surface. These findings are discussed with respect to the conversion of the Mg anode to a $MgCl_2$ configuration which provides the thermodynamical driving force for the discharge of a Cl-ion battery.

O 16.3 Mon 15:30 MA 141

Copper Surface Electro-Oxidation via Large-Scale Atomistic Simulations — •Felix Riccius, Nicolas Bergmann, Simon Wengert, Nicolas G. Hörmann, Hendrik H. Heenen, and Karsten Reuter — Fritz-Haber-Institut der MPG, Berlin

Copper (Cu) is a versatile electrocatalyst material. However, the catalytically active metal phase is susceptible to oxidation either during electrode preparation or within the reaction environment. The degree of oxidation and the concomitant structural and morphological transformations are presently still largely unclear, let alone how these changes affect the electrocatalytic performance. With the formed surface structures certainly far from ideal, determining atomistic structural models requires an efficient sampling in vast configurational spaces. We address this challenge by capturing predictive-quality density-functional theory calculations using a MACE machine learning interatomic potential. Enabled by the resulting fast energetics, parallel tempering is employed to sample a large surface phase space of Cu surface structures at increasing degree of oxidation. Combining the resulting data within *ab initio* thermodynamics we arrive at surface phase diagrams that point to amorphous Cu(I)O-like sites as a key to understand the reactivity and stability changes.

O 16.4 Mon 15:45 MA 141

Exploring dynamic catalyst restructuring with adaptive kinetic Monte Carlo: Oxidation of the Pd(100) surface — •Patricia Poths, King Chun Lai, Christoph Scheurer, Sebastian Matera, and Karsten Reuter — Fritz-Haber-Institut der MPG, Berlin

Modern operando catalyst characterization approaches have revealed that even single crystal model catalysts are not static, but adapt their surface morphology and composition in response to applied reactions conditions. One such adaption is the possible formation of surface oxides, for which the mechanism and kinetics must be regarded as largely unknown. In this talk, we take the first steps towards explicit dynamic modeling of this process employing a newly developed adaptive kinetic Monte Carlo (AKMC) approach. This technique automatically determines the complex reaction mechanism on-the-fly instead of relying on human intuition. Employing machine-learned potentials as the basis for AKMC, we investigate for the first time the explicit kinetics of surface oxide formation on the highly debated Pd(100) surface. We discuss key aspects of surface restructuring during the initial phase of oxide formation.

O 16.5 Mon 16:00 MA 141

Realistic Representations of IrO2 Catalyst Surfaces through Extensive Sampling — •Hao Wan, Hendrik H. Heenen, Christoph Scheurer, and Karsten Reuter — Fritz-Haber-Institut der MPG, Berlin

Iridium oxide catalyzes the oxygen evolution reaction with unparalleled activity and stability, even under harsh acidic conditions. However, this performance is sensitively correlated to strong structural, compositional and morphological changes of the working catalyst. At the atomic level little is presently known about the true active state, if only that it is unlikely ideal rutile IrO2.

This situation spans a vast configurational space, the extensive sampling of which e.g. by means of parallel tempering would be intractable with direct predictive-quality first-principles calculations. Training of a machine-learning interatomic potential (MLIP) as an efficient surrogate is in turn challenged by an unprecedented required diversity of training structures if not even the bulk structure and composition can be assumed known. To this end, we create a comprehensive training set by first assembling prototype bulk structures for various IrO* stoichiometries from existing databases. In an active learning loop, this set is then augmented through extensive sampling of diverse surface structures created from the prototypes. The thus trained MLIP successfully reproduces the known stability reversal of the rutile (110) and (111) facets with increasing potential, while its computational efficiency allows to rapidly probe the activity and stability of the plethora of sampled surface sites using established descriptors.

O 16.6 Mon 16:15 MA 141

Nonadiabatic quantum dynamics of molecules scattering from metal surfaces — •Riley Preston[1], Yaling Ke[1], Samuel Rudge[1], Reinhard Maurer[2], and Michael Thoss[1] — [1]Institute of Physics, University of Freiburg, Germany — [2]Department of Physics, University of Warwick, UK

A precise theoretical understanding of reactivity at metal surfaces has the possibility of providing valuable theoretical insights into the dynamical processes which emerge due to the coupling of a molecule to a surface, as well as the prospect of profound consequences to a range of applications. However, the modelling of molecular scattering off metal surfaces is a theoretical challenge, since a single scattering process will traverse regimes of both strong and weak coupling, while there is no guarantee of a time-scale separation between electronic and nuclear degrees of freedom, rendering perturbative approaches invalid [1].

We simulate the scattering of molecules off metal surfaces using the hierarchical equations of motion approach in the framework of a matrix product state formulation, which aids the computational efficiency

[2]. The approach constitutes a numerically exact, quantum approach which includes all nonadiabatic and quantum nuclear effects. The method is applied to a system described by a Newns-Anderson Hamiltonian, from which we derive valuable insights about the behaviour of important observables such as the adsorption probability to the surface under different conditions.

[1] A. M. Wodtke, Chem. Soc. Rev. 45, 3641-3657 (2016).

[2] Y. Ke, R. Borrelli, M. Thoss, J. Chem. Phys. 156, 194102 (2022).

O 16.7 Mon 16:30 MA 141

Electronic Friction of Interacting Systems Near Metal Surfaces — •Martin Mäck, Samuel Rudge, and Michael Thoss — Institute of Physics, University of Freiburg, Germany

Mixed quantum-classical methods are an efficient way of modeling the dynamics of molecules near metal surfaces, such as in molecular junctions or scattering problems. Since a full quantum mechanical description of the vibrational degrees of freedom is numerically expensive, semiclassical methods such as Langevin dynamics based on electronic friction can greatly reduce the computational cost.

Until now, however, a description based on electronic friction was either limited to noninteracting systems or position independent metal-molecule couplings. In this contribution, we further develop the hierarchical equations of motion (HEOM) approach to electronic friction [1] to systems with a position dependent molecule-metal coupling. We derive new expressions for the Markovian friction tensor. Moreover, we apply our approach to a system with strong intrasystem interactions. Here, we demonstrate the usefulness of our extended approach, since the HEOM remains numerical exact [2], even for interacting systems. Our extended approach could be useful for the semiclassical description of a wide variety of problems, such as desorption dynamics and quantum shuttles.

[1] S. L. Rudge, Y. Ke and M. Thoss, Physical Review B **107**, 115416 (2023)

[2] C. Schinabeck, A. Erpenbeck, R. Härtle and M. Thoss,
Physical Review B **94**, 201407 (2016)

O 16.8 Mon 16:45 MA 141

How a long-range attraction influences desorption kinetics — •Florian Schneider[1], Lukas Höltkemeier[1], Andrea Floris[2], Lev Kantorovich[3], Ralf Bechstein[1], and Angelika Kühnle[1] — [1]Physical Chemistry I, Bielefeld University, 33615 Bielefeld, Germany — [2]School of Chemistry, University of Lincoln, Brayford Pool, Lincoln LN6 7TS, UK — [3]Department of Physics, Kings's College London, London WC2R 2LS, UK

Desorption of molecules from surfaces is omnipresent both in nature and technology. Despite its omnipresence and conceptual simplicity, fundamental details can be surprisingly complex and are often poorly understood. Here, we study the desorption of a submonolayer film of 3-nitrophenol from the natural cleavage plane of calcite kept in ultrahigh vacuum. Interestingly, two distinctly different desorption regimes are observed during isothermal desorption monitored by dynamic atomic force microscopy. Initially, at high coverages, the coverage decreases almost linearly in time, indicating a constant desorption rate. Beyond this linear regime, at low coverages, a drastic increase in desorption rate is observed until the surface is completely empty. The transition between these two regimes is associated with a critical island width. The increase in desorption rate when falling below a critical island width indicates that a long-range attractive interaction between the molecules is present. The herein observed phenomenon of two different desorption regimes is expected to be of general significance when interactions beyond next-neighbor attraction are present.

O 16.9 Mon 17:00 MA 141

Supercooled 1D-metal nanocrystallites prepared by orbital-selective photodoping — •Hannes Böckmann[1,2], Jan Gerrit Horstmann[1,2], Felix Kurtz[1,2], Stefan Wippermann[3], and Claus Ropers[1,2] — [1]Max Planck Institute for Multidisciplinary Sciences, Göttingen, Germany — [2]4th Physical Institute, University of Göttingen, Göttingen, Germany. — [3]Faculty of Physics, Philipps University of Marburg, Marburg, Germany.

Photochemical reactions in molecules are brought about by targeted transitions into reactive electronic states. Much like molecular levels, the electronic bands of a material are formed due to the hybridization of atomic orbitals at different energies. In this sense, the most fundamental solid state state analogy is described by the optical quench of a Peierls insulator, in which the periodic dimerization of a one-dimensional atomic chain is lifted by resonant transitions between electronic states at the accompanying band gap. However, in real materials, their light-induced occupation is typically mediated by delocalized higher energy bands, effectively rendering structural transformations independent of the light frequency and polarization. Here, we use orbital-selective photodoping to drive the quench of a prototypic quasi one-dimensional Peierls insulator, formed by atomic indium wires on the (111) face of silicon. Polarized excitation exploits the anisotropic interactions along specific crystallographic directions to directly address electronic states at the cdw gap. The targeted excitation minimizes excess energy, leading to an unprecented control over the nanoscale phase texture and metastable lifetime.

O 16.10 Mon 17:15 MA 141

A continuous-wave/pulsed X-band electron spin resonance spectrometer operating in ultra-high vacuum for the study of low dimensional spin ensembles — Franklin Cho[1,2], Juyoung Park[1,2], Soyoung Oh[1,2], Jisoo Yu[1,2], Yejin Jeong[1,2], Luciano Colazzo[1,2], Lukas Spree[1,2], Caroline Hommel[1,2], Arzhang Ardavan[3], Giovanni Boero[4], and •Fabio Donati[1,2] — [1]IBS Center for Quantum Nanoscience, South Korea — [2]Department of Physics, Ewha Womans University, South Korea — [3]Clarendon Laboratory, University of Oxford, UK — [4]Microsystems Laboratory, EPFL, Switzerland

Surface-adsorbed molecular spin qubits have been attracting growing interest as a new platform for coherently controlled phenomena at the atomic scale. The localization at the surface of the molecular spins imposes to employ specifically designed setups to achieve sufficient spin sensitivity and operate in ultra-high vacuum. In this work, we report the development of a UHV continuous-wave and pulsed X-band electron spin resonance (ESR) spectrometer operating down to 10 K. The use of a half-wavelength microstrip line resonator made of epitaxially grown copper films on single crystal Al_2O_3 substrates allows achieving a sensitivity of 6.5×10^{10} spins G^{-1} $Hz^{-1/2}$ on a surface of about 1 mm^2. Surface characterizations using atomic force microscopy, low-energy electron diffraction, and scanning tunneling microscopy show that the Cu surface is atomically clean, flat, and single crystalline. Advanced pulsed ESR experimental capabilities including dynamical decoupling and electron-nuclear double resonance are demonstrated.

O 17: Semiconductor Substrates II: Structure, Epitaxy, Growth

Time: Monday 15:00–17:00 Location: MA 144

O 17.1 Mon 15:00 MA 144

Bimodal Growth of Fe islands on graphene — •Yisheng Gu — Shanghai Jiao Tong University, Shanghai, China

Nucleation of different materials on graphene may improve our cognizing of further application of graphene related systems, among which the combinations of graphene and magnetic materials are promising in spin related technics. We have prepared Fe islands on epitaxial graphene on SiC by molecular beam epitaxy, and then directly examine the topography by scanning tunneling microscope. At room temperature, Fe forms cluster. However, by annealing at relatively high temperature, Fe flat islands and Fe polyhedrons arise. The electronic states with certain patterns that differs from normal lattice are detected on top of the flat islands. This difference of Fe topography between room temperature growth and relatively high temperature annealing may be generalized to deposition of other metals on graphene.

O 17.2 Mon 15:15 MA 144

Local GaAs growth on patterned Si(001) surfaces by Laser-assisted MOVPE — •Christian Bruckmann, Jürgen Bläsing, Armin Dadgar, and André Strittmatter — Institut für Physik, Otto-von-Guericke-Universität Magdeburg, PF4120 Magdeburg, Germany

Monolithic integration of group III-V compound semiconductors on a silicon-based platform is the ultimate solution for combining optoelectronics based on compound semiconductors with Si-based integrated circuit technology. Heteroepitaxy of group III-V semiconductors on Si wafers is a common approach as it facilitates large-scale production. However, for the realization of complex networks by implementing different high performance devices on the same chip a local growth approach of III-V-compound semiconductors can be a viable path to achieve cost-effective monolithic integration. The newly developed Laser-assisted Metal Organic Vapor Phase Epitaxy features local heating via high-power laser radiation enabling controllable local epitaxial growth[1]. In order to improve the crystalline quality of GaAs islands, the local growth is now performed on patterned Si(001)-wafers. In addition n-/p-type doping of GaAs-based structures is also presented.

[1] M. Trippel et al., "Laser-assisted local metal-organic vapor phase epitaxy", Rev. Sci. Instrum. 93, 113904 (2022)

O 17.3 Mon 15:30 MA 144

Growth of vanadium on Si(111) — •Dang Liu — School of Physics and Astronomy, Shanghai Jiao Tong University, Shanghai, China

Vanadium is a type-II superconductor with a Tc=5.3 K. Due to its high melting temperature, it is difficult to grow Vanadium material by thermal evaporation. We use molecular beam epitaxy method to deposit Vanadium on silicon substrate, and then characterized by low temperature scanning tunnelling microscope. Vanadium islands with many facets are successfully grown on Si(111) with ultra-high substrate temperature. The lateral dimensions of Vanadium islands are from dozens to several hundred nanometers, and their heights are around several tens of nanometers. Atomic images of different facets are obtained by constant current mode of STM. We also get scanning tunneling spectroscopy on the surface of vanadium island, and a small gap shows around zero bias, which is a signal of superconductivity.

O 17.4 Mon 15:45 MA 144

In-situ analysis of phase transitions in ultrathin nickel silicides by time-of-flight medium energy ion scattering — •Carolin Frank[1], Kevin Vomschee[1], Radek Holeňák[2], Eleni Ntemou[2], and Daniel Primetzhofer[2] — [1]Faculty of Physics and CENIDE, University of Duisburg-Essen, Duisburg, Germany — [2]Department of Physics and Astronomy, Uppsala University, Uppsala, Sweden

We present first results obtained during the commissioning phase of recent upgrades to the Time-of-Flight Medium Energy Ion Scattering (ToF-MEIS) set-up at Uppsala University: A new UHV sample preparation chamber enables growth of thin films with a high precision in thickness by deposition from electron beam evaporators. We focus on calibration of the growth rate, in-situ deposition of thin nickel films on silicon and subsequent ToF-MEIS analysis of these films regarding phase transitions. Prior to deposition of nickel, the silicon substrates were treated ex-situ with hydrofluoric acid and annealed. To study the phase transition from the nickel layer deposited on silicon to epitaxial nickel silicide step-by-step, the samples were subsequently annealed up to 1500 K by an electron impact heater. During the stepwise annealing process, the transition to a well-ordered cubic nickel disilicide phase was found at 648 K. 2D scattering maps enable the analysis of intensity distributions of backscattered ions for selected energies. Since each of these energies corresponds to different chemical elements and - for a specific element signal - to specific depths within the sample, the epitaxy of the nickel silicide films due to blocking effects is verified.

O 17.5 Mon 16:00 MA 144

XPS study on composition and band structure of aluminum alloyed β-gallium oxide bulk crystals and thin films — •Lukas Schewe[1], Jana Rehm[2], Ming Chao Kao[3], Vedran Vonk[3], Zbigniew Galazka[2], Saud Bin Anooz[2], Andreas Popp[2], and Jan Ingo Flege[1] — [1]Fachgebiet Angewandte Physik und Halbleiterspektroskopie, BTU Cottbus-Senftenberg — [2]Leibnitz-Institut für Kristallzüchtung, Berlin — [3]CXNS-Center for X-ray and Nano Science, DESY Hamburg

Beta-phase gallium oxide is a wide gap semiconductor with promising applications in high-power devices. To further increase the high-power capabilities of a material with dielectric constant ϵ, as suggested by the Baliga figure of merit $BFOM = \epsilon\mu E_b^3$, it is desirable to increase the electric breakdown field E_b, e.g., by alloying with aluminum.

The present work discusses structural and electronic properties of $\beta - (Al_xGa_{1-x})_2O_3$ thin films grown by metal organic vapour-phase epitaxy and bulk crystals grown by the czochralski method. The predicted Al content ranges up to 30%. The Al concentration has been measured by X-ray photoelectron spectroscopy (XPS) and will be compared to the Al content calculated from the lattice parameter measured by X-ray diffraction for thin films and inductively coupled plasma-optical emission spectrometry for bulk samples. Additionally the thin films have been investigated for homogeneity through XPS depth profiling obtained by sequential Ar ion sputtering. Furthermore the band gap has been determined by electron loss spectra from XPS depending on Al content and compared to optical absorbance measurements.

O 17.6 Mon 16:15 MA 144

Atomic structure of As-modified Si(100) surfaces prepared in MOCVD ambience utilizing background arsenic — Chris Yannick Bohlemann[1], Agnieszka Paszuk[1], Manali Nandy[1], Aaron Flötotto[2,3], Max Grossmann[2,3], Oleksandr Romanyuk[4], •Kai Daniel Hanke[1], Peter Kleinschmidt[1], Erich Runge[2,3], and Thomas Hannappel[1] — [1]TU Ilmenau, Institute of Physics, Fundamentals of Energy Materials — [2]TU Ilmenau, Institute of Physics,Theoretical Physics I — [3]TU Ilmenau, Centre of Micro- and Nanotechnologies — [4]Institute of Physics, Academy of Sciences of the Czech Republic, 182 00 Prague 8

A low-defect III-V nucleation layer and a well-defined atomically abrupt interface between the Si(100) substrate and the III-V nucleation layer are essential prerequisites for subsequent low-defect III-V layer growth. Preparation of a well-ordered Si(100) surface in industrially-relevant MOCVD ambience with arsenic benefits in a significant temperature reduction during the deoxidation step. In this study, we investigate the atomic structure of Si(100) surfaces prepared in As-rich MOCVD reactor, employing background arsenic as the arsenic source. The preparation of the samples in the MOCVD reactor was monitored in situ by surface sensitive optical spectroscopy and the surfaces were characterized in UHV by FTIR and STM. The measurements are supported by complementary DFT calculations. We confirm presence of hydrogen on the surface and mixed As-Si-H dimers, which was previously unrecognized.

O 17.7 Mon 16:30 MA 144

Towards an ab initio kinetic Monte Carlo model for the growth of β-Ga 2 O 3 (100) — •Qaem Hassanzada[1], Konstantin Lion[1,2], Claudia Draxl[2], and Matthias Scheffler[1] — [1]The NOMAD Laboratory at the FHI of the Max-Planck-Gesellschaft and IRIS-Adlershof of the Humboldt-Universität zu Berlin — [2]Physics Department and IRIS Adlershof, Humboldt-Universität zu Berlin, Berlin, Germany

Ga2O3 is gaining attention in diverse applications [1]. Insights from molecular beam epitaxy (MBE) indicate that varying Ga-to-O ratios influence its growth modes [2], and volatile suboxide (Ga2O) desorption limits growth rate under Ga-rich conditions [3]. However, the atomic-level mechanisms underlying these observations remain elusive. This project focuses on studying β-Ga2O3(100) growth using density functional theory (DFT) calculations. The study uncovers distinct stable sites for single adatoms and clusters. Ga adatoms diffuse more favorably than O adatoms at both stoichiometric terminations. Both adatoms and Ga-O pairs preferentially diffuse along the b crystallographic direction. This directional mobility aligns with experimental observations of elongated islands along the b direction in MBE growth [4]. The results lay the groundwork for developing a comprehensive ab initio kinetic Monte Carlo model for Ga2O3 growth.

[1] D. Guo et al. Materials Today Physics 11, 100157 (2019) [2] P Mazzolini et al., J. Phys. D: Appl. Phys. 53, 354003 (2020). [3] P. Vogt et al., Appl. Phys. Lett. 106, 081910 (2015). [4] Z. Cheng et al, Nanotechnology 29, 395705 (2018).

O 17.8 Mon 16:45 MA 144

Exploring surface properties of hexagonal Si and Ge — •Martin Keller[1], Jürgen Furthmüller[1], Friedhelm Bechstedt[1], and Silvana Botti[1,2] — [1]Friedrich-Schiller-Universität Jena, Germany — [2]Ruhr-Universität Bochum, Germany

The determination of surface energy is essential to provide guidelines for choosing proper substrates for high-quality epitaxial growth. At the same time interface electronic states are crucial to determine the functionality of a heterostructure. We present ab initio calculations, using density functional theory in a slab geometry, that explore structural and electronic properties of the surfaces of hexagonal silicon and germanium, which are novel materials for active optoelectronic applications. We study the a, m, c and r-plane orientations, which are being considered as preferential facets for the growth of hexagonal SiGe nanowires. Our focus lies on understanding surface and interface stability and its implications on material and device design. Our calculations and the comparison with experimental findings offer practical insights for substrate selection for thin film growth and the construction of heterostructures.

O 18: Solid-Liquid Interfaces I: Structure and Spectroscopy

Time: Monday 15:00–18:00 Location: H 1012

O 18.1 Mon 15:00 H 1012

Reporting activity and stability for the oxygen evolution reaction — •Marcel Risch — Helmholtz-Zentrum Berlin, Berlin, Germany

The oxygen evolution reaction (OER) is a key enabler of sustainable chemical energy storage. It occurs on the anode of a fuel-producing device such as electrolyzers or photoelectrochemical cells and provides the protonated ions for the formation of the fuel molecule. The OER has been studied for more than 230 years. Yet, there is no consensus how to report the key performance parameters of electrocatalytic activity and stability [1,2], which thwarts, e.g., correlating these parameters to materials properties for systematic mechanistic insight [3]. The author assesses the current sore spots when reporting activity and stability and touches on the related topic of determining the electrochemical surface area [4]. Ideas are presented to improve upon the current sore spots.

[1] M. Risch, Commun Chem 6, 221 (2023) [2] M. Risch, Cur. Op. Electrochem. 38, 101247 (2023) [3] D. Antipin, M. Risch, JPhys Energy 2, 032003 (2020) [4] D.M. Morales, M. Risch, JPhys Energy 3, 034013 (2021)

O 18.2 Mon 15:15 H 1012

Interfacial water structure effects during CO2 electroreduction — •Ya-Wei Zhou[1,2], Beatriz Roldan Cuenya[2], and Christopher Seiji Kley[1,2] — [1]Helmholtz Young Investigator Group Nanoscale Operando CO2 Photo-Electrocatalysis, Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, 14109 Berlin, Germany — [2]Department of Interface Science, Fritz Haber Institute of the Max Planck Society, 14195 Berlin, Germany

We combine on-line differential electrochemcial mass spectroscopy (DEMS), in situ attenuated total reflection surface enhanced Infrared spectroscopy (ATR-SEIRAS), operando shell-isolated nanoparticle-enhanced Raman spectroscopy (SHINERS) and density functional theory (DFT) to investigate the structure and dynamic process of interfacial water on Au and Cu surfaces during CO2 electroreduction reaction (CO2RR). Direct ATR-SEIRAS evidence and D2O labelling DEMS data reveals that H2O is the proton donor of CO2RR and HER, while bicarbonate is solely involved in the Heyrovsky step. We reveal that hydrated CO32- increases the ordering of the hydration water network, with shorter H-bond leading to rapid delivery of electrons and protons at the electrode-electrolyte interface. This, in turn, suppresses the CO2RR due to promoted H* formation through the increased connectivity of the CO32- induced interfacial water network in the high current density region. We observe a carbonate anion radical (CO3.-) on both Au and Cu surfaces that we propose to originate from hydrated CO32-, acting as additional carbon source under reducing condition.

O 18.3 Mon 15:30 H 1012

Cu/Au(111) Surfaces and AuCu Intermetallics for Electrocatalytic Reduction of CO_2 in Ionic Liquid Electrolytes — •Björn Ratschmeier[1], Christian Paulsen[1], Klaus Stallberg[2], Gina Ross[1], Winfried Daum[2], Rainer Pöttgen[1], and Björn Braunschweig[1] — [1]University of Münster, Germany — [2]TU Clausthal, Germany

Room-temperature ionic liquids (RTIL) are important alternatives for reducing high overpotentials and modifying product selectivities in CO_2 reduction reactions (CO_2RR). The activity of Au(111), Cu(111), Cu-modified Au(111) electrodes, and AuCu intermetallics in contact with 1-butyl-3-methylimidazolium trifluorosulfonylimide [BMIM][NTf$_2$] electrolytes was investigated in terms of CO_2RR. *In operando* IR absorption spectroscopy (IRAS) of the bulk electrolyte reveals the formation of a 2-imidazolium carboxylic acid intermediate that can lower the overpotential for CO_2RR and does not require stabilization of the CO_2 radical anion as an alternative intermediate at the interface. On Au(111) we observe the formation of H_2 and mainly CO, while on Cu(111) the only reduction product is H_2. By systematically varying the copper content at the catalyst surfaces, we are able to tune the H_2/CO syngas ratio to a maximum of 1.8 for Cu-modified Au(111) electrodes and 3.2 for $AuCu_3$ catalysts, demonstrating a large tunability of the syngas ratio with electrode potential. The observed H_2/CO ratios approach the ideal value of 2 needed for the Fischer-Tropsch process and a ratio of 3 required for methanation.

O 18.4 Mon 15:45 H 1012

The Influence of Mesoscopic Surface Structure on the Electrocatalytic Selectivity of CO2 Reduction with UHV-Prepared Cu(111) Single Crystals — •Khanh-Ly Nguyen, Jared P. Bruce, Aram Yoon, Juan Navarro, Fabian Scholten, Felix Landwehr, Clara Rettenmaier, Markus Heyde, and Beatriz Roldan Cuenya — Fritz Haber Institute of the Max Planck Society, Faradayweg 4-6, 14195 Berlin, Germany

Electrocatalytic conversion of CO2 to higher order hydrocarbons has been proposed as a promising approach to re-utilize the greenhouse CO2. The only pure metal that is capable of reducing CO2 to C2+ hydrocarbons is copper although it suffers from low selectivity. Cu(111) single crystal surfaces prepared under ultra-high vacuum (UHV) conditions were found to display different product selectivity during CO2RR despite similar chemical and local microscopic surface features. In particular, changes in selectivity from hydrogen-dominant to hydrocarbon-dominant product distributions were observed based on the number of CO2RR electrolysis and subsequent UHV treatments. Significant mesostructural changes were observed through a micron scale microscopic analysis, including a higher density of irregular steps on the samples producing hydrocarbons. Thus, our findings highlight that step edges are key for C-C coupling in CO2RR and that not only atomistic but also mesoscale characterization of electrocatalytic materials is needed in order to comprehend complex selectivity trends.

O 18.5 Mon 16:00 H 1012

Oxygen reduction reaction on adlayer covered surfaces: A

case study on Ru(0001) in acid — •Albert K. Engstfeld[1,2], Stephan Beckord[1], Stefan Fuchs[1], and R.Jürgen Behm[1,3] — [1]Institute of Surface Chemistry and Catalysis, Ulm, DE — [2]Institute of Electrochemistry, Ulm, DE — [3]Institute of Theoretical, Chemistry, Ulm, DE

On bare Ruthenium (Ru) electrodes, the oxygen reduction reaction (ORR) primarily leads to the formation of water. Depending on the electrode structure and availability of surface (hydr)oxides, in certain potential regions also H_2O_2 can be formed. Recently we have shown that the electrode surface is never free from adsorbates.[1] In this work, we will show how such adlayers influence the ORR activity and discuss the limitations of the commonly accepted reaction pathway, which involves free surface sites. The electrodes are prepared under ultrahigh vacuum conditions and their structural integrity is determined by scanning tunnelling (STM) measurements performed before and after the electrocatalytic investigation. The electrocatalytic measurements are performed in O_2-free and O_2-saturated acid electrolytes ($HClO_4$ and H_2SO_4). We will discuss the impact of bisulfate on the reactivity, which almost completely inhibits the reactivity of the surface. Furthermore, we illustrate that adsorbed OH plays a major role in the formation of H_2O_2 on such surfaces. Additional differential electrochemical mass spectrometry (DEMS) measurements allow disentangling the ORR from the hydrogen evolution reaction at high overpotentials.
[1] A.K. Engstfeld *et al.*, Electrochimica Acta, **389** (2021) 138350

O 18.6 Mon 16:15 H 1012

Initial Stages of Alkali Metal Deposition on Au(111) from [MPPip][TFSI] — •Maren-Kathrin Heubach[1], Fabian M. Schuett[1], Areeg Abdelrahman[1], Ludwig A. Kibler[1], Toni Moser[2], Julia Kunze-Liebhäuser[2], and Timo Jacob[1,3,4] — [1]Institute of Electrochemistry, Ulm University, Ulm, Germany — [2]Institute of Physical Chemistry, University of Innsbruck, Innrain 52c, Innsbruck, 6020 Austria — [3]Helmholtz-Institute-Ulm (HIU), Ulm, Germany — [4]Karlsruhe Institute of Technology, Karlsruhe, Germany

The cycling stability of alkali metal batteries using a metal anode is often limited by dendritic growth.[1,2] Hereby, the initial stages of metal deposition and dissolution play a key role since they have a huge impact on the macroscopic morphology of the deposits.[3]

In this study, the initial stages of sodium and lithium deposition on Au(111) from the ionic liquid *N*-methyl-N-propylpiperidinium bis(trifluoromethane-sulfonyl)imide ([MPPip][TFSI]) are investigated by cyclic voltammetry and *in situ* scanning tunnelling microscopy. The deposition of both alkali metals starts at underpotentials of approximately 1 V with island formation and the following underpotential deposition includes more than two monolayers.[4]

[1] A. Jana, R. E. García, Nano Energy **2017**, *41*, 552–565. [2] B. Lee, E. Paek, D. Mitlin, S. W. Lee, Chemical Reviews **2019**, *119*, 5416–5460. [3] C. A. Berger, M. U. Ceblin, T. Jacob, ChemElectroChem **2017** *4*, 261–265. [4] M.-K. Heubach, F. M. Schuett, L. A. Kibler, A. Abdelrahman, T. Jacob, ChemElectroChem **2022**, *9*, e202200722.

O 18.7 Mon 16:30 H 1012

Self-organized structures of carboxylic acids on the Au (111) electrode surface — •Kristin Gratzfeld[1], Anna J. Kny[1], Tomasz Kosmala[2], Radosław Wasielewski[2], Marek Nowicki[2], Klaus Wandelt[1], and Moritz Sokolowski[1] — [1]Clausius-Institute of Physical and Theoretical Chemistry, University of Bonn, Germany — [2]Institute of Experimental Physics, University of Wrocław, Poland

We investigated the self-ordered structures of 3,4,9,10-perylene-tetracarboxylic acid (PTCA) and 1,4,5,8-naphthalene-tetracarboxylic acid (NTCA) on the Au(111) electrode interface in an aqueous electrolyte. Electrochemical studies of PTCA were perfomed before [1], however no structural data was reported, yet. We used cyclic voltammetry (CV) with in situ electrochemical STM (EC-STM): By tuning the electrode potential, we can observe different ordered phases of both molecules on the Au(111) electrode surface. We will discuss the geometrical differences of the ordered phases and why we observe the transitions between phases at different electrode potentials on the Au(111) surface. When shifting to more positive electrode potentials, we observe that the PTCA molecules in the ordered phases change from an upright to a tilted and finally to a flat orientation. In contrast, for NTCA we observe only ordered phases of tilted molecules.

This work was supported by the DFG through the research training group 2591, the DAAD (Deutscher Akademischer Austauschdienst)program Ostpartnerschaften and the NAWA (National Agency for Academic Exchange) program.

[1] Journal of American Society, 138, 1490 -1493, 2016.

O 18.8 Mon 16:45 H 1012

In situ electrochemical atomic force microscopy studies of a copper surface during lithium plating and dissolution — •Luca Kaufer[1], Ronja Haas[2], Thomas Göddenhennrich[1], Daniel Ebeling[1], Jürgen Janek[2], and André Schirmeisen[1] — [1]Institute of Applied Physics, Justus-Liebig-University, Gießen, Germany — [2]Institute of Physical Chemistry, Justus-Liebig-University, Gießen, Germany

The increasing concern about climate change and the transition to sustainable energy sources has led to a increased demand for high energy density batteries for energy storage. Lithium is considered a favourable anode material[1]. However, challenges like dendrite growth when used in batteries are still present[2]. This investigation focuses on the processes involved in dendrite formation and growth and upcoming side reactions in a lithium-copper system with in situ electrochemical atomic force microscopy as a method. We will cover two topics. Firstly, the technical development of an appropriate measurement setup for in situ investigation of cells in atomic force microscopy. Secondly, the presentation of initial findings on the processes of formation and growth of dendrites within a lithium-copper cell system.

[1]Yan, K.; Lu, Z.; Lee, H.-W.; Xiong, F.; Hsu, P.-C.; Li, Y.; Zhao, J.; Chu, S.; Cui, Y. Nat Energy 2016, DOI: 10.1038/nenergy.2016.10

[2] TAKEDA, Y.; YAMAMOTO, O.; IMANISHI, N. Electrochemistry 2016, DOI: 10.5796/electrochemistry.84.210

O 18.9 Mon 17:00 H 1012

Size Effects and Active State Formation in Cobalt Oxide Nanoparticles during Oxygen Evolution Reaction — •Arno Bergmann[1], Felix T. Haase[1], Travis E. Jones[2], Janis Timoshenko[1], and Beatriz Roldan Cuenya[1] — [1]Department of Interface Science, Fritz-Haber-Institut, Berlin, Deutschland — [2]Department of Inorganic Chemistry, Fritz-Haber-Institut, Berlin, Deutschland

Water electrolysis for a CO_2-neutral H_2 production remains challenging due to the large overpotential required for the anodic oxygen evolution reaction (OER). Cobalt oxides exhibit high catalytic activity for OER but the underlying principles of the oxygen-evolving surface is unkown. Thus, we investigated size-selected $CoO_x(OH)_y$ nanoparticle (NP) catalysts down to 1 nm using operando X-ray absorption spectroscopy (XAS) combined with DFT calculations.[1] The mass-based activity showed an increasing OER current with decreasing NP size which correlates with a larger accessible surface area. XAS revealed reversible changes in charge density at the Co-O ligand system during OER, being more pronounced with decreasing NP size. Size-dependent changes in the Co-O distance during OER were correlated to charge-transfer between Co and O ligand determined by DFT suggesting electrophilic O sites and allow to extract quantitative information on the near-surface atomic and electronic structure of the oxygen evolving $CoO_x(OH)_y$ NP near-surface. [1] F.T. Haase et al, Nat Energy 7, 765 (2022).

O 18.10 Mon 17:15 H 1012

Tuning the HER activity by using ion confinement in layered MoS2 — •Marco Schönig[1], Simon Fleischmann[2], and Marc Koper[1] — [1]Leiden University, Leiden, The Netherlands — [2]HIU, Ulm, Germany

The optimization of energy storage and conversion materials caused a decrease of the pore size in these materials to the nanometer scale. On this length scale the ions can not simply enter the pores but are forced to (partially) shed their solvation shell [1]. This so called "ion confinement" was shown to be promising way to increase the storage capacity of supercapacitors [2]. However, this effect is scarcely exploited in electrocatalysis, although ions close to the surface play an important role in several electrocatalytic reactions, including the HER [3]. In this contribution, we study the effect of the confinement of different ions on the activity of the HER at MoS2. This is achieved by tuning the ion confinement via the interlayer distance of the layered MoS2, as well as the ion diameter.

[1]Chmiola, J.; Yushin, G.; Gogotsi, Y.; Portet, C.; Simon, P.; Taberna, P.-L. Anomalous increase in carbon capacitance at pore sizes less than 1 nanometer Science, 2006, 313, 1760-1763. [2]Fleischmann, S.; Spencer, M. A.; Augustyn, V. Electrochemical Reactivity under Confinement Enabled by Molecularly Pillared 2D and Layered Materials Chem. Mater., 2020, 32, 3325-3334. [3]Monteiro, M. C. O.; Goyal, A.; Moerland, P.; Koper, M. T. M. Understanding Cation Trends for Hydrogen Evolution on Platinum and Gold Electrodes in Alkaline Me-

dia ACS Catal., 2021, 11, 14328-14335.

O 18.11 Mon 17:30 H 1012

Impedance Spectroscopy with Microscopic Reference Electrodes as a Technique to Study Electrode Kinetics in Dye-Sensitized Solar Cells — •Daniel Holzhacker and Derck Schlettwein — Institut für Angewandte Physik, Justus-Liebig-Universität Gießen

A detailed impedance analysis of dye-sensitized solar cells (DSSC) is essential to precisely optimize performance-limiting steps in these cells. Two-electrode impedance measurements as typically used, however, often do not allow a separate analysis of charge transfer reactions at different electrodes that occur with similar time constants. The use of a microscopic quasi-reference electrode (μRE) offers the possibility to separately record impedance spectra of the photoanode (WE) and the counter electrode (CE) of a DSSC under operating conditions in one experiment. In three-electrode experiments, the potential was modulated between the μRE and WE. The respective impedance (Z_{WE}) and, simultaneously, the impedance between μRE and CE (Z_{CE}) were recorded and the overall cell impedance Z calculated. The latter was identical to Z determined in a two-electrode measurement, proving the validity of the experimental approach. Thus, one experiment yielded spectra for the full cell and both respective half-cells, which allowed to simultaneously study the interactions of novel redox mediators with photoanodes and counter electrode materials in detail.

O 18.12 Mon 17:45 H 1012

Arrhenius follows Frumkin to describe Atomic Diffusion involved Peaks in Cyclic Voltammograms: the Reversible Place-Exchange on Pt(111) — Jon Bjarke Valbaek Mygind[1], Francesc Valls Mascaró[2], and •Marcel J. Rost[3] — [1]Department of Physics, Technical University of Denmark, 2800 Kongens Lyngby, Denmark — [2]Leiden Institute of Chemistry, Leiden University, Leiden, The Netherlands — [3]Huygens-Kamerlingh Onnes Laboratory, Leiden University, Leiden, The Netherlands

CVs are one of the most powerful tools for the experimental investigation and characterization of electron transfer processes in electrochemistry. Determined by the driving force, thus the potential of the working electrode, electrochemical interfaces can be assessed on their structure as well as activity. As the unique electrochemical fingerprint of a particular system is given by specific (current) peaks in the CV, it is not surprising that great effort is done to understand, describe, and model these peaks analytically. However, the underlying involved thermodynamic and kinetic processes makes this a difficult task, even if specific changes in the fingerprint occur upon change of the voltammogram's sweep rate. Here we show on the example of the reversible Place-Exchange peak, a precursor in the electrochemical oxidation of the Pt(111) surface, that a combination of a Frumkin isotherm followed by an Arrhenius (atomic) diffusion process delivers a rather good description. All peaks of all CVs, measured with different sweep rates, are simultaneously fitted together with only three (basic description) or six (extended description) fit variables.

O 19: Overview Talk Felix Baumberger

Time: Tuesday 9:30–10:15 Location: HE 101

Invited Talk O 19.1 Tue 9:30 HE 101

Quasiparticle dynamics and spin-orbital texture of 2D electron liquids at surfaces — •Felix Baumberger — Department of Quantum Matter Physics, University of Geneva, Switzerland — Swiss Light Source, Paul Scherrer Institute, Switzerland

Two-dimensional electron liquids (2DELs) at interfaces of transition metal oxides are foundational for the field of oxide electronics but proved notoriously hard to study spectroscopically. Here, we show that surface doping of $SrTiO_3$ and $KTaO_3$ can be used to replicate the electrostatic boundary conditions stabilizing the prototypical interface 2DELs in these oxides. Our angle resolved photoemission data on different $SrTiO_3$ and $KTaO_3$ surfaces provide direct evidence for subband formation of the *d*-electrons with a surface orientation dependent lifting of the orbital degeneracy induced by quantum confinement. Modeling the experimental electronic structure, we uncover a complex spin-orbital texture on the Fermi surface and show that it determines the large spin charge conversion effect in $SrTiO_3$. Tuning the 2DEL carrier density of $SrTiO_3$(001) by controlling the oxygen vacancy concentration on the surface, we find a remarkably complex evolution of electron-phonon coupling from a coherent polaronic liquid at low carrier density to a more conventional metal with modest short range electron-phonon coupling at high density. These results support the notion that superconductivity at the $LaAlO_3$ / $SrTiO_3$ interface is phonon mediated.

O 20: Focus Session: Molecular Nanostructures on Surfaces: On-Surface Synthesis and Single-Molecule Manipulation I

Surface-confined molecular nanostructures hold promise for applications in nanoscale (opto-) electronics, spintronics, solar cells, energy storage, and more. The bottom-up fabrication involves molecular self-assembly using non-covalent interactions, covalent on-surface synthesis, or direct molecule manipulation. Self-assembly yields highly ordered nanostructures influenced by non-covalent and adsorbate-substrate interactions, thermodynamics, and kinetics. Covalent on-surface synthesis on metallic, semi-conducting, or insulating surfaces offers new routes for complex low-dimensional nanostructures with unique properties. Direct manipulation with a scanning probe microscope enables unprecedented chemical transformations. The Focus Session provides a platform to discuss current trends in these interconnected fields from diverse experimental and theoretical perspectives. It addresses recent advancements in on-surface synthesis, manipulation, characterization, and comprehension of complex molecular architectures.

Organizer: Michael Gottfried (U Marburg), Sabine Maier (U Erlangen-Nürnberg)

Time: Tuesday 10:30–13:00 Location: HE 101

Topical Talk O 20.1 Tue 10:30 HE 101

Generation and Characterization of Cyclocarbons — Florian Albrecht[1], Igor Roncevic[2], Yueze Gao[2], Fabian Paschke[1], Shantanu Mishra[1], Harry L. Anderson[2], and •Leo Gross[1] — [1]IBM Research Europe - Zurich, Rüschlikon, Switzerland — [2]Oxford University, Oxford, United Kingdom

The family of carbon allotropes was expanded by tip-induced synthesis and on-surface characterization of cyclo[N]carbons, molecular rings composed of N carbon atoms [1-3]. Cyclocarbons possess two orthogonal delocalized pi-systems and can undergo different Jahn-Teller distortions rendering them excellent systems for benchmarking theory [4-6]. Even-N cyclocarbons can be doubly aromatic [1,3] or doubly anti-aromatic [2], and can exhibit polyynic [1,2] or cumulenic [3] structures. Odd-N cyclocarbons [5,7] do not allow a symmetric polyynic structure, might be open-shell systems, and will be discussed in the talk.

References: [1] K. Kaiser et al. Science 365, 1299-1301 (2019). [2] Y. Gao. et al. Nature 623, 977-981 (2023). [3] L. Sun, et al. Nature 623, 972-976 (2023). [4] G. V. Baryshnikov et al. J. Phys. Chem. A 124, 10849-10855 (2020). [5] G. V. Baryshnikov et al. J. Phys. Chem. A 126, 2445-2452 (2022). [6] I. Rončevic et al. (2023) 10.26434/chemrxiv-2023-8kc5t-v2. [7] F. Albrecht et al. (2023) 10.26434/chemrxiv-2023-ddrh7.

O 20.2 Tue 11:00 HE 101

Probing charge redistribution at the interface of self-assembled cyclo-P5 pentamers on Ag(111) — •Remy Pawlak[1], Outhmane Chahib[1], Yulin Yin[2], Jung-Ching Liu[1], Chao Li[1], Thilo Glatzel[1], Feng Ding[2], Qinghong Yuan[3], and Ernst Meyer[1] — [1]Department of Physics, University of Basel, Basel, Switzerland — [2]Chinese Academy of Sciences, Shenzhen, China — [3]East China Normal University, Shanghai, China

Phosphorus pentamer (cyclo-P$_5^-$) ions are unstable in nature but can be synthesized at the Ag(111) surface. Unlike monolayer black phosphorous, little is known about their electronic properties when in contact with metal electrodes, although this is crucial for future applications. Here we characterize the atomic structure of cyclo-P$_5$ assembled on Ag(111) using atomic force microscopy with functionalized tips and density functional theory. Combining force and tunneling spectroscopy, we find that a strong charge transfer induces an inward dipole moment at the cyclo-P$_5$/Ag interface as well as the formation of an interface state. We probe the image potential states by field-effect resonant tunneling 1and quantify the increase of the local change of work function of 0.46 eV at the cyclo-P$_5$ assembly. Our results suggest that the high-quality of the cyclo-P$_5$/Ag interface might serve as a prototypical system for electric contacts in phosphorus-based semiconductor devices.

O 20.3 Tue 11:15 HE 101

On-Surface Synthesis and Characterization of Pentadecacene — •Zilin Ruan[1], Tim Naumann[1], Jakob Schramm[2], John B. Bauer[3], Holger F. Bettinger[3], Ralf Tonner-Zech[2], and J. Michael Gottfried[1] — [1]Department of Chemistry, University of Marburg, 35043 Marburg (Germany) — [2]Wilhelm Ostwald Institute of Physical and Theoretical Chemistry, University of Leipzig, 04103 Leipzig (Germany) — [3]Institute for Organic Chemistry, University of Tübingen, 72076 Tübingen (Germany)

Acenes represent a unique class of polycyclic aromatic hydrocarbons that have fascinated chemists and physicists due to their exceptional potential for use in organic electronic applications. Here, we demonstrate the on-surface synthesis of pentadecacene, the longest acene known to date and here reported for the first time, via atom-manipulation-induced dissociation of a trietheno-bridged precursor on a Au(111) surface under ultra-high vacuum conditions. The geometric and electronic structures of the generated acene have been investigated by combined scanning tunneling microscopy/spectroscopy and non-contact atomic force microscopy. We observed an antiferromagnetic open-shell ground state electron configuration for pentadecacene from a spin-excitation feature at low bias, giving a singlet-triplet gap of around 124 meV. Alternatively, upon a thermal cracking of the trietheno-bridges at higher temperature, pentadecacene can be also generated and spontaneously forms complex structure with up to 6 gold atoms on Au(111) surface, suggesting a considerable multiradical contribution to its electronic ground state.

O 20.4 Tue 11:30 HE 101

On-Surface Stepwise Double Dehydrogenation for the Formation of an Undecacene Isomer — •Suchetana Sarkar[1], Kwan Ho Au-Yeung[1], Berta Álvarez[2,3], Agustín Cobas[3], Roberto Robles[4], Nicolas Lorente[4,5], Diego Peña[2,3], Dolores Pérez[2,3], and Francesca Moresco[1] — [1]Center for Advancing Electronics Dresden, TU Dresden, 01062 Dresden, Germany — [2]Centro de Investigación en Química Biolóxica e Materiais Moleculares (CiQUS), Universidade de Santiago de Compostela, Santiago de Compostela 15782, Spain — [3]Departamento de Química Orgánica, Universidade de Santiago de Compostela, Santiago de Compostela 15782, Spain — [4]Centro de Física de Materiales CFM/MPC (CSIC-UPV/EHU), 20018 Donostia-San Sebastián, Spain — [5]Donostia International Physics Center, 20018 Donostia-San Sebastián, Spain

We present the on-surface synthesis of an isomer of undecacene bearing two four-membered rings and two para-quinodimethanes. The transformation involves a thermal double deoxygenation followed by on-surface stepwise double dehydrogenation reaction of the methyl groups, locally induced by inelastic tunneling electrons on an Au (111) substrate. The structure and electronic properties of the intermediates and the final product have been investigated at the single molecule level with high spatial resolution, using both low temperature scanning tunneling microscopy/spectroscopy and non-contact atomic force microscopy. The experimental results are supported by density functional theory calculations.

O 20.5 Tue 11:45 HE 101

High-Yield On-Surface Synthesis of Isokekulene Facilitated by Strong Molecule-Substrate Interaction — •Sabine Wenzel[1], Qitang Fan[2], Anja Haags[1], Alexander Reichmann[3], Larissa Egger[3], Hans Kirschner[4], Tim Naumann[2], Simon Werner[2], Tobias Vollgraff[2], Jörg Sundermeyer[2], Jose Martinez[1], Felix Lüpke[1], Xiaosheng Yang[1], Dominik Brandstetter[3], François C. Bocquet[1], Christian Kumpf[1], Georg Koller[3], Alexander Gottwald[3], Mathias Richter[4], Michael Ramsey[3], Peter Puschnig[3], Serguei Soubatch[1], F. Stefan Tautz[1], and J. Michael Gottfried[2] — [1]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich — [2]Fachbereich Chemie, Philipps-Universität Marburg — [3]Institute of Physics, University of Graz — [4]Physikalisch-Technische Bundesanstalt (PTB), Berlin

Recent progress in on-surface synthesis has enabled the formation of novel aromatic molecules such as nanographenes, nanoribbons, and cycloarenes. Thereby, the choice of metal crystal and its surface termination can have a strong influence on the selectivity of the synthesis reaction. Here, we report the formation of the novel nonplanar cycloarene isokekulene on Cu(110) and compare it to the planar kekulene which forms on Cu(111) from the same precursor. Combining density functional theory, scanning tunneling microscopy with a CO-functionalized tip, and photoemission orbital tomography, we identify two adsorption configurations of isokekulene and reveal a strong molecule-substrate interaction including charge transfer, which account for the more favorable energetics of isokekulene on Cu(110).

O 20.6 Tue 12:00 HE 101

On-surface Synthesis of Nitrogen-Doped Carbon Nanostructures — •Tim Naumann[1], Zilin Ruan[1], Olaf Kleykamp[1], Alix Kaczmarek[2], Eugen Sharikow[1], Jörg Sundermeyer[1], Doreen Mollenhauer[2], and J. Michael Gottfried[1] — [1]Department of Chemistry, University of Marburg, 35043 Marburg, Germany — [2]Department of Chemistry, University of Giessen, 35392 Giessen, Germany

The electronic and magnetic properties of carbon-based nanomaterials can be tailored by altering the topology, introducing defect sites like vacancies, or doping with heteroatoms. Here, we present an on-surface synthesis route to a variety of nitrogen-containing 0D and 1D carbon nanostructures starting from one single precursor. Annealing 4-([1,1'-biphenyl]-2-yl)-2,6-dibromopyridine on Au(111) leads to Ullmann-coupling of the precursor molecules followed by dehydrocyclization to yield cycloarenes with different cavity sizes as well as a nitrogen-doped gulf-edge graphene nanoribbon. Ring formation of six precursor molecules leads to a flat cycloarene, which can host a metal atom in its central cavity. Filling the cavity causes an energetic shift of the cycloarene's frontier orbitals. The direction of the shift depends on the metal used. A curved cycloarene emerges from a ring formed by five precursor molecules, whereas a higher surface coverage favors chain formation. The electronic and geometric properties of the products were investigated by low-temperature scanning tunneling microscopy/spectroscopy (STM/STS) and non-contact atomic force microscopy (nc-AFM).

O 20.7 Tue 12:15 HE 101

Screening Nonalternant π-Electron Systems for Metal-Organic Interfaces: Interplay between Topology, Aromaticity, and Adsorption Behavior — •Jakob Schramm and Ralf Tonner-Zech — Wilhelm-Ostwald-Institut, Universität Leipzig, Deutschland

Metal-organic interfaces play an important role in organic electronics. Topology, aromaticity and interface properties are closely related as previously shown by experimental and theoretical studies of isomer pairs of nonalternant and alternant adsorbates. While nonalternant azulene and azupyrene chemisorb on Cu(111), the alternant isomers naphthalene and pyrene physisorb.[1,2]

This promises to be a general trend and can be exploited for interface design. Therefore, we utilize first-principles in-silico screening on a set of nonalternant and alternant molecules. We report data on molecular properties, especially aromaticity, based on simple topological approaches as well as DFT gas phase calculations. Furthermore, interface properties on the Cu(111) surface are reported based on periodic DFT calculations using a slab approach. We are able to show that interface properties are determined by corresponding gas phase properties which can already be derived from simple topological considerations.

[1] B. P. Klein, R. Maurer, R. Tonner, C. Kumpf, I. Swart, J. M. Gottfried et al., Phys. Rev. X. 2019, 9, 011030. [2] B. P. Klein, R. Maurer, G. Hilt, R. Tonner-Zech, A. Schirmeisen, J. M. Gottfried et al., ACS Nano 2022, 16, 11979-11987.

O 20.8 Tue 12:30 HE 101

On-Surface Synthesis of Nanostructures With Non-Alternant Topology — •Lukas Heuplick[1], Qitang Fan[1], Alexander Ihle[2], Pengcai Liu[3], Miguel Wiche[2], Xing-Yu Chen[3], Jiawen Cao[3], Jan Herritsch[1], Daniel Ebeling[2], Xiao-Ye Wang[3], André Schirmeisen[2], and J. Michael Gottfried[1] — [1]Department of Chemistry, University of Marburg, Marburg, Germany — [2]Institute of Applied Physics (IAP), University of Gießen, Gießen, Germany — [3]State Key Laboratory of Elemento-Organic Chemistry, Nankai University, Tianjin, China

In contrast to benzenoid and alternant structures, molecules with non-alternant binding motifs, such as azulene, show drastic changes in their electronic and optical properties. Acepleiadylene (APD), a constitutional isomer of pyrene, consists of 5-, 6-, and 7-membered rings and represents such a non-alternant structure. Here, we report the sucessful on-surface synthesis of nanostructures with non-alternant topology based on APD derivatives. Upon annealing 1,2-dibromoacepleiadylene on Ag(111), non-alternant nanographenes, most notably a covalent trimer, are formed by an Ullmann coupling. Annealing 1,2-dicyanoacepleiadylene on Cu(111), on the other hand, yields acepleiadylenocyanine - a phthalocyanine with non-alternant backbone - by cyclotetramerization. To characterize these reactions, we performed scanning tunneling microscopy (STM) and x-ray photoelectron spectroscopy (XPS) experiments. To provide further evidence of the actual structures, we used non-contact atomic force microscopy (nc-AFM).

O 20.9 Tue 12:45 HE 101

Deceptive orbital confinement at the edges and pores of carbon-based 1D and 2D nanoarchitectures — •Ignacio Piquero-Zulaica[1], Eduardo Corral-Rascón[1], Xabier Diaz de Cerio[2], Alexander Riss[1], Aran Garcia-Lekue[2], Hiroshi Sakaguchi[3], Willi Auwärter[1], and Johannes V. Barth[1] — [1]Physics Department E20, Technical University of Munich, D-85748 Garching, Germany — [2]Donostia International Physics Center (DIPC), E-20018 Donostia-San Sebastian, Spain — [3]Institute of Advanced Energy, Kyoto University, 611-0011, Kyoto, Japan

The electronic structure defines the properties of graphene-based nanomaterials. Scanning tunneling microscopy/spectroscopy (STM/STS) experiments on graphene nanoribbons (GNRs), nanographenes and nanoporous graphene (NPG) often determine an apparent electronic orbital confinement into the edges and nanopores, leading to dubious interpretations such as image potential states or super-atom molecular orbitals. Here we show that these measurements are subject to a wave function decay into the vacuum that masks the undisturbed electronic orbital shape. We use Au(111)-supported semiconducting gulf-type GNRs and NPGs as model systems fostering frontier orbitals that appear confined along the edges and nanopores in STS measurements. DFT calculations confirm that these states originate from valence and conduction bands. The deceptive electronic orbital confinement observed is caused by a loss of Fourier components, corresponding to states of high momentum. This effect can be generalized to other 1D and 2D carbon-based nanoarchitectures.

O 21: Focus Session: Spins on Surfaces studied by Atomic Scale Spectroscopies III

Time: Tuesday 10:30–13:15 Location: MA 004

Topical Talk O 21.1 Tue 10:30 MA 004

Theory of Electron Spin Resonance in Scanning Tunneling Microscopy — •Juan Carlos Cuevas[1], Christian R. Ast[2], Piotr Kot[2], Maneesha Ismail[2], Sebastián de-la-Pena[1], and Antonio I. Fernandez-Dominguez[1] — [1]Departamento de Fisica Teorica de la Materia Condensada and Condensed Matter Physics Center (IFIMAC), Universidad Autonoma de Madrid, 28049 Madrid, Spain — [2]Max-Planck-Institut für Festkörperforschung, Heisenbergstraße 1, 70569 Stuttgart, Germany

Electron spin resonance (ESR) spectroscopy in scanning tunneling microscopy (STM) has enabled probing the electronic structure of single magnetic atoms and molecules on surfaces with unprecedented energy resolution. Despite this remarkable success, the field could still greatly benefit from a more quantitative understanding of the ESR-STM physical mechanisms. Here, we present a theory of ESR-STM which quantitatively models not only the ESR signal itself, but also the full background tunneling current, from which the ESR signal is derived. We show that this theory is able to quantitatively reproduce the experimental results for a spin 1/2 system (TiH molecules on MgO) across many orders of magnitude in tunneling current, providing access to the relaxation and decoherence rates that govern the spin dynamics due to intrinsic mechanisms and to the bias voltage. More importantly, and with the help of additional electromagnetic simulations, our work establishes that the transitions in our ESR-STM experiments can be driven by the ac magnetic field at the junction.

O 21.2 Tue 11:00 MA 004

Coherent spin dynamics between electron and nucleus within a single atom — •Lukas M. Veldman[1,2], Evert W. Stolte[1], Mark P. Canavan[1], Rik Broekhoven[1], Philip Willke[3], Laëtitia Farinacci[1], and Sander Otte[1] — [1]Delft University of Technology, Delft, The Netherlands — [2]University of Stuttgart, Stuttgart, Germany — [3]Karlsruhe Institute of Technology, Karlsruhe, Germany

The study of single atom electron spins has led to both fundamental insight into the building blocks of matter as well as shown the tantalizing potential to construct functional magnetic devices atom-by-atom. The nuclear spin forms the next frontier in this effort, offering insight into intra-atomic magnetic interactions while, at the same time, promising prolonged coherence times for coherent quantum operations compared to their electronic counterparts. Here, we present coherent dynamical access to a single nuclear spin by combining electron spin resonance measurements with direct-current pump-probe spectroscopy inside a scanning tunneling microscope. We initialize both spins by means of the spin polarized tunneling electrons and control the resulting dynamics by tuning the amount of entanglement between nuclear and electron spin with the magnetic probe tip. This local spin control provides a pathway towards utilizing single nuclei for dynamic quantum simulation in extended atomic arrays.

O 21.3 Tue 11:15 MA 004

Single-shot measurement of the nuclear spin state of a single atom using ESR-STM — •Jinwon Lee[1], Evert W. Stolte[1], Hester Vennema[1], Rik Broekhoven[1], Esther Teng[1], Philip Willke[2], and Sander Otte[1] — [1]Department of Quantum Nanoscience, Kavli Institute of Nanoscience, Delft University of Technology, 2628 CJ Delft, The Netherlands — [2]Physikalisches Institut, Karlsruhe Institute of Technology, 67131 Karlsruhe, Germany

Individual nuclear spins have arisen as promising candidates for the building blocks for quantum memory because they have longer lifetime and coherence time compared to electronic spin states. Most studies on individual nuclear spins have focused on the nuclear spins embedded in solids and single-molecule magnets, which have limited controllability due to their environment. Scanning tunneling microscopy with electron spin resonance (ESR-STM), which allows for precise placement of individual atoms on a surface, recently observed the nuclear spin state through the hyperfine interaction. However, time-resolved measurements for its relevant timescales have not been reported. In this work, we achieve single-shot measurements of the nuclear spin state of ^{49}Ti atom with $S=1/2$ and $I=7/2$, adsorbed on MgO/Ag using ESR-STM. We apply continuous-wave RF electric field, which can drive ESR only when the atom has a certain nuclear spin state and observe whether ESR is driven or not by measuring tunneling conductance. This new approach enables time-resolved measurements of the nuclear spin state, and we measure its dwell time to be on the order of 100 ms, 6 orders of magnitude longer than the electronic spin in the same atom.

O 21.4 Tue 11:30 MA 004

Unveiling second harmonic resonances in ESR/STM Setups using a time dependent Anderson impurity model. — •Jose Reina Galvez[1], Roberto Robles[2], Nicolas Lorente[2], and Christoph Wolf[1] — [1]Center for Quantum nanoscience, Seoul, South Korea — [2]Centro de Fisica de Materiales, San Sebastian, Spain

The achievement of reproducible single-atom and single-molecule electron spin resonance (ESR) in a scanning tunneling microscope (STM) junction opened new possibilities in the analysis of surface science at the atomic scale [1,2]. The predominant feature of a spin driven on resonance is a resonance at the first harmonic of the Larmor frequency in the current spectrum. Higher harmonic excitations of the spin precession can emerge as long as the driving is sufficiently strong due to nonlinearity in the transport. This leads to a 2nd harmonic resonance at half the Larmor frequency.

In this talk, I will discuss the theoretical aspects of the phenomenon above. Our framework utilizes an open quantum systems formalism for a modified Anderson impurity Hamiltonian where the quantum impurity is connected by time dependent hopping terms to two fermionic baths, a polarized STM tip and a metal substrate [3,4,5]. This barrier modulation in the tunneling allows for efficient driving of the spin-1/2 and the emergence of a 2nd harmonic at driving amplitudes VRF in good agreement with the experiment.

References: [1] S. Baumann et al, Science (2015). [2] S. Kovarik et al, Nano Lett. (2022) [3] J. Reina-Gálvez et al, PRB (2021) [4] J. Reina-Gálvez et al, PRB (2023) [5] https://github.com/qphensurf/

O 21.5 Tue 11:45 MA 004

DC-bias gating of single spin electron paramagnetic resonance — Hong T. Bui[1,2], We-hyo Seo[1,2], Valeria Sheina[1,2], and •Soo-hyon Phark[1,2] — [1]Center for Quantum Nanoscience, Institute for Basic Science, Seoul, Korea — [2]Ewha Womans University, Seoul, Korea

Ti and Fe atoms in the tunnel junction of a scanning tunneling microscope combined with electron paramagnetic resonance (EPR) are prototypical quantum systems to study single spin dynamics. However, influence of DC electric bias (Vdc), followed by the spin-polarized current passing through the spins, on the EPR signal have not been studied in depth yet. Here, we introduce DC-bias gating of EPR resonance observed from single Ti and Fe adsorbates on ultrathin MgO layers. We find that a negative Vdc, inducing reversed magneto-tunneling transport, considerably suppresses EPR of Ti and accompanies a reversal of peak asymmetry. This is even more pronounced in the EPR of Fe, where the sign of the peak is inversed with a considerable intensity. Simulations on spin-dependent electron transport through a RF-driven single spin reproduce the experiments on both Ti and Fe in good agreement, revealing that DC-bias-driven spin pumping via the inelastic channels plays a key role in the RF-driven spin dynamics of the EPR-relevant two levels. In addition, a long-lived inversed population in a Fe spin can promote EPR resonance with a reversed sign as observed in the measured spectra.

O 21.6 Tue 12:00 MA 004

Pulse scheme to create and detect entanglement in surface spin systems using a tunneling microscope — •Rik Broekhoven[1], Curie Lee[2,3], Soo-hyon Phark[2,4], Sander Otte[1], and Christoph Wolf[2,4] — [1]Department of Quantum Nanoscience, Kavli Institute of Nanoscience, Delft University of Technology, 2628 CJ Delft, the Netherlands — [2]Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul 03760, Korea — [3]Department of Physics, Ewha Womans University, Seoul 03760, Korea — [4]Ewha Womans University, Seoul 03760, Korea

Being able to certify quantum entanglement is an essential element of quantum-coherent applications which has not yet been shown in spins on a surface. Here, we present a way to unambiguously identify entanglement by exploiting that entangled states undergo a free evolution with a characteristic time constant different from any other evolution in the system. Our approach is compatible with multi-spin resonance control in a scanning tunneling microscope (STM). We suggest a pulse scheme that first creates an entangled spin pair outside of the STM junction and subsequently projects the accumulated phase during free evolution on the population of one of the spins, which in turn can be read out through a weakly coupled sensor spin. The resulting measurement signal is an oscillation in the STM current at a time scale proportional to the spin pair exchange coupling if and only if the pair was entangled. We show that this scheme can be implemented using realistic simulation parameters and study the expected measurement contrast as a function of temperature and decoherence times.

O 21.7 Tue 12:15 MA 004

Syncing of stochastically switching atomic orbital memory — •Kira Junghans[1], Hermann Osterhage[1], Werner M. J. van Weerdenburg[1], Ruben Christianen[1], Eduardo J. Domínguez Vázquez[2], Hilbert J. Kappen[2], and Alexander A. Khajetoorians[1] — [1]Institute for Molecules and Materials, Radboud University Nijmegen, the Netherlands — [2]Donders Institute for Neuroscience, Radboud University Nijmegen, the Netherlands

Stochastically fluctuating multi-well systems are a promising route toward designing neuromorphic hardware. One challenge is finding tunable material platforms that exhibit such multi-well behavior.

The recently discovered atomic Boltzmann machine provides such a platform, where orbital memory states represent binary stochastic units [1,2]. Here, we investigate the response of orbital memory states of Fe and Co atoms on black phosphorus [2,3] to a sinusoidal input voltage using scanning tunneling microscopy. For both species, we observe synchronization of the state occupation to the sinusoidal signal. The state favorability of Fe atoms also shows a frequency-dependent response to the drive, which can be tuned by the input parameters. In contrast to Fe, there is no significant frequency-dependence in the state favorability for Co atoms. The response of the state favorability can be traced to the difference in the voltage-dependent switching rates of the two species based on the Poisson process model.

[1] B. Kiraly et al., Nat. Nanotechnol. 16, 414 (2021).

[2] B. Kiraly et al., Nat. Commun. 9, 3904 (2018).

[3] B. Kiraly et al., Phys. Rev. Research 4, 33047 (2022).

O 21.8 Tue 12:30 MA 004

Manipulation of Orbital States on Titanium Atoms on MgO/Ag(100) — •Susanne Baumann[1], Daria Sostina[2], Lukas Arnhold[1], Fernando Delgado[3], Philip Willke[2], and Sebastian Loth[1] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]Karlsruhe Institute of Technology, Physikalisches Institut, Karlsruhe, Germany — [3]Instituto de estudios avanzados IUDEA, Departamento de Física, Universidad de La Laguna, Spainh

The orbital and spin properties of quantum systems at the nanoscale are heavily influenced by their interaction with the surrounding environment. For atoms adsorbed on a surface, this interaction commonly leads to a partial suppression of the orbital moment, a phenomenon known as orbital quenching. In this study, we investigate and modify the orbital quenching of titanium atoms (Ti) on two monolayers of MgO/Ag(100), the experimental "fruit fly" system for electron spin resonance measurements in conjunction with scanning tunneling microscope (STM). Our study demonstrates how we can finely adjust the spin and orbital excitations by the proximity of the STM tip, ranging from a nearly unquenched orbital moment to a highly quenched state. In addition, we use multiplet and transport calculations to gain a deeper understanding of the intricate interplay between atom adsorption on a surface and the quenching of their orbital moments.

O 21.9 Tue 12:45 MA 004

From single electron ratio rules to atoms on surfaces – A relativistic investigation of hyperfine splittings — •Katharina Lorena Franzke, Wolf Gero Schmidt, and Uwe Gerstmann — Paderborn University, Warburger Str. 100, 33098 Paderborn

The hyperfine splitting of a given system is caused by the interaction of the electronic spin with the magnetic moments of the nuclei and leads to comparably small energy shifts in the absorption spectra. These shifts can be split into the isotropic Fermi contact, the anisotropic dipolar and the orbital part. In many systems, especially in semiconductors, the hybridization and deformation of the atomic orbitals by the crystal field leads to the orbital moment being no longer well defined, but effectively averaged out. This so-called orbital quenching occurs in many, but not all physical systems. We have developed a non-perturbative relativistic method which allows to calculate the contribution of the orbital part for complex structures [1]. We show that the orbital part actually scales with spin-orbit coupling if orbital quenching is hindered by local symmetry, i.e. in case of dimers or atoms at surfaces. This holds true in particular when the unpaired electron is localized in quasi-atomic p-like orbitals. Here, the orbital part is by far not negligible, but becomes dominant by surpassing the dipolar contribution by a factor of five. It is thus required to achieve overall accuracy in predicting the hyperfine interactions.

[1] Franzke K.L. *et al.*, *Relativistic calculation of the orbital hyperfine splitting in complex microscopic structures*, accepted in J. Phys. Com., 2023

O 21.10 Tue 13:00 MA 004

Atomic scale study on electronic configurations of single titanium atoms on MgO — •Hong T. Bui[1,2], We-hyo Seo[1,2], Curie Lee[1,2], Christoph Wolf[1,2], Nicolas Lorente[3,4], and Soo-hyon Phark[1,2] — [1]center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul, Korea — [2]Ewha Womans University, Seoul, Korea — [3]Centro de Física de Materiales, Donostia-San Sebastián, Spain — [4]Donostia International Physics Center, Donostia-San Sebastián, Spain

A single Ti adsorbate on two-monolayer (ML) MgO on Ag(100) is a prototypical electron spin two-level system (S = 1/2), which has recently been used to demonstrate quantum gates in on-surface atomic-scale qubit platforms using scanning tunneling microscopy (STM) combined with an electron spin resonance (ESR) [1]. Further advanced multiple-order quantum gates, however, are limited by its short coherence mainly due to the insufficient decoupling from the substrate. In this work, we investigated spin-dependent electronic properties of single Ti adsorbates on a 3-ML MgO using tunneling and ESR spectroscopies. Measurements on Ti on an O-O bridge site show a differential conductance similar to that on the same adsorption site of the 2-ML MgO and reveal S = 1/2, a promising candidate of on-surface spin qubit but with longer coherence. On the other hand, Ti adsorbed on an O-atop, in contrast to that on the 2-ML MgO (S = 1/2), shows inelastic tunneling features and uniaxial out-of-plane magnetic anisotropy, a signature of S > 1/2, which are well supported by DFT and multiplet calculations for the electronic ground state of 3d2 and S = 1.

O 22: 2D Materials II: Electronic Structure (joint session O/TT)

Time: Tuesday 10:30–12:15 Location: MA 005

O 22.1 Tue 10:30 MA 005

On the origin of circular dichroism from graphene, WSe$_2$, and other quantum materials — •Lukasz Plucinski — PGI-6 Forschungszentrum Jülich

On the example of graphene, within the dipole approximation, we discuss various contributions to the circular-dichroic angle-resolved photoemission (CD-ARPES) [1] which include phase shifts of the participating partial waves in the atomic photoionization [2], the finite inelastic mean free path induced CD [3], the interatomic phase shifts [4], and the CD due to multiple scattering of the excited electron [5]. Using tabulated phase shifts and radial integrals [6], we predict photon energies at which CD signal might exhibit sign changes, and compare the prediction to experimental results. Subsequently, we perform similar analysis for WSe$_2$, a material where orbital characters are relatively well-defined, however, varying over Brillouin zone, with different contributions at K, K', and Γ points. This can be translated into understanding CD-ARPES from topological materials. Within this context, we briefly discuss how various components needed to derive the Berry curvature, the spin characters [4], and the phases of the wave functions, can be accessed through CD-ARPES and its spin-polarized variant.

[1] Plucinski, arXiv:2309.02187 (2023), [2] Dubs et al., Phys. Rev. B 32, 8389 (1985) [3] Moser, JESRP 214, 29 (2017) [4] Heider, et al., PRL 130, 146401 (2023) [5] Daimon et al., JJAP 32, L1480 (1993) [6] Goldberg et al., JESRP 21, 285 (1981)

Topical Talk O 22.2 Tue 10:45 MA 005

Bias free extraction of orbital angular momentum from two-dimensional materials by dichroic photoemission — Jonas Erhardt[1,2], Cedric Schmitt[1,2], Philipp Eck[2,3], Philipp Kessler[1,2], Kyungchan Lee[1,2], Giorgio Sangiovanni[2,3], Ralph Claessen[1,2], and •Simon Moser[1,2] — [1]Physikalisches Institut, Universität Würzburg, D-97074 Würzburg, Germany — [2]Würzburg-Dresden Cluster of Excellence ct.qmat, Universität Würzburg, D-97074 Würzburg, Germany — [3]Institut für Theoretische Physik und Astrophysik, Universität Würzburg, D-97074 Würzburg, Germany

Topological band inversion is at the heart of the quantum spin Hall insulator (QSHI) but is difficult to demonstrate experimentally. In the bulk, this band inversion is characterized by the Berry curvature, a gauge-invariant fingerprint of the wave function's geometric properties. Intimately tied to orbital angular momentum (OAM), the Berry curvature can be in principle extracted from circular dichroism in angle-resolved photoemission spectroscopy (CD-ARPES), were it not for interfering photoelectron emission channels that obscure the OAM signature. Here, we outline a full-experimental strategy to avoid such interference artifacts and isolate the clean OAM from the CD-ARPES response. Bench-marking this strategy for the recently discovered atomic monolayer QSHI indenene, we establish CD ARPES as scale-able bulk probe to experimentally classify the topology of two dimensional quantum materials with time reversal symmetry.

O 22.3 Tue 11:15 MA 005

Orbital angular momentum of Cr$_2$Ge$_2$Te$_6$ bands using circular dichroism — •Honey Boban[1], Mohammed Qahosh[1], Xiao Hou[1], Tom G Saunderson[2,3], Yuriy Mokrousov[2,3], Claus Michael Schneider[1], and Lukasz Plucinski[1] — [1]Peter Gruenburg Institute-6, Forschungszentrum Juelich, Germany — [2]Peter Gruenburg Institute-1, Forschungszentrum Juelich, Germany — [3]University of Mainz, Germany

We present high-resolution circular-dichroic angle-resolved photoemission (CD-ARPES) spectra from graphene, WSe$_2$, and Cr$_2$Ge$_2$Te$_6$ (CGT), materials that exhibit different levels of band structure complexity. In graphene, where bands near the Fermi level are of single C 2p$_z$ orbital nature, we investigated various contributions to dichroism, such as phase shifts and final state scattering, by varying the photon energy. In WSe$_2$, near the valence band maximum Γ and K points we found strong contribution of dichroism derived from the experimental geometry, in the light incidence angle dependent CD-ARPES. We use these findings to study the OAM texture of a 2D ferromagnetic material CGT [1], in which the time reversal symmetry is broken and mirror planes are absent. CGT is interesting as it offers a platform to study the interplay of ferromagnetism and spin orbit coupling, and is a potential candidate for future spintronic devices where both spin degree of freedom and orbital degree of freedom can be utilized. Therefore, understanding the initial band OAM in CGT is crucial, as it is closely linked to its quantum transport properties [2]. Ref: [1] J. Phys. Condens. Matter **7**, 69(1995), [2] Rev. Mod. Phys. **82**, 1539(2010)

O 22.4 Tue 11:30 MA 005

Evolution of band structure in 2D Transition Metal Dichalcogenide alloy $Mo_x W_{1-x}Se_2$ — •Sarath Sasi[1], Laurent Nicolaï[1], Aki Pulkkinen[1], Christine Richter[2,3], Karol Hricovini[2,3], and Ján Minár[1] — [1]New Technologies Research Centre, University of West Bohemia, Pilsen, Czech Republic — [2]LPMS, CY Cergy Paris Université, Neuvillesur- Oise, France — [3]Université Paris-Saclay, CEA, CNRS, LIDYL, Gif-sur-Yvette, France

In the realm of two-dimensional (2D) materials research, transition metal dichalcogenides (TMDCs) have emerged as significant subjects, especially WSe_2 and $MoSe_2$, which are notable for their similar band structures. Our research aims to explore the band structure evolution in $Mo_x W_{1-x}Se_2$ alloys (x= 0 to 1) utilizing a blend of Angle-Resolved Photoemission Spectroscopy (ARPES) experimentally and its complementary theoretical one-step model photoemission calculations employing the *SPR-KKR* package [1]. Furthermore, circular dichroism [2] ARPES measurements provide an insight into the orbital characteristics, revealing Mo concentration-dependent effects that are substantiated through photoemission calculations using the coherent potential approximation(*CPA*). For homogeneous random alloys, *CPA* effectively models average scattering properties and, within the KKR formalism, ensures no extra scattering when embedding an alloy component. Studying these systems with inherent disorders uncovers fundamental insights, enhancing their potential applications.

[1]Braun, J., Minar, J., Ebert, H. (2018). Physics Reports, 740.

[2]Beaulieu, S. *et al* . (2020). Physical Review Letters, 125(21).

O 22.5 Tue 11:45 MA 005

Photon energy dependence of circular dichroism in the topologically nontrivial surface states of WTe$_2$ — •Aki Pulkkinen[1], Ján Minár[1], Shoresh Soltani[2], Khadiza Ali[2], Craig Polley[2], Balasubramanian Thiagarajan[2], and Saroj Dash[2] — [1]New Technologies-Research Centre, University of West Bohemia, Pilsen, Czech Republic — [2]MAX IV Laboratory, Lund University, Lund, Sweden

We present a theoretical and experimental study of the distorted octahedral transition metal dichalcogenide T_d-WTe_2, a type-II Weyl semimetal renowned for its intriguing physical properties, such as strong spin-orbit coupling, giant magnetoresistance, and superconductivity. Utilizing the full potential formulation of the one-step model of photoemission, our investigation focuses on elucidating the electronic and topological properties of WTe_2. By employing the theoretical model of angle-resolved photoemission spectroscopy (ARPES) and circular dichroism (CD), our results provide detailed insights into the spin texture of electronic states near the Fermi level. These findings contribute to the ongoing exploration of WTe_2, emphasizing its potential applications in quantum computing and advanced electronic devices while bridging the gap between theoretical predictions and experimental observations, including the characteristics of surface states.

O 22.6 Tue 12:00 MA 005

Ultrafast Hidden Spin Polarization Dynamics of Bright and Dark Excitons in 2H-WSe2 — •Mauro Fanciulli[1,2], David Bresteau[2], Jérome Gaudin[3], Shuo Dong[4], Romain Géneaux[2], Thierry Ruchon[2], Olivier Tcherbakoff[2], Jan Minar[5], Olivier Heckmann[1,2], Maria Christine Richter[1,2], Karol Hricovini[1,2], and Samuel Beaulieu[3] — [1]LPMS, CY Cergy Paris Université, 95031 France — [2]Université Paris-Saclay, CEA, CNRS, LIDYL, 91191 France — [3]Université de Bordeaux - CNRS - CEA, CELIA, 33405 France — [4]Beijing National Laboratory for Condensed Matter Physics, 100190 China — [5]University of West Bohemia, 30100 Czech Republic

We performed spin-, time- and angle-resolved extreme ultraviolet photoemission spectroscopy of excitons prepared by photoexcitation of inversion-symmetric 2H-WSe2 with circularly polarized light.

The very short probing depth of XUV photoemission permits selective measurement of photoelectrons originating from the top-most WSe2 layer, allowing for direct measurement of hidden spin polarization of bright and momentum-forbidden dark excitons.

Our results reveal efficient chiroptical control of bright excitons' hidden spin polarization. Following optical photoexcitation, intervalley scattering between nonequivalent K-K' valleys leads to a decay of bright excitons' hidden spin polarization. Conversely, the ultrafast formation of momentum-forbidden dark excitons acts as a local spin polarization reservoir, which could be used for spin injection in van der Waals heterostructures involving multilayer transition metal dichalcogenides.

O 23: Ultrafast Electron Dynamics at Surfaces and Interfaces III

Time: Tuesday 10:30–13:15 Location: MA 041

O 23.1 Tue 10:30 MA 041

Emergence of Floquet band structure in Dirac Hamiltonians by short pulse irradiation — •Yuriko Baba[1], Alejandro Sebastián Gómez[2], Vanessa Junk[3], Francisco Domínguez-Adame[4], Rafael A. Molina[2], and Klaus Richter[3] — [1]Universidad Autónoma de Madrid, Madrid, Spain — [2]Instituto de Estructura de la Materia, IEM-CSIC, Madrid, Spain — [3]Fakultät für Physik, Universität Regensburg, Germany — [4]GISC, Universidad Complutense de Madrid, Madrid, Spain

Floquet theory is a well-established approach to describe time-dependent quantum systems driven by a periodic external field. In the presence of such a driving, the spectrum of the system is described by replicas of the original dispersion, shifted by integers of the driving frequency [1]. Using the Floquet formalism, this work studies the emergence of the Floquet structure in the energy spectrum of Dirac Hamiltonians subject to short pulses in order to understand the limits of this formalism as well as to interpret recent results of subcycle lightwave-ARPES [2].

[1] V. Junk, P. Reck, C. Gorini, K. Richter, Physical Review B, 101, 134302 (2020). [2] S. Ito, et al, Nature, 616, 696 (2023).

O 23.2 Tue 10:45 MA 041

The challenge of realizing Floquet effects in epitaxial graphene — •Leonard Weigl[1], Niklas Hofmann[1], Johannes Gradl[1], Peter Richter[2], Thomas Seyller[2], and Isabella Gierz[1] — [1]University of Regensburg — [2]Technische Universität Chemnitz

Floquet engineering where the band structure of a solid is modified by periodic driving with strong laser pulses is a promising method for tailoring the electronic properties of solids on ultrafast time scales. One particularly tempting proposal is the opening of a topologically non-trivial gap in the Dirac cone of graphene with circularly polarized light [1]. The size of the dynamical gap is predicted to scale linearly with the inverse driving frequency [1]. On the other hand, charge carriers inside the Dirac cone need to be able to follow the driving field coherently over many cycles, which requires driving frequencies that are large compared to the inverse scattering time of the carriers [2]. Broers et al. [3] recently predicted a sweet spot for the driving frequency around 30 THz. We implemented a setup for time- and angle-resolved photoemission spectroscopy that combines a strong-field variable-wavelength multi-THz pump source with extreme ultraviolet probe pulses for putting these predictions to the test. In this talk we present our progress and discuss remaining challenges towards the experimental observation of Floquet effects in epitaxial graphene.

[1] T. Oka and H. Aoki, Phys. Rev. B 79, 081406(R) (2009) [2] Dunlap et al., Phys. Rev. B 34, 3625 (1986) [3] L. Broers and L. Mathey, Phys. Rev. Research. 4, 013057 (2022)

O 23.3 Tue 11:00 MA 041

Subcycle time-resolved THz-ARPES of Bi_2Te_3 in two-dimensional momentum space — •Tim Bergmeier, Suguru Ito, Jens Güdde, and Ulrich Höfer — Fachbereich Physik, Philipps-Universität Marburg

Angle-resolved photoemission spectroscopy (ARPES) combined with Terahertz (THz) excitation and subcycle temporal resolution offers unique capabilities for investigating the ultrafast electron dynamics of Dirac currents in the surface band of topological insulators [1]. At field strengths that are strong enough for high-harmonic generation [2], it has recently been shown in Bi_2Te_3 that this technique can even track the buildup and dephasing of Floquet-Bloch states along one particular direction parallel to the surface [3]. A full two-dimensional (2D) momentum mapping of optically excited Dirac currents, however, has been so far demonstrated only without subcycle time resolution [4].

Here, we present first results on subcycle-resolved THz-ARPES on Bi_2Te_3 in the full 2D momentum space of the surface with a recently developed experimental setup. It combines the generation of few-cycle THz-pulses in the frequency range of 12-90 THz, which allow for field strengths of several MV/cm on the sample surface, with an ultrashort 400-nm two-photon probe of less than 15 fs duration at a repetition rate of 200 kHz.

[1] J. Reimann *et al.*, Nature **562**, 396 (2018).
[2] C. P. Schmid *et al.*, Nature **493**, 385 (2021).
[3] S. Ito *et al.*, Nature **616**, 696 (2023).
[4] J. Reimann *et al.*, Sci. Rep. **13**, 5796 (2023).

O 23.4 Tue 11:15 MA 041

Nonperturbative Floquet-Landau-Zener Mechanism in Multiphoton Photoemission on Metal Surface — •Yun Yen[1,2], Marcel Reutzel[3], Andi Li[4], Hrvoje Petek[4], and Michael Schüler[1,5] — [1]Laboratory for Materials Simulations, Paul Scherrer Institute, Villigen PSI, Switzerland — [2]Ecole Polytechnique Fédérale de Lausanne (EPFL), Lausanne, Switzerland — [3]Physikalisches Institut, Georg-August-Universitat Gottingen, Gottingen, Germany — [4]Department of Physics and Astronomy and Pittsburgh Quantum Institute, University of Pittsburgh, Pittsburgh, Pennsylvania, USA — [5]Department of Physics, University of Fribourg, Fribourg, Switzerland

In the Floquet engineering picture, time periodic optical fields perturbatively replicate states shifted by photon energy quanta, and cause field-dependent Autler-Townes splitting. As the field intensifies, light matter interaction shows more non-perturbative nature. Here we reveal the onset of non-perturbative responses in multiphoton photoemission (mPP) process for a driven two-level system of Cu(111) surface states. With strong enough driving, Floquet side bands form avoided crossing gaps, and thus lead to Landau-Zener (LZ) non-adiabatic tunneling within subcycle time scale. We further simulate the population dynamics with instantaneous Floquet state (IFS) formalism, and successfully reproduce experimental mPP features. Interpretation of the mPP process by Floquet-LZ theory elaborates the importance of non-adiabatic dynamics in strong field regime.

O 23.5 Tue 11:30 MA 041

Quantification of local electric fields in ultrafast photoemission experiments — •Marco Merboldt[1], Michael Schüler[2], David Schmitt[1], Jan Philipp Bange[1], Wiebke Bennecke[1], David Ramcke[1], Karun Gadge[3], Salvatore R. Manmana[3], Sabine Steil[1], G. S. Matthijs Jansen[1], Daniel Steil[1], Michael Sentef[4], Marcel Reutzel[1], and Stefan Mathias[1] — [1]Georg-August-Universität Göttingen, I. Physikalisches Institut, Germany — [2]Department of Physics, University of Fribourg, Fribourg, Switzerland — [3]Georg-August-Universität Göttingen, Institut für Theoretische Physik, Germany — [4]Institute for Theoretical Physics, University of Bremen, Bremen, Germany

The optical control of material properties using femtosecond laser pulses is one of the driving goals in ultrafast condensed matter physics. Optical parameters to control, e.g., far-from-equilibrium phases or hidden states, are the optical wavelength, the pulse duration and sequence, the polarization, and the electric field strength. For the latter, it is important to realize that the dielectric properties of the sample screen the incident light fields. Therefore, it is desirable to be able to determine the local field strength directly from the data of the time-resolved measurement.

Here, we show how to quantify the local electric field strength in trARPES experiments. We achieve this by analyzing the polarization- and momentum-resolved photoemission intensity originating from band replica generated via the laser-assisted photoelectric effect (LAPE).

O 23.6 Tue 11:45 MA 041

Momentum transfer in the pondermotive potential of VIS/NIR laser pulses detected by time-resolved ARPES — •Christian Strüber, Xinwei Zheng, and Martin Weinelt — Freie Universität Berlin, Berlin, Germany

Pump-probe photoemission measurements of ultrafast phenomena at reflective surfaces, e.g. demagnetization process in 3d or rare earth metals detected by tr-ARPES, are affected by the interaction of electrons with the ponderomotive potential of the light field. Oscillatory energy shifts have been detected when the probe laser pulse arrives before the pump pulse [1].

In our time- and angle-resolved photoemission spectroscopy (tr-ARPES) setup [2] using a hemispherical energy analyzer we measure the ponderomotive momentum transfer in dependence of the pump intensity and wave vector, the lateral momentum of the photoelectron and energy of the electronic state in the solid. We employ sub-50fs near infrared-pulses at 1300 nm to create the ponderomotive potential. Extreme ultraviolet pulses at 35 eV allow to detect a large range of lateral and perpendicular momenta. Additional to energy shifts of energetically narrow electronic states we observe squeezing of extended electron wavepackets leading to intensity modulations. Measurement results are simulated and fitted with high accuracy. Complete reversion of the ponderomotive momentum transfer allows for retrieval of the undisturbed initial state.

[1] U. Bovensiepen et al., *Phys. Rev. B* **79** (2009) 045415

[2] B. Frietsch et al., *Rev. Sci. Instrum.* **84** (2013) 075106

O 23.7 Tue 12:00 MA 041

Three-dimensional electron dynamics in PtBi2 — Paulina Majchrzak[1], •Charlotte Sanders[2], Yu Zhang[2], Andrii Kuibarov[3], Oleksandr Suvorov[3], Emma Springate[2], Bernd Büchner[3], Alexander N. Yaresko[4], Sergey Borisenko[3], and Philip Hofmann[1] — [1]Department of Physics & Astronomy, Interdisciplinary Nanoscience Centre, Aarhus University, 8000 Aarhus C, Denmark — [2]Central Laser Facility, STFC Rutherford Appleton Laboratory, Research Complex at Harwell, Harwell, OX11 0QX, UK — [3]Leibniz IFW Dresden, Helmholtzstr.. 20, 01069, Dresden, Germany — [4]Max-Planck-Institute for Solid State Research, D-70569, Stuttgart, Germany

Interplay between the three-dimensionally dispersing Weyl states of trigonal PtBi2 and complex many-body physics at the surface has been the subject of an intriguing recent study [1]. Key to these interesting physics is a set of Weyl points in conduction band states that are not occupied at room temperature and therefore cannot be directly probed by traditional methods of angle-resolved photoemission spectroscopy (ARPES). Using pump-probe time-resolved ARPES at the UK Artemis facility, along with the capability at Artemis for photon-energy scanning, we have been able to investigate three-dimensional dynamics in the conduction band of this interesting material, and to compare the results to theory predictions relating to the quasiparticle band dispersion and topologically non-trivial states. [1] arXiv:2305.02900 [cond-mat.supr-con]

O 23.8 Tue 12:15 MA 041

Efficient carrier multiplication in the topological insulator Bi2Se3 — •Michael Herb[1], Leonard Weigl[1], Niklas Hofmann[1], Johannes Gradl[1], Jason Khoury[2], Leslie Schoop[2], and Isabella Gierz[1] — [1]University of Regensburg — [2]Princeton University

Carrier multiplication, where the absorption of a single photon results in the formation of multiple electron-hole pairs, significantly increases the quantum yield of photodetectors and solar cells. It commonly occurs in semiconductors for incident photon energies that significantly exceed the size of the band gap. This makes the topological insulator Bi2Se3 with a bulk band gap of only ~300meV a promising candidate for efficient carrier multiplication in the visible spectral range. Previous investigations of the non-equilibrium carrier dynamics in Bi2Se3 mainly focused on the topological surface state [1] and photovoltage effects [2]. We use time- and angle-resolved photoemission spectroscopy with visible 2eV pump pulses to investigate carrier dynamics inside the bulk bands. We find that the both the hole concentration in the valence band and the electron concentration in the conduction band keep increasing after the pump pulse is gone, providing direct evidence for the occurrence of carrier multiplication.

[1] Y. H. Wang et al., Phys. Rev. Lett. 2012

[2] M. Neupane et al., Phys. Rev. Lett. 2015

O 23.9 Tue 12:30 MA 041

Doping-Induced Modulation of Charge Density Wave States in $1T$-TaS$_2$: Ultrafast Insights — •Jesumony Jayabalan[1], Florian K. Diekmann[2], Negar Najafianpour[1], Ping Zhou[1], Walter Schnelle[3], Martin Eckstein[4], Kai Rossnagel[2], and Uwe Bovensiepen[1] — [1]Universität Duisburg-Essen, Germany — [2]Christian-Albrechts-Universität zu Kiel, Germany — [3]Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany — [4]University of Hamburg-CFEL, 22761 Hamburg, Germany

$1T$-TaS$_2$ displays diverse charge density wave (CDW) ordered states influenced by temperature-dependent electron-electron and electron-phonon coupling. Chemically doping $1T$-TaS$_2$ provides a potential means to adjust these couplings, thus modifying the material's properties. This presentation reports the ultrafast response of doped $1T$-TaS$_2$ mainly in incommensurate CDW state, aiming to understand changes in electronic and structural properties. We present results on periodic lattice distortions induced by an infra-red ultrafast pulse, measured using ultraviolet time-resolved photoemission spectroscopy. Samples were doped with W or Mo by substituting a fraction of Ta during growth. Alongside temperature-dependent static photoemission, resistivity, and low energy electron diffraction studies, we demonstrate that the amplitude of oscillation and damping time are strongly influenced by CDW formation, dependent on the level of doping. We find that the defect atoms act as scattering centers which hinders the oscillations causing an increased damping with doping. Funding by the DFG through FOR 5249 QUAST is gratefully acknowledged.

O 23.10 Tue 12:45 MA 041

Sub-picosecond photo-induced local distortion reduction in Sb$_2$Te chalcogenide phase-change material — •Zhipeng Huang[1], Xinxin Cheng[2], Hazem Daoud[3], Wen-Xiong Song[4], R. J. Dwayne Miller[3], Klaus Sokolowski-Tinten[1], and R. Kramer Campen[1] — [1]Faculty of Physics and Center of Nanointegration (CENIDE), University of Duisburg-Essen, Duisburg, Germany — [2]SLAC National Accelerator Laboratory — [3]Departments of Chemistry and Physics, University of Toronto — [4]Shanghai Institute of Microsystem and Information Technology, Chinese Academy of Sciences

Chalcogenide phase-change materials can be switched reversibly between a low resistive opaque crystalline phase and a high resistive transparent amorphous phase by heating, electrical or optical pulses with (sub-)nanosecond switching speed. Femtosecond laser excitation provides an ultrafast, energy-efficient way for precisely manipulating the optical, electronic and structural properties of these materials.

Here we present our ultrafast electron diffraction and femtosecond-resolved sum frequency generation spectroscopy studies on Sb_2Te material under femtosecond UV pulse irradiation. We observed a subpicosecond photo-induced non-thermal structural change in the crystallized Sb_2Te sample. Through careful structural factor calculations, we quantified the structural change corresponding to Sb/Te atom movement along the [111] plane to a less-distorted local structure by coupling to the A1g coherent optical phonon mode.

O 23.11 Tue 13:00 MA 041

Ultrafast Electron-Phonon Scattering in Dirac Antiferromagnets — Marius Weber[1,2], •Kai Leckron[1], Bärbel Rethfeld[1], and Hans Christian Schneider[1] — [1]Department of Physics and Research Center OPTIMAS University of Kaiserslautern - Landau, Campus Kaiserslautern, Germany — [2]Institut für Physik, Johannes Gutenberg University Mainz, Germany

In topological antiferromagnets the corresponding band and spin structure strongly depends on the orientation of the order parameter, for instance, two Dirac-cones with nearly linear dispersion at the border of the Brillouin zone for in-plane orientation of the Néel-vector may become a gapped system in another direction. This poses a challenge for the calculations of the electronic dynamics in these bandstructures, but the problem also applies more generally to all cases in which the electronic dynamics in the whole Brillouin zone are of interest and in which the bandstructure exhibits pronounced anisotropies. For this case and effectively 2D momentum spaces, we have developed a numerical approach which we present here for electron dynamics due to electron-phonon interactions in a model antiferromagnet. After a simulated instantaneous excitation of the electronic system, which leads to anisotropic carrier distributions, we demonstrate the effects of anisotropy on the electronic distribution functions and on macroscopic quantities like magnetization, and band-resolved carrier densities. We highlight the numerical accuracy of our implementation of the electron-phonon scattering which fully accounts for the density conservation requirements necessary for a dynamic spin-resolved calculation.

O 24: Plasmonics and Nanooptics III: Light-Matter Interaction and Spectroscopy

Time: Tuesday 10:30–13:00 Location: MA 042

O 24.1 Tue 10:30 MA 042

Identification of weak molecular absorption on single-wavelength s-SNOM images — •Iris Niehues[1,2], Lars Mester[3], Edoardo Vincentini[2], Daniel Wigger[4], Martin Schnell[2,5], and Rainer Hillenbrand[2,5,6] — [1]Institute of Physics, University of Münster, Germany — [2]CIC nanoGUNE BRTA, Donostia-San Sebastián, Spain — [3]attocubes systems AG, Munich-Haar, Germany — [4]Department of Physics, University of Münster, Germany — [5]IKERBASQUE, Basque Foundation for Science, Spain — [6]Department of Electricity and Electronics, UpV/EHU, Donostia-San Sebastián, Spain

Scattering-type scanning near-field optical microscopy (s-SNOM) and nanoscale IR point spectroscopy (nano-FTIR) allow for nanoscale optical mapping of manifold material properties. Both techniques are based on elastic light scattering at an atomic force microscope tip that is illuminated with monochromatic or broadband laser illumination. For dielectric samples, the near-field amplitude and phase signals of the tip scattered field reveal the local reflectivity and absorption, respectively. Importantly, absorption in s-SNOM imaging corresponds to a positive phase contrast relative to a non-absorbing reference sample. Interestingly, a negative phase contrast (NPC) can be observed when imaging a non-absorbing material on a highly reflecting substrate. We explore the origin of the NPC using representative test samples and demonstrate straightforward simple correction methods that remove the NPC and that allow for the identification of weak absorption contrasts. [Opt. Express 31(4), 7012 – 7022 (2023)]

O 24.2 Tue 10:45 MA 042

Near - field optical microscopy of complex plasmonic excitations — •Farid Aghashirinov, Anant Mantha, Florian Mangold, Julian Schwab, Bettina Frank, and Harald Giessen — 4-th Physics Institute, University of Stuttgart, Stuttgart, Germany

We experimentally and theoretically study surface plasmon polaritons (SPPs) on single crystalline gold platelets of both long- and short-range type. Short-range surface plasmon polaritons (SR-SPPs) have received less attention compared to long-range SPPs (LR-SPPs) due to high attenuation losses, which shortens their propagation length. To investigate the complex SPP near - field pattern on single crystalline gold platelet, we utilize a reflection s - SNOM combined with a tunable broadband laser source. We disentangle excitations coming both from the scanning tip and the gold platelet edges by applying a Fourier analysis method. This allows us to determine the SPP wavelength and furthermore identify hidden excitations that are covered by interference of other waves. Fourier filtering makes it possible to identify the propagation direction of short-range surface plasmon polaritons, as well as their propagation length. In addition, we explore the impact of platelet thickness on the short - range SPP wavelength, which in future will give us another tuning parameter for scaling and combining complex near - field optical microscopy with topological plasmonics.

O 24.3 Tue 11:00 MA 042

Near-field Fano spectroscopy of MaPbI3 nanoparticles — Jinxin Zhan[1], •Tom Jehle[1], Sven Stephan[1], Sam Nochowitz[1], Ekaterina Tiguntseva[2], Sergey Makarov[2], Juanmei Duan[1], Petra Gross[1], and Christoph Lienau[1] — [1]Universität Oldenburg, D-26129, Germany — [2]St. Petersburg, Russia

Dielectric nanoparticles particles have optical shape resonances that confine light on the nanoscale in localized modes with well-defined spatial field profiles. A particularly interesting example are halide perovskite nanoparticles, for which the coupling between excitons and Mie modes results in Fano lineshapes in the spectral domain [1]. Here, we use a new broadband, interferometric sSNOM technique [2] to probe the time dynamics of the local optical near-fields of such particles. We measure amplitude and phase of the scattered light field in a broad spectral range and with 10 nm spatial resolution. Direct Fourier transformation gives the time dynamics of the local electric field, recorded with sub-cycle resolution. We uncover biexponential near-field decays with a characteristic destructive interference dip after a few fs. In the spectral domain, this corresponds to a Fano resonance with an unual 2π phase jump. We show that this signature arises from the intereference between spectrally broad dipole and narrow quadrupole resonances of the particles. Our results give new insight into the optical properties of high-index, active semiconductor nanoparticles with intriguing applicatoins for nanoscale all-opticals switching and lasing. [1] Tiguntseva, E. Y., et al. Nano Lett. 2018, 18 (2), 1185-1190. [2] Zhan, J., et al. Advanced Photonics 2020, 2 (04).

O 24.4 Tue 11:15 MA 042

Direct programming of confined Surface Phonon Polariton Resonators with the plasmonic phase-change material In_3SbTe_2 — Lukas Conrads, Luis Schüler, Konstantin Wirth, Matthias Wuttig, and •Thomas Taubner — I. Institute of Physics (IA), RWTH Aachen University

Tailoring light-matter interaction is essential to realize nanophotonic components and can be achieved with surface phonon polaritons (SPhPs), an excitation of photons coupled with phonons of polar crystals. Ultra-confined resonances are observed by restricting the SPhPs to cavities. Phase-change materials (PCMs) enable non-volatile programming of these cavities based on a change in the refractive index [1]. Recently, the new plasmonic PCM In_3SbTe_2 (IST) was introduced which can be reversibly switched from an amorphous dielectric to a crystalline metallic state in the infrared to realize numerous nanoantenna geometries [2]. However, the application potential of IST to create and modify SPhP resonators has not been exploited yet. Here, we demonstrate direct programming of confined SPhP resonators by phase-switching IST on top of a polar silicon carbide crystal and investigate the strongly confined resonance modes with scanning near-field optical microscopy. Reconfiguring the size of the resonators themselves result in enhanced mode confinements up to a value of $\lambda/35$ [3]. This study is a first step towards rapid prototyping of reconfigurable SPhP resonators even with hyperbolic and anisotropic 2d materials.

[1] Wuttig et al., *Nat. Photon.* **11**, 465 (2017) [2] Heßler et al., *Nat.*

Commun. **12**, 924 (2021) [3] Conrads et al. *arXiv:2310.12841* (2023)

O 24.5 Tue 11:30 MA 042

Observation of Anisotropic Phonon Polariton Propagation with Sum-Frequency Generation Microscopy — •Richarda Niemann[1], Sören Wasserroth[1], Gonzalo Álvarez-Pérez[2], Javier Martín-Sánchez[2], Pablo Alonso-Gonzáles[2], Martin Wolf[1], and Alexander Paarmann[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin — [2]Department of Physics, University of Oviedo, Oviedo 33006, Spain

Surface phonon polaritons (SPhPs) have been proven an interesting tool in the field of nanophotonics as they can be used in applications in the mid- to far-IR regime with lower losses than their plasmonic counterparts.[1] Commonly, propagating SPhPs are imaged using tip-based near-field techniques like scanning near-field optical microscopy which suffer from long image acquisition times due to the point-scanning approach and thus are often limited to only a few selected frequencies.[2] Here, we observe propagating phonon polaritons launched on an m-cut AlN substrate in a far-field approach by interferometric wide-field imaging. Our technique of sum-frequency generation (SFG) spectromicroscopy offers high spatial and spectral resolution at rapid acquisition speed.[3] We are able to extract the anisotropic dispersion of the propagating polaritons and compare them to numerical simulations.

[1] Caldwell et al., Nanophotonics 4, 1 (2015)
[2] Barnett et al., Appl. Phys. Lett. 120, 211107 (2022)
[3] Niemann et al., Appl. Phys. Lett. 120, 131102 (2022)

O 24.6 Tue 11:45 MA 042

Surface phonon polariton ellipsometry — •Giulia Carini, Richarda Niemann, Niclas Sven Müller, Martin Wolf, and Alexander Paarmann — Fritz Haber Institute, Berlin, Germany

Phonon polaritons are light-matter electromagnetic waves emerging in polar crystals from the hybridization of infrared impinging photons with IR-active phonon resonances. The light-matter coupling appears in the bulk polariton dispersion as an avoided crossing and gives rise to a spectral region - known as reststrahlen band (RB) - where the material optically behaves like a metal, only supporting evanescent surface waves, while light propagation inside the crystal is prohibited. The excitation of surface phonon polaritons (SPhPs) requires special experimental schemes to overcome the momentum mismatch between SPhPs and free-space radiation.

In our contribution, we investigate surface phonon polaritons at the interface between gallium phosphide (GaP) and air in momentum-domain by means of the Otto-type prism coupling geometry, a total internal reflection-based approach. Furthermore, we combine this technique with spectroscopic ellipsometry to enable the acquisition of both amplitude and phase information of the reflected waves along the whole RB of GaP. The adjustability of the prism-sample air gap width enables a systematic study of the ellipsometry parameters' dependence on the optical coupling efficiency. In particular, we show that combined observation of both ellipsometry parameters - amplitude and phase - provides a powerful toolkit for the detection of SPhPs, even in the presence of high optical losses.

O 24.7 Tue 12:00 MA 042

Ultra-confined THz hyperbolic phonon polaritons in $HfSe_2$ — Ryan Kowalski[1], Gonzalo Alvarez-Perez[2], Niclas S. Müller[3], Maximilian Obst[4], Giulia Carini[3], Saurabh Dixit[1], Katja Diaz-Granados[1], Aditha S. Senarath[1], Lukas M. Eng[4], Martin Wolf[3], Thomas G. Folland[5], Pablo Alonso-Gonzalez[2], Susanne C. Kehr[4], •Alexander Paarmann[3], and Joshua D. Caldwell[1] — [1]Vanderbilt University, Nashville, TS, USA — [2]University of Oviedo, Oviedo, Spain — [3]Fritz Haber Institute of the Max Planck Society, Berlin, Germany — [4]Institute of Applied Physics, TUD Dresden University of Technology, Dresden, Germany — [5]The University of Iowa, Iowa City, IA, USA

Hyperbolic phonon polaritons have recently attracted tremendous attention due to their ability to confine infrared- to terahertz light to deeply subwavelength dimensions. So far, however, high-quality polaritons in the THz region have only been reported for very few naturally hyperbolic materials.[1] Here, we experimentally demonstrate ultraconfined hyperbolic phonon polaritons in the 3-5 THz spectral range in ultrathin flakes of the van der Waals crystal $HfSe_2$,[2] with confinement factors of up to ≈80 below free space wavelength. Additonally, we also observe a unique case of strong coupling between the hyperbolic mode with the intrinsic epsilon-near-zero polariton emerging from the out-of-plane longitudinal phonon resonance within the hyperbolic band.

[1] T. de Oliveira, et, Adv. Mat. 33, 2005777 (2021).
[2] R.A. Kowalski, Adv. Opt. Mat. 10, 2200933 (2022).

O 24.8 Tue 12:15 MA 042

THz Cavities for Investigation of Strong Light-Matter Coupling — •Michael S. Spencer, Maximilian Frenzel, Joanna M. Urban, and Sebastian F. Maehrlein — Fritz Haber Institute of the Max Planck Society, Berlin, Germany

Next-generation materials science aims for precise and ultrafast manipulation of structural properties and fundamental excitations to alter material properties on-demand, in search of emergent, enhanced, or even hidden states of matter. A novel pathway for achieving these goals is through modulation of light-matter coupling, experimentally achieved using electromagnetic cavities. Modification of low-energy material properties using Terahertz (THz) cavities is a nascent research focus, enabled in recent years by the availability of high-intensity THz radiation sources.

Here, we present our recent development of new Fabry-Pérot cavity designs for experimental realization of THz cavities. These designs allow for full tunability of the THz cavity resonance frequencies, while simultaneously allowing a sample to be positioned at a field-maximum, boosting light-matter interaction. By making use of electrooptic sampling, we measure the full cavity mode spectrum in amplitude and phase. Our experimental and analytical results can be generalized to complex, multi-layered cavities, allowing for a rigorous description of samples beyond the thin-layer approximation. Furthermore, we will experimentally demonstrate how the practical design of such double-cavities provides a unique avenue for simple and direct investigations of light-matter interactions in the THz and mid-infrared spectral regions.

O 24.9 Tue 12:30 MA 042

Generative inverse design of functional molecules for plasmonic nanodevices — •Zsuzsanna Koczor-Benda, Shayantan Chaudhuri, Joe Gilkes, and Reinhard J. Maurer — University of Warwick, Coventry, United Kingdom

The interaction between molecules and strongly confined electromagnetic fields at metallic nanostructures results in extreme enhancement of molecular spectroscopic signals. This effect can be utilized in new nanoscale devices such as molecular terahertz (THz) detectors. However, to achieve high efficiency, molecules with highly specialized properties are required. We explore how quantum chemistry and machine learning methods can provide good candidate molecules for these applications. In particular, we investigate a promising new THz detection technique based on frequency upconversion by molecular vibrations. By screening databases containing millions of molecules, a two-orders-of-magnitude improvement of spectral intensity can be achieved. Generative machine learning provides a route for going beyond existing molecular databases, to instead design new functional molecules by biasing towards the desired properties. We discuss how recent developments in inverse property-driven design open the way for the targeted generation of molecules for THz detection.

O 24.10 Tue 12:45 MA 042

A microscopic approach for active plasmonics in THz-pumped metal nanoparticles — •Jonas Grumm[1], Robert Salzwedel[1], Holger Lange[2], and Andreas Knorr[1] — [1]Institut für Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Technische Universität Berlin, Berlin, Germany — [2]Institut für Physikalische Chemie, Universität Hamburg, Hamburg, Germany

The optical response of metal nanoparticles is dominated by the formation of collective electronic resonances, forming localized plasmons. To gain insight into their dynamics and for switching applications, it would be beneficial to influence the plasmonic resonance actively.

Here, we present theoretical models for an active tuning of the plasmonic resonance in optical spectra by using strong THz fields. We discuss theoretical descriptions with different complexity, ranging from a numerical solution of kinetic equations for the electron dynamics to coarse-grained hydrodynamic descriptions of metal nanoparticles. In all cases, we merge the electron dynamics and Maxwell's equations to obtain self-consistent solutions. A renormalization of the optical metal nanoparticle plasmon resonance by the THz field can be realized by THz-induced spatial gradients.

O 25: Organic Molecules on Inorganic Substrates III: Adsorption & Growth

Time: Tuesday 10:30–13:00 Location: MA 043

O 25.1 Tue 10:30 MA 043

(Meta)stable monolayers on metals: Kinetic trapping of the flat-lying phase — •Anna Werkovits[1], Simon B. Hollweger[1], Max Niederreiter[2], Thomas Risse[3], Johannes J. Cartus[1], Martin Sterrer[2], Sebastian Matera[4], and Oliver T. Hofmann[1] — [1]Institute of Solid State Physics, TU Graz — [2]Institute of Physics, KFU Graz — [3]Institut für Chemie und Biochemie, Freie Universität Berlin — [4]Theory Department, Fritz Haber Institute of the MPG

While conventional wisdom dictates that conjugated organic molecules on metals adopt a flat-lying configuration, recent studies indicate coverage-dependent shifts to upright-standing phases, which exhibit distinctly different physical characteristics. Our investigation challenges the prevailing notion by arguing that, from an energetic standpoint, thermodynamically stable upright-standing phases may be more widespread than previously believed. However, these phases are often not found experimentally due to kinetically trapping under typical experimental conditions. Using a combination of first principles kinetic Monte Carlo simulations and experiment, we show that during the growth process of organic molecules, phases with a lower molecular density (i.e., flat-lying molecules) are formed before the thermodynamically more stable upright standing phase. Notably, the transition to the upright-standing phases is frequently hindered kinetically in standard surface science conditions. Our exploration of growth conditions and interface energetics reveals that the phase transition time is primarily dictated by the deposition rate, and largely independent of the nature of the organic molecule.

O 25.2 Tue 10:45 MA 043

CoPc/F_{16}CuPc on Ag(100): Triggering the condensation of a 2D molecular gas — •Thorsten Wagner[1], Blazej Golyszny[2], Grazyna Antczak[2], and Peter Zeppenfeld[1] — [1]Institute of Experimental Physics, Johannes Kepler University Linz, Austria — [2]Institute of Experimental Physics, University of Wroclaw, Poland

When metal phthalocyanine molecules are deposited on metallic surfaces, they often form a 2D molecular gas in the submonolayer regime: Instead of a fixed position as in a condensed phase such as 2D islands, they are very mobile on the surface and change frequently their adsorption sites and geometry. The reason for this is a so-called surface-mediated repulsion, which may stabilizes the 2D gas phase even far beyond a coverage of 0.5 ML. This repulsion can be compensated by using mixtures of hydrogen- and fluorine-termined phthalocyanines. Here, we use photoelectron emission microsopy (PEEM) to investigate ultrathin films of cobalt-Pc (CoPc) and perfluorinated copper-Pc (F_{16}CuPc) on Ag(100) surfaces. By following the evolution of the standard deviation[1] of the image sequences acquired during subsequent deposition of the molecules, the 2D molecular gas and the condensed bi-molecular phase can be identified. Regardless of the sequence of deposition, first CoPc and then F_{16}CuPc or vice versa, we can trigger the condensation of the 2D gas into a dense structure with a 1:1 ratio of the two molecules. The mixing of the molecules is actually not limited to the first layer on the substrate.

[1] Th. Wagner et al. in Ultramicroscopy 233, 113427 (2022)

O 25.3 Tue 11:00 MA 043

Constructing chiral overlayers: From single molecules to closed layers — •Jonas Brandhoff[1], Fumi Nishino[2,3], Keisuke Fukutani[2,3], Marco Gruenewald[1], Maximilian Schaal[1], Felix Otto[1], Roman Forker[1], Satoshi Kera[2,3], and Torsten Fritz[1] — [1]Institute of Solid State Physics, Friedrich Schiller University Jena, Helmholtzweg 5, 07743 Jena, Germany — [2]Institute for Molecular Science, Myodaiji, 444-8585, Okazaki, Japan — [3]The Graduate University of Advanced Studies, Hayama-cho, 240-193 Kanagawa, Japan

Recently, the interest in chiral molecules has spiked. The chirality-induced spin selectivity (CISS) promises a high spin-polarizability for electrons traversing a chiral molecule. However, the CISS effect is yet not fully understood. To be able to probe the CISS effect with area averaging methods like photoelectron spectroscopy a well-defined chiral surface, consisting of chiral molecules, is needed. In this study the chiral molecule BINAP is investigated on a Au(111) surface. To understand how chirality from one single molecule evolves into a fully chiral overlayer, a coverage dependent study, showing many different structural motifs of BINAP, was done. This structural evolution was investigated using scanning tunneling microscopy (STM) and low-energy electron diffraction (LEED). In combination, these methods reveal a chiral lattice and the role of the different molecule-molecule and molecule-substrate interactions will be discussed. This study offers a deeper insight in the engineering of chiral surfaces and opens a possible pathway towards spintronic applications.

O 25.4 Tue 11:15 MA 043

Vibrational and structural analysis of P2O and P4O monolayers on Ag(111) and Ag(110) — •Ravi Priya, Keda Jin, Weishan Wu, and Peter Jakob — Department of Physics, Philipps-Universität Marburg, Germany

Various phases and associated phase transformations have been studied for 6,13-pentacenequinone (P2O) and 5,7,12,14-pentacenetetrone (P4O) on Ag(111) and Ag(110) using SPA-LEED, FTIRAS & TDS. For P2O, sub-monolayer regime is governed by formation of long rows, aligned along the [110] direction with varying distances between rows. The interaction along the rows is attractive and either weak or slightly repulsive between rows. Close to completion of the monolayer, the streaky LEED patterns turn into sharp, well-defined reflexes. For P4O, attractive intermolecular and molecule-metal interactions lead to ordered and uniform structures for sub-monolayers on both substrates. Lateral ordering is governed by orientation-dependent intermolecular interactions including steric hindrance and hydrogen bonding leading to distinct coverage and temperature-dependent phases on both substrates. IRAS suggests more prominent Interfacial Dynamical Charge Transfer (IDCT) for P4O vs P2O on Ag(111) due to a notably higher DOS of the P4O-LUMO at ε_F. Molecule-metal interaction is enhanced on Ag(110) vs Ag(111) for both molecules. Intensity, asymmetry, and line shape changes of IDCT-bands reflect the dynamics of vibrational modes and are associated with the DOS of the molecule-LUMO at ε_F along with the overall strength of respective molecule-metal interaction.

O 25.5 Tue 11:30 MA 043

Impact of alkyl side groups on the structure formation of merocyanines on the Ag(100) surface studied by STM, SPA-LEED, and NIXSW — •Anna Juliana Kny[1], Anja Haags[1], Sergey Subach[2], and Moritz Sokolowski[1] — [1]Clausius Institut für Physikalische und Theoretische Chemie, Universität Bonn, Germany — [2]Forschungszentrum Jülich, Germany

The control over the molecular packing is an important key aspect for the properties of molecular optoelectronic devices. In our study, we focus on the self-organization of merocyanine (MC) molecules which are in focus for applications due to high transition dipole moments.[1] Starting from the prototype MC 2-[5-(5-dibutylamino-thiophen-2-yl-methylene)-4-*tert*-butyl-5*H*-thiazol-2-ylidene]-malononitrile (HB238) [2] we systematically altered the length of the alkyl side groups and investigated their influence on the adsorption and ordering on the Ag(100) surface. We used SPA-LEED, STM, XPS, and NIXSW. Remarkably, a formation of homochiral tetrameric aggregates is observed for all investigated MCs, independently from their alkyl side groups. However, the side groups have a large impact on the lateral packing of the aggregates and the registry to the underlying Ag(100) surface. We present NIXSW data which give an additional insight to the surface bonding of the tetrameric aggregates. The MC were kindly provided by the Meerholz group (Cologne). We acknowledge experimental support from D. Duncan (DLS). Supported by the DFG through the research training group 2591 and the Diamond Light Source, UK. [1] JACS 137 (2015) 13524. [2] Nanoscale 15 (2023) 10319.

O 25.6 Tue 11:45 MA 043

Adsorption and Orientation of N-heterocyclic Carbenes and Olefines on Metallic Surfaces — •Felix Landwehr[1], Mowpriya Das[2], Sergio Tosoni[3], Juan Navarro[1], Ankita Das[2], Maximilian Koy[2], Markus Heyde[1], Gianfranco Pacchioni[3], Frank Glorius[2], and Beatriz Roldan Cuenya[1] — [1]Fritz-Haber Institute of the MPG, Berlin, Germany — [2]Westfälische Wilhelms-Universität, Münster, Germany — [3]Università di Milano-Bicocca, Milano, Italy

N-Heterocyclic carbenes (NHCs) have attracted scientific interest in recent years due to their excellent properties as surface modifiers with applications in materials science and catalysis. Structurally similar to

NHCs are N-heterocyclic olefins (NHOs), comprising of an additional terminal alkylidene moiety (CH2) at the carbene position. NHOs possess highly polarizable and remarkably electron-rich double bonds and have been effectively utilized as exceptional anchors for surface modifications. We investigated the adsorption, orientation and electronic properties of the sterically low-demanding IMe-NHO compared to its analogous IMe-NHC counterpart on a Cu(111) surface. High-resolution electron energy-loss spectroscopy (HREELS) measurements show a flat-lying ring adsorption configuration for both molecules. While the NHC adopts a dimer configuration including a Cu adatom, the NHO chemisorbs over a C-Cu bond perpendicular to the surface. This distinct difference leads for the NHOs to a higher net electron transfer from the surface compared to the NHCs. These results highlight the role of NHOs in surface science as they extend the functionalization capabilities of NHCs into stronger electronic modification.

O 25.7 Tue 12:00 MA 043

Passivation of a metal surface strengthens the binding of an N-heterocyclic carbene to a metalloporphyrin pedestal — •Dennis Meier[1], Peter Knecht[1], Peter Feulner[1], Francesco Allegretti[1], Joachim Reichert[1], Johannes V. Barth[1], and Anthoula C. Papageorgiou[1,2] — [1]Technical University of Munich, Germany — [2]National and Kapodistrian University of Athens, Greece

The extension of well-ordered 2D self-assemblies to more complex 3D architectures in ultra-high vacuum will bridge the gap between atomically precise supramolecules and extended materials. It is therefore crucial to understand the binding of out-of-plane linker molecules. We recently introduced a strategy to use Ru-porphyrins as pedestals of N heterocyclic carbenes (NHCs) on a metal surface. Here, we present engineering of the Ru-porphyrin metal interface for a significant improvement in the thermal stability of such pedestal systems. The enhanced binding strength is achieved by introducing a layer of O atoms between the Ru porphyrins and the metal surface. We characterize unambiguously the chemical state, structure and thermal stability of NHC molecules adsorbed to Ru octaethyl porphyrin on Cu(110) and Cu(110)-(2x1)O by means of photoelectron spectroscopy, scanning tunneling microscopy and temperature programmed desorption. On Cu(110)-(2x1)O, more extended 2D domains of pedestals and an increment of NHC binding stability by around 130 K was realized.

O 25.8 Tue 12:15 MA 043

Molecular self-assembly of C_{60} on $CsPbBr_3(100)$ — •Hannah Loh, Andreas Raabgrund, and M. Alexander Schneider — Universität Erlangen-Nürnberg, 91058 Erlangen, Germany

$CsPbBr_3$ is a promising alternative to organic-inorganic perovskites in solar cells. Aiming at higher efficiencies, the band alignment of the perovskites and the transport layers within the solar cell are a crucial parameter [1]. Evaporating the single components CsBr and $PbBr_2$ in a 1:1 stoichiometry, thin $CsPbBr_3$ films grow epitaxially in a (2×2) cell with respect to Au(100). For coverages greater than 5 ML two surface reconstructions, (2×2) and $\left(2\sqrt{2}\times\sqrt{2}\right)$ R45°, of the $CsPbBr_3(100)$ are found [2].

In this contribution we exploit these films to investigate the self assembly of C_{60}, which is a known electron transport material in solar cells, on $CsPbBr_3$/Au(100) by STM, LEED and DFT. For submonolayer coverage of C_{60} we find hexagonal closed-packed islands with different alignments with respect to the substrate that are not influenced by the reconstruction. The C_{60} molecules within the islands show rotational orientation without apparent lateral order. After annealing to 420 K the islands become multilayer structures. STS measurements reveal a bandgap of 2.8 ± 0.1 eV for $CsPbBr_3$ and molecular states at 0.9 ± 0.1 eV (LUMO) and at 2.3 ± 0.1 eV (LUMO+1) in the monolayer C_{60} configuration.

[1] Schmidt-Mende et al., APL Materials, **9**, 109202 (2021)

[2] Rieger J. et al., Phys. Rev. Mat, **7**, 035403 (2023)

O 25.9 Tue 12:30 MA 043

Molecular adsorption on multidomain h-BN/Pt(111): Exploring 2D material behavior for different rotational domains — •Rocío Molina-Motos[1] and Antonio J. Martínez-Galera[1,2,3] — [1]Departamento de Física de Materiales, Universidad Autónoma de Madrid, E-28049 Madrid, Spain. — [2]Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid, E-28049 Madrid, Spain — [3]Instituto de Ciencia de Materiales Nicolás Cabrera, Universidad Autónoma de Madrid, E-28049 Madrid, Spain

Exploration into the domain of molecular adsorption on two-dimensional (2D) materials carries significant expectations, offering insights into unprecedented physicochemical phenomena and paving the way for the future development of nanodevices [1,2]. The present work explores the intricacies of the adsorption of the prototypic molecule Perylenetetracarboxylic dianhydride (PTCDA) on single layers of polycrystalline hexagonal boron nitride (h-BN) grown on Pt(111) surfaces, focusing on the structural and electronic properties of adsorbed molecules near domain boundaries. This research has been conducted under ultrahigh vacuum conditions, utilizing a variable temperature scanning tunneling microscope as the main characterization tool.

References:

[1] A. Kumar, K. Banerjee and P. Liljeroth. Nanotechnology 28, 082001 (2017).

[2] H. Guo, M. D. Jiménez-Sánchez, A. J. Martínez-Galera, J. M. Gómez-Rodríguez. Nanoscale 15, 5083-5091 (2023).

O 25.10 Tue 12:45 MA 043

Analysis of thiahelicene non-covalent molecular wires using nc-AFM — •Gema Navarro[1], Antoine Hinaut[1], Shuyu Huang[1], Yunbin Hu[2], Thilo Glatzel[1], Akimitsu Narita[2], and Ernst Meyer[1] — [1]Department of Physics, University of Basel, Klingelbergstrasse 82, 4056 Basel, Switzerland. — [2]Max Plank Institute for Polymer Research, Ackermannweg 10, 55128, Mainz, Germany.

Helicene derivatives are widely used in the field of Circularly Polarized Organic Light-Emitting Diodes (CP-OLEDs)[1] due to their chiroptical properties. According to previous reports an enhancement in their optical response have been obtained by means of the self-assembled formation. The surface science studies are beneficial for the further comprehension of this phenomena. On surface, the zigzag twin row configuration constitutes by far the predominant molecular phase formed on metal surfaces, however factors such as temperature, surface nature and heteroatoms incorporation have been shown to have an strong influence in the self assembled pattern.

In this work, we addressed the study of the adsorption of a sulfur-containing double helicene on Cu(111) surface. Formation of non-covalent molecular wires, widely spread on the terraces, were observed after the deposit and gentle annealing. The wires symmetry indicates an strong influence with the surface. Furthermore, this configuration was observed at low, medium and high coverage regimen. All the images presented were acquired at room temperature by means a home-built nc-AFM microscope under UHV conditions.

[1] Meng, G. et al. Adv. Mater. 2023, 2307420.

O 26: Focus Session: Spin Phenomena in Chiral Molecular Systems I (joint session O/TT)

Spin phenomena in monolayers and thin films of enantiopure chiral molecules have recently attracted great attention. These phenomena are attributed to the chiral induced spin selectivity (CISS) effect, which leads to, for instance, different transmissions for the longitudinal orientation of the electron spin through helical molecules arranged with their molecular axes perpendicular to the surface. A microscopic theoretical understanding of the CISS effect has not been achieved so far. Therefore, new tools are being developed to uncover the different influences of electronic structure, spin-orbit coupling, and (chiral) phonons on the CISS effect. Applications have already been demonstrated in spin-directed chemistry, spintronics, and quantum technologies. The purpose of this Focus Session is to review the state-of-the-art and also to report new results.

Organizers: Katharina Franke (FU Berlin), Benjamin Stadtmüller (TU Kaiserslautern),
Helmut Zacharias (U Münster)

Time: Tuesday 10:30–13:15 Location: MA 141

Topical Talk O 26.1 Tue 10:30 MA 141

The Electron's Spin and Chirality - a Miraculous Match — •Ron Naaman — Dep. of Chemical and Biological Physics, Weizmann Institute, Rehovot, Israel

Spin based properties, applications, and devices are commonly related to magnetic effects and to magnetic materials. However, we established that chiral material could act as spin filters for photoelectrons transmission, in electron transfer, and in electron transport. The effect, termed Chiral Induced Spin Selectivity (CISS), has interesting implications in wide range of fields like spintronics, spin controlled chemistry, long range electron transfer, enantio-recognition, and enantio-separation. The basic effect, and its applications and implications, will be presented.

Topical Talk O 26.2 Tue 11:00 MA 141

Electrons, Vibrations and Chirality — •Martin B. Plenio — Institut of Theoretical Physics and Center for Quantum Biosciences, Ulm University, Germany

Chirality - the handedness of molecules and larger structures - plays an important role in a plethora of biological, chemical, and physical processes. Peptides, DNA-helices, and small chiral molecules are identical in their constitution apart from their handedness and can have completely different interactions with biological receptors. For example, their taste can vary between chocolate or mint (menthol) or their effect can be either tranquilising or cause birth defects (thalidomide) depending on their chirality.

In 2011, it was observed that photo-electrons passing through a self-assembled monolayer of double-stranded DNA with a fixed helicity on a gold surface acquire significant spin polarisation. This chirality-induced spin selectivity (CISS) has been confirmed in various molecules.

However, there is no consensus regarding the microscopic mechanism that can explain the experimental data quantitatively. Here I would like to discuss theoretical models that combine vibrational molecular dynamics and momentum conservation to provide a possible mechanism to explain CISS and discuss potential experimental signatures that one may explore to confirm or refute this hypothesis.

Topical Talk O 26.3 Tue 11:30 MA 141

Electrical Dipole Moment Governs Spin Polarization in Charge Transport in Single α-helical Peptides Junctions — •Ismael Diez-Perez — Department of Chemistry, Faculty of Natural and Mathematical Science, Kings College London, Britannia House, 21 Swan St

The recent discovery of the CISS (Chirality-induced Spin Selectivity) has many implications for our understanding of biological ET (electron transfer/transport). In this contribution, we are presenting our latest experimental results on spin-dependence ET through single-molecule electrical contacts made of chiral alpha-helical peptide backbones. We synthesize two alpha-helical peptides with homologous sequences of different lengths, 17 and 22 amino acids, and with their two corresponding D- and L- optical isomer. The latter includes the retro-versions of the same sequence, i.e., the same peptide sequence but in an inverted order. To measure spin-dependent single-molecule ET in such structures, we use a magnetic STM break-junction approach we have previously exploited to measure magnetoresistance in single-molecule contacts. The results are explained by an intuitive picture that includes both CISS and interfacial effects. The simple picture has enough flexibility to accommodate the description of the observed differences in magnetoresistance as a function of the magnetization direction of the Ni electrode, the chirality of the peptide, and the peptide dipole orientation on the electrode surface.

O 26.4 Tue 12:00 MA 141

Spin-Resolved Photoemission Studies of Heptahelicene and Tetrapyrrole (Sub-)Monolayers — •Paul V. Möllers[1], Bianca C. Baciu[2], Rafael Rodriguez[3], Johannes Seibel[4], Adrian J. Urban[5], Albert Guijarro[2], Jeanne Crassous[3], Karl-Heinz Ernst[4], Hiroshi M. Yamamoto[5], and Helmut Zacharias[1] — [1]Center for Soft Nanoscience (SoN), Univ. Münster, Germany — [2]Inst. Universitario de Síntesis Orgánica, Univ. de Alicante, Spain — [3]Univ. of Rennes, CNRS, Inst. des Sciences Chimiques de Rennes (ISCR), France — [4]Eidgenössische Materialprüfungs- und Forschungsanstalt (EMPA), Swiss Federal Laboratories for Materials Science and Technology, Switzerland — [5]Inst. for Molecular Science, Research Center of Integrative Molecular Systems, Japan

We present recent results of spin-resolved photoemission studies [1] performed with layers of dithia-heptahelicene molecules [2]. A photoelectron spin polarization (SP) of more than $|P| = 30\%$ was measured at full monolayer (ML) coverage. The SP was furthermore measured at varying submonolayer coverages to probe the role of cooperative effects in its emergence. Indeed, a sharp, nonlinear increase of the SP was found at high surface coverages above 80% of a ML. We interpret this behavior not as a direct effect of intermolecular interactions on CISS, but argue that it is mediated through a phase transition in the molecular ordering within the ML. Similar measurements were performed with layers of helical tetrapyrrole complexes. [1] Möllers et al., *Isr. J. Chem.* **2022**, 62, e202200062 [2] Baciu et al., *Nanoscale Adv.* **2020**, 2, 1921 [3] Urban et al., *Chem. Eur. J.* **2023**, 29, e202300940

O 26.5 Tue 12:15 MA 141

Spin-dependent transmission and CISS effect in PdGa — •Mayra Peralta[1], Iñigo Robredo[1,2], Xia Wang[1], Maia Vergniory[1,2], and Claudia Felser[1] — [1]Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany — [2]Donostia International Physics Center, 20018 Donostia-San Sebastian, Spain

Chiral crystalline materials are an excellent framework to study Chiral Induced Spin Selectivity-related effects, since they can be grown and designed in a more controllable way and they are ordered systems where impurities and localization effects are minimised with respect to molecules. Besides, chiral crystals present topologically protected surface states and momentum locked spin states that open the way for using chiral couplings to control quantum information. In this talk I will present an analytical-computational model using a two-terminal setup and the Landauer formalism to compute spin-dependent transmission in chiral crystals, specifically in those of the group B20 as PdGa. Based on the results obtained with this model, I will evaluate the importance of chirality and spin orbit coupling in this material, to give rise to the spin-dependent transmission observed.

O 26.6 Tue 12:30 MA 141

Chiral-induced spin selective transmission (CISS) on naturally chiral surfaces — •Chetana Badala Viswanatha[1], Johannes Stöckl[1], Benito Arnoldi[1], Sebastian Becker[1], Ka Man

Yu[1], Martin Mitkov[1], Iulia Cojocariu[2], Vitaliy Feyer[2], Martin Aeschlimann[1], and Benjamin Stadtmüller[1] — [1]Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau, 67663 Kaiserslautern, Germany — [2]Peter Grünberg Institute (PGI-6), Forschungszentrum Jülich GmbH, 52428 Jülich, Germany

The chiral-induced spin selectivity (CISS) effect refers to the spin-selective electron transmission via chiral molecules. So far, CISS has been studied for helical molecules on noble metal surfaces. Here, we focus on CISS in molecules grown on a chiral Cu surface. Using spin- and momentum-resolved photoelectron spectroscopy, we show that the spin-dependent electron transmission through a point-chiral molecule on the chiral Cu(643) surface depends on all three components of the electron's spin [1]. Swapping the enantiomers alters the electrons' spin component parallel to the terraces of the chiral surface. This emphasizes the role of enantiomer-specific adsorption configurations on chiral surfaces. To understand the role of substrate chirality in CISS, we focus on the effect of a surface's chirality on the electronic properties of the adsorbed molecules. We use the highly symmetric pentacene on the Cu(643) surface as an example. Our momentum-resolved photoemission data show adsorption-induced changes in the orbital emission pattern that can be attributed to the chiral nature of the Cu surface. [1] J. Phys. Chem. Lett. 2022, 13, 26, 6244-6249.

Topical Talk O 26.7 Tue 12:45 MA 141

First-principles approaches to chiral induced spin selectivity — •Carmen Herrmann[1,2], Sumit Naskar[1], Ulrich Pototschnig[1], Aida Saghatchi[1], and Vladimiro Mujica[3] — [1]University of Hamburg, Department of Chemistry, HARBOR Bldg. 610, Luruper Chaussee 149, 22761 Hamburg, Germany — [2]The Hamburg Centre of Ultrafast Imaging, Hamburg, Germany — [3]School of Molecular Sciences, Arizona State University, Tempe, Arizona 85287-1604, USA

Exploring the spin degree of freedom offers fascinating options for nanoscale functionality, and also provides new experimental data for improving our insight into fundamental aspects of nonequilibrium physics at that scale. Chiral induced spin selectivity (CISS) is a particularly intriguing example of this, as it leads to spin preferences in electrons transported through chiral molecules, even though the molecules themselves are diamagnetic. Its underlying mechanism is still not understood [1]. We discuss recent progress in the first-principles description of CISS [2-6], such as the importance of exchange and the buildup of nonequilibrium spin in the junction. [1] F. Evers et al., Adv. Mater. 34, 2106629 (2022). [2] V. V. Maslyuk, R. Gutierrez, A. Dianat, V. Mujica, G. Cuniberti, J. Phys. Chem. Lett. 9, 5453 (2018). [3] Y. Liu, J. Xiao, J. Koo et al, Nat. Mater. 20, 638 (2021). [4] M. Zöllner, S. Varela, E. Medina, V. Mujica, C. Herrmann, J. Chem. Theory Comput., 16, 2914 (2020) [5] M. Zöllner, A. Saghatchi, V. Mujica, C. Herrmann, J. Chem. Theory Comput., 16, 7357 (2020) [6] S. Naskar, V. Mujica, C. Herrmann, J. Phys. Chem. Lett. 14, 694 (2023).

O 27: Tribology

Time: Tuesday 10:30–12:30 Location: MA 144

O 27.1 Tue 10:30 MA 144

Anisotropic Friction of Snake Scales Analyzed by Atomic Force Microscopy - From Fundamentals to Applications — Weibin Wu[1,2], K. M. Samaun Reza[1], Patrick Weiser[1], Cornelia Friederike Pichler[1], Richard Thelen[1], and •Hendrik Hölscher[1] — [1]Karlsruhe Institute of Technology, Kalsruhe, Germany — [2]now at: Institute of Noise and Vibration, Naval University of Engineering, Wuhan, P.R. China

As snakes are limbless, they have to locomote in direct contact with the ground. Therefore, the ventral scales of many snake species are equipped with nano-step structures to achieve anisotropic friction for efficient locomotion. Here, we present our study of this structural frictional anisotropy by atomic force microscopy showing that the frictional anisotropy correlates with the height of the nanosteps. The frictional anisotropy of the nano-stepped surface can be employed for the unidirectional transport of microscale particles through small random vibrations. Due to the frictional anisotropy, the micro-particles drift in the direction of lower friction. This feature can be employed for the dry self-cleaning of surfaces like photovoltaic modules in sunny but dry areas where soiling is an issue.

O 27.2 Tue 10:45 MA 144

Dynamic Friction Unraveled by Observing an Unexpected Intermediate State in Controlled Molecular Manipulation — Norio Okabayashi[1], Thomas Frederiksen[2,3], Alexander Liebig[4], and •Franz J. Giessibl[4] — [1]Graduate School of Natural Science and Technology, Kanazawa University, Ishikawa 920-1192, Japan — [2]Donostia International Physics Center (DIPC), San Sebastián 20018, Spain — [3]IKERBASQUE, Basque Foundation for Science, Bilbao 48013, Spain — [4]Institute of Experimental and Applied Physics, University of Regensburg, Regensburg D-93053, Germany

The pervasive phenomenon of friction has been studied at the nanoscale via a controlled manipulation of single atoms and molecules with a metallic tip, which enabled a precise determination of the static friction force necessary to initiate motion. However, little is known about the atomic dynamics during manipulation. Here, we reveal the complete manipulation process of a CO molecule on a Cu(110) surface at low temperatures using a combination of noncontact atomic force microscopy and density functional theory simulations. We found that an intermediate state, inaccessible for the far-tip position, is enabled in the reaction pathway for the close-tip position, which is crucial to understanding the manipulation process, including dynamic friction. Our results show how friction forces can be controlled and optimized, facilitating new fundamental insights for tribology [1].

[1] N. Okabayashi, Th. Frederiksen, A. Liebig, F.J. Giessibl, Phys. Rev. Lett. 131, 148001 (2023).

O 27.3 Tue 11:00 MA 144

Atomic Friction on 1T-TaS2 over a Phase Transition — •Yiming Song, Dirk Dietzel, and Andre Schirmeisen — Institute of Applied Physics (IAP), Justus-Liebig-Universität Gießen, 35392 Gießen, Germany

Friction force microscopy experiments were performed on 1T phase tantalum disulfide (1T-TaS2) surfaces as a function of temperature over the phase transition from nearly commensurate charge density wave (CDW) phase to commensurate CDW phase. While the superstructure can be revealed both in friction and in topography images, friction variation between these two phases is negligible, which is supported by detailed investigation on atomic stick-slip motion of the single asperity AFM tip sliding over 1T-TaS2. Besides, the load and velocity dependence of atomic friction on 1T-TaS2 surface in CCDW and NCCDW phases have been revealed. Using conductive atomic force microscopy, we were able to show the local electric conductivity on NCCDW surface with atomic resolution.

O 27.4 Tue 11:15 MA 144

Tribological properties of 2D materials — •Oscar Javier Gutiérrez Varela and José Guilherme Vilhena Albuquerque D Orey — Departamento de Física Teórica de la Materia Condensada, Facultad de Ciencias, Universidad Autónoma de Madrid, Madrid, España

2D materials are pivotal in diverse technological areas, necessitating a thorough examination of their physical properties in order to guarantee practical applications and to gain insights into their exhibited intriguing phenomena. We delved into the tribological properties of 2D materials, specifically graphene and MoS2. Using fully atomistic Molecular Dynamics (MD) simulations to mimic Friction Force Microscopy (FFM) experiments, we examine different tribological properties. Scenarios include suspended and substrate placements of monolayers or multiple layers, with analyses of pristine and defective 2D material presentations to discern different types of vacancies through FFM. We also explore various scanning directions and the impact of thermophoresis. Preliminary results confirm known characteristics, such as higher frictional forces in suspended graphene than on a substrate. Variations in lateral force patterns are observed in MoS2 with vacancies, hinting at the potential to distinguish defects using FFM. Additionally, scanning graphene and MoS2 against the direction of thermal flow increased hysteresis and lateral force, associated with higher friction. This comprehensive analysis allows for comparisons with experimental groups, contributing to a deeper understanding of the tribological properties of 2D materials.

O 27.5 Tue 11:30 MA 144

Friction Force Microscopy of Graphene on a Platinum Surface — •Thilo Glatzel[1], Zhao Liu[2], Guilherme Vilhena[3], Antoine Hinaut[1], Sebastian Scherb[4], Enrico Gnecco[5], and Ernst Meyer[1] — [1]University of Basel, Dep. of Physics, Klingelbergstr. 82, 4056 Basel, Switzerland — [2]Nankai University, School of Materials Science and Engineering, 300350 Tianjin, China — [3]Universidad Autónoma de Madrid, Dep. of Theoretical Condensed Matter Physics, 28049 Madrid, Spain — [4]Institute for Molecules and Materials, Radboud University, 6525 AJ Nijmegen, Netherlands — [5]M. Smoluchowksi Institute of Physics, Jagiellonian University in Krakow, 30-348 Krakow, Poland

Friction control and technological advancement are deeply connected. Two dimensional materials play a significant role in achieving near-frictionless contacts. However, there is a challenge in adjusting the sliding of superlubric materials. Taking inspiration from twistronics, we studied the control of superlubricity through moiré patterning. Through friction force microscopy and molecular dynamics simulations, we demonstrated that different twist angles of graphene moirés on a Pt(111) surface lead to a transition from superlubric to dissipative sliding regimes under various normal forces. This is due to a new mechanism at the superlattice level, where moiré tiles undergo a highly dissipative shear process connected to the twist angle beyond a critical load. Importantly, the atomic-level dissipation associated with moiré tile manipulation allows bridging different sliding regimes in a reversible manner, offering a way to subtly control superlubricity.

O 27.6 Tue 11:45 MA 144

Analyzing the tribological combination of microstructure and lubricant in beetle joints — •Cornelia Friederike Pichler[1], Richard Thelen[1], Matthias Mail[2], Thomas van de Kamp[3], and Hendrik Hölscher[1] — [1]Institute of Microstructure Technology, Karlsruhe Institute of Technology, Karlsruhe, Germany — [2]Institute of Nanotechnology, Karlsruhe Institute of Technology, Karlsruhe, Germany — [3]Institute for Photon Science and Synchrotron Radiation, Karlsruhe Institute of Technology, Karlsruhe, Germany

In order to gain inspiration for the development of environmentally friendly lubricants, we characterize the sophisticated friction-reducing system found in the joints of beetles. By combining microstructures with a lubricating substance, beetles optimize friction in their joints. We aim to research both the lubricant and the microstructure of the joints to gain inspiration for a degradable (and hopefully superior) alternative to mineral-oil-based lubricants. However, restrained by the tiny quantities of the beetle's lubricant and the compactness of their joints, the analysis is challenging. We, therefore, apply atomic force microscopy (AFM) to image topographical features of the joints and the frictional properties of the lubricating substance. Furthermore, we develop an artificial surface mimicking the beetle's microstructures and determine its frictional properties utilizing colloidal probes. Finally, we discovered the origin of the lubricant leaking through pores into the beetle's joint with focused ion beam (FIB) tomography.

O 27.7 Tue 12:00 MA 144

Superfluidity meets the solid-state: frictionless mass-transport through a (5,5) carbon-nanotube — •Alberto Ambrosetti, Pier Luigi Silvestrelli, and Luca Salasnich — Università degli Studi di Padova (Italy)

Superfluidity is a well-characterized quantum phenomenon which entails frictionless-motion of mesoscopic particles through a superfluid, such as ^{4}He or dilute atomic-gases at very low temperatures. As shown by Landau, the incompatibility between energy- and momentum-conservation, which ultimately stems from the spectrum of the elementary excitations of the superfluid, forbids quantum-scattering between the superfluid and the moving mesoscopic particle, below a critical speed-threshold. Here we predict that frictionless-motion can also occur in the absence of a standard superfluid, i.e. when a He atom travels through a narrow (5,5) carbon-nanotube (CNT). Due to the quasi-linear dispersion of the plasmon and phonon modes that could interact with He, the (5,5) CNT embodies a solid-state analog of the superfluid, thereby enabling straightforward transfer of Landau's criterion of superfluidity. As a result, Landau's equations acquire broader generality, and may be applicable to other nanoscale friction phenomena, whose description has been so far purely classical.

O 27.8 Tue 12:15 MA 144

Effect of Amorphous-Crystalline Phase Transition on Superlubric Sliding — Ebru Cihan[1,2], •Dirk Dietzel[1], Benedykt Jany[3], and Andre Schirmeisen[1] — [1]Institute of Applied Physics, Justus-Liebig-Universität Giessen, Germany — [2]Institute for Materials Science and Max Bergmann Center for Biomaterials, TU Dresden, Germany — [3]Marian Smoluchowski Institute of Physics, Jagiellonian University, Krakow, Poland

'Structural superlubricity' describes a state of ultralow friction, that can be attributed to the lattice mismatch between two incommensurate surfaces in sliding motion. It is commonly anticipated that structural superlubricity is most effective for crystalline surfaces, while the irregular structure of amorphous surfaces leads to less efficient lateral force cancellations and thus higher friction. To verify this fundamental assumption, we analyzed friction of antimony nanoparticles sliding on HOPG for temperatures between 300 and 750K[1] under UHV conditions. At about 420K a distinct and irreversible decrease in friction was observed. Based on complementary EBSD-analysis, this decrease was linked to a phase transition from amorphous to crystalline structure of the nanoparticles. More quantitatively, the results are described based on the Prandtl-Thomlinson model, where the relative changes of the effective energy barrier are correlated to a characteristic scaling factor γ that represents a measure for the crystalline state of the interface. γ is found to decrease by 20% and thereby also corroborates the influence of the phase transition on structural superlubricity.

[1] Cihan et al., PRL 130, 126205 (2023)

O 28: Solid-Liquid Interfaces II: Structure and Spectroscopy

Time: Tuesday 10:30–13:15 Location: H 1012

O 28.1 Tue 10:30 H 1012

Just How Anisotropic is the Air-Water Interface? An SFG/DFG Study — •Martin Thämer[1], Alexander Fellows[1], Álvaro Díaz Duque[1], Vasillios Balos[2], Louis Lehmann[3], Roland Netz[3], and Martin Wolf[1] — [1]Fritz-Haber-Institut der MPG, Berlin, Germany — [2]IMDEA Nanociencia, Madrid, Spain — [3]Freie Universität, Berlin. Germany

The air-water interface belongs to the most abundant interfaces on our planet and its unique properties are at the heart of numerous chemical processes in both nature and industrial applications. However, despite decades of research, some of the most fundamental aspects of the air-water interface still remain controversial, particularly the thickness of the interfacial region.

Here we directly address this question by employing our recently developed depth-resolved vibrational spectroscopy, which is based on the simultaneous measurement of phase-resolved sum- and difference-frequency signals and allows for precise depth-profiling on the sub-nm scale. Through isotopic exchange experiments the second-order spectra are separated into their resonant and non-resonant contributions, allowing the origins of each to be determined along with the length-scales of their decaying contributions. These results provide the first direct experimental measurement of the anisotropic layer thickness and yield important insight into the different signal contributions in second-order spectroscopy on water.

O 28.2 Tue 10:45 H 1012

Surface stratification determines the interfacial water structure of simple electrolyte solutions — •Yair Litman[1,2], Kuo-Yang Chiang[2], Takakazu Seki[2], Yuki Nagata[2], and Mischa Bonn[2] — [1]Yusuf Hamied Department of Chemistry, University of Cambridge, U. K. — [2]MPI for Polymer Research, Mainz, Germany.

For the past twenty years, the electrical-double layer (EDL) model has been the primary framework for analyzing how ions behave at the interface between water and air and interpreting various types of surface-selective experimental measurements. In this talk [1], I will present a combination of surface-sensitive heterodyne-detected vibrational sum frequency generation (VSFG) and *ab initio* based molecular dynamics simulations to study the liquid/air interface of several aqueous electrolyte solutions. Our VSFG measurements highlight the shortcomings of the EDL model for a range of electrolytes, including but not restricted to NaCl, NaBr NaI, NaOH, and CsF aqueous solutions. We rationalize our findings by proposing a new microscopic picture in which the surface of simple electrolyte solutions is stratified and consists of an ion-depleted outer surface and an ion-enriched subsurface layer, jointly determining the water interfacial structure.

[1] Y. Litman, K-Y. Chang, T. Seki, Y. Nagata, M. Bonn, submitted.

O 28.3 Tue 11:00 H 1012

Depth-Resolved SFG/DFG Spectroscopy of Charged Aqueous Interfaces — •Álvaro Díaz Duque, Alexander Fellows, Martin Wolf, and Martin Thämer — Fritz Haber Institut der MPG, Berlin, Germany

Charged aqueous interfaces are omnipresent in our world, being a crucial ingredient on both natural systems and technical devices. The charges at such interfaces generate electric fields which interact with the electrolyte and can extend over relatively large length scales. The evolution of the potential in such systems is typically described by the Gouy-Chapman-Stern (GCS) theory. However, much less in known about the depth-dependent structural properties of the main constituent of the electrolyte, the water.

Vibrational Sum-frequency generation spectroscopy is commonly used to study aqueous interfaces. The particular strength of this technique is its sensitivity to structural anisotropy and its ability to report on the molecular orientation and intermolecular environment of water. Here, we utilize our recent development that combines phase-resolved SFG and DFG which additionally yields depth resolution on the nm scale. We study the details of the depth-dependent water structure within the electric double layer and correlate these to the defining thermodynamic and electrostatic quantities in the investigated electrolyte. Based on the results we demonstrate that the continuum solvent model that is typically assumed within the GCS theory is clearly insufficient to accurately describe water in such systems.

O 28.4 Tue 11:15 H 1012

Exploring Dynamic Solvation Effects at the Electrochemical IrO_2/Water Interface — •Nikhil Bapat, Hendrik H. Heenen, and Karsten Reuter — Fritz-Haber-Institut der MPG, Berlin

Upon electrification, solid-liquid interfaces are known to undergo significant alterations, not least due to the potential-induced adsorption of surface species. Computational studies conveniently explore this phenomenon via an *ab initio* thermodynamics based Pourbaix analysis, which compares the potential-dependent stability of multiple structural and compositional configurations. To render the computational cost tractable, solvation effects are thereby presently if at all mostly only implicitly treated.

Here, we assess this approximation for the highly hydrophilic $IrO_2(110)$ oxide catalyst surface by comparing to results from extensive molecular dynamics simulations including explicit water. This approach is computationally enabled at first-principles predictive-quality level by the use of efficient and accurate machine-learning potentials that are trained to density functional-theory data via an active learning approach. The detailed comparison to the analog Pourbaix diagram obtained with implicit solvation sheds light on the effect of dynamic H-bonding networks at the complex, corrugated oxide surface, while the analysis of the potential-dependent interfacial water structure and mobility gives insight to which degree these networks may be mimicked by static ice overlayers.

O 28.5 Tue 11:30 H 1012

Molecular Dynamics Modeling of Pulsed Laser Fragmentation of Solid and Porous Si Nanoparticles in Liquid Media — •Dmitry Ivanov and Martin Garcia — University of Kassel, Kassel, Germany

The production of non-toxic and homogeneous colloidal solutions of nanoparticles (NPs) for biomedical applications is of extreme importance nowadays. Among the various methods for generation of NPs, pulsed laser ablation in liquids (PLAL) has proven itself as a powerful and efficient tool in biomedical fields, allowing chemically pure silicon nanoparticles to be obtained. Due to a number of complex and interrelated processes involved in the laser ablation phenomenon, however, the final characteristics of the resulting particles are difficult to control, and the obtained colloidal solutions frequently have broad and multimodal size distribution. Therefore, the subsequent fragmentation of the obtained NPs in the colloidal solutions due to pulsed laser irradiation can be utilized. The resulting NPs characteristics, however, depend on the parameters of laser irradiation as well as. Thus, reliable knowledge of the mechanism of NP fragmentation is necessary for generation of a colloidal solution with NPs of predesigned properties. To investigate the mechanism of a laser-assisted NP fragmentation process, in this work, we perform a large-scale molecular dynamics (MD) modeling of FS laser interaction with colloidal solution of Si NPs. The corresponding conclusion about the relative input of the properties of different laser-induced processes and materials to the mechanism of NP generation is drawn.

O 28.6 Tue 11:45 H 1012

Visualizing solvent structures at electrified solid-liquid interfaces by electrochemical atomic force microscopy — •Andrea Auer[1,2], Thorben Eggert[3], Nicolas G. Hörmann[3], Karsten Reuter[3], and Franz J. Giessibl[1] — [1]Institute of Experimental and Applied Physics, University of Regensburg, Germany — [2]Institute of Physical Chemistry, University of Innsbruck, Austria — [3]Fritz-Haber-Institut der Max-Planck-Gesellesschaft, Berlin, Germany

To design efficient electrochemical interfaces for sustainable energy conversion, it is crucial to resolve the molecular structures of the electric double layer, where inevitably all electrocatalytic reactions take place. Here, we employ electrochemical atomic force microscopy (AFM) with stiff qPlus sensors[1,2], which allows us to probe the potential-dependent, vertical solvent layering at well-defined electrified solid-liquid interfaces with high spatial resolution. Investigations of a Au(111) electrode in various aqueous electrolytes reveal distinct oscillatory frequency shift modulations in z-direction. Depending on the applied potential, the charge of the electrode and the type of ions, we attribute these oscillations to water and/or ion layering close to the electrode, which we can correlate with atomistic molecular dynamics simulations.

[1] F.J. Giessibl, Rev. Sci. Instrum. 90, 011101 (2019). [2] A. Auer, B. Eder and F.J. Giessibl, J. Chem. Phys. 159, 174201 (2023).

O 28.7 Tue 12:00 H 1012

A Comparative Study of Classical Force Fields and the MB-Pol Water Model in Understanding Water/Metal Interfaces — •Saeyeon Lee, Yevhen Horbatenko, Seungchang Han, and Stefan Ringe — Department of Chemistry, Korea University

Metal/water interfaces hold profound significance in the material or energy science fields. They form pivotal junctures where the behavior of metals and their interactions with aqueous environments give reference to various industrial, technological, and environmental applications. At these interfaces, water-water interactions are expected to be very distinctive compared to the bulk, raising a question about the validity of classical force fields. In this study, we attempt to follow this question and compare classical force fields for the description of interfacial water with a high-level reference, the MB-Pol water model. This model is based on the many-body expansion of the water interaction energy based on coupled-cluster calculations. We use this model and a simplified description of water-metal interactions to study the impact of the water force field on water-metal interactions and present a detailed analysis and justification for using many-body force fields.

O 28.8 Tue 12:15 H 1012

Atomistic modeling of the electric double layer at water/Ag(111) interfaces — •Sung Sakong and Axel Gross — Institut für Theoretische Chemie, Universität Ulm, 89081 Ulm, Germany

The structures and processes at the electrode/electrolyte interface are crucial components of electrochemical cells play a critical role in energy conversion and storage. Recently, the subject has attracted significant interest from a first-principles perspective. Despite their significance, no conclusive consensus exists on how to include the electrochemical environment in atomistic simulations. Here, we address the structure and properties of electrochemical interfaces from an atomistic perspective based on ab initio molecular dynamics simulations in the presence of ionic species. We use the water/Ag(111) interface as an example and explore how to take the electrode potential into account properly in first-principles simulations. In particular, we discuss whether local charges, e.g., surface charges, can correspond to physical observables. Finally, we address the driving force for forming electric double layers at electrochemical interfaces.

[1] A. Groß and S. Sakong, Chem. Rev. **122** , 10746 (2022).

[2] A. Groß and S. Sakong, Curr. Opinion Electrochem. **14**, 1 (2019).

O 28.9 Tue 12:30 H 1012

Unraveling electrocatalysts and electrochemical interfaces in situ at the nanoscale — •Christopher Kley — Helmholtz Young Investigator Group Nanoscale Operando CO2 Photo-Electrocatalysis, Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, 14109 Berlin, Germany — Department of Interface Science, Fritz Haber Institute of the Max Planck Society, 14195 Berlin, Germany

Recent advances in electrochemical atomic force microscopy will be presented, with a focus on deciphering the local properties of electrocatalysts under liquid phase reaction conditions. Reactions include the electroreduction of CO2 (CO2RR) and CO (COR), as well as the oxygen evolution reaction (OER). Selected results provide insights into (i) the complex surface structures of Cu electrodes during CO2RR and COR in distinct electrolytes, (ii) the electrical conductivity, chemical-frictional, and morphological properties of electrocatalysts, and (iii) the in situ surface structure and catalytic activity of OER catalysts. Complementary spectroscopic approaches and findings will be discussed and a perspective given on the potential of scanning probe microscopy for nanoscale catalysis and rational innovation of energy conversion materials.

O 28.10 Tue 12:45 H 1012

Electrokinetic Measurement: A Tool for Characterization of Piezoelectric Materials in liquid media — •Mohsen Sadeqi-Moqadam and Julia Glaum — Department of Materials Science and Engineering, Norwegian University of Science and Technology

Piezoelectric materials have pervasive applications across industries like telecommunications, automotive, electronics, and aerospace. Recently, their use in liquid environments for biomedical engineering, electrochemistry, and electromechanical systems has surged, prompting inquiries into their behavior and performance in such conditions. To probe this, an innovative experimental setup was devised, employing electrokinetic measurements to characterize polarized piezoelectric materials like lead zirconate titanate under mechanical stress (up to 150 MPa) in liquid media. This setup revealed a direct link between applied stress and the piezoelectric sample's electrokinetic response. Dynamic stress variations closely mirrored changes in the sample's electrokinetic behavior and affected the flow cell's resistance. Conversely, the non-piezoelectric alumina sample showed negligible alterations in both electrokinetics and resistance during stress application. Finite element modeling further supported these experimental findings, showcasing consistent outcomes for the electrical processes induced by mechanical stress variations on piezoelectric samples. This exploration sheds light on understanding and predicting piezoelectric material responses within liquid environments, impacting diverse fields reliant on their functionality.

O 28.11 Tue 13:00 H 1012

Efficient and Accurate Description of Solid-Liquid Interface Dynamics Using High-Dimensional Neural Network Potentials — •Knut Nikolas Lausch[1,2], Marco Eckhoff[3], Peter Blöchl[4], and Jörg Behler[1,2] — [1]Theoretische Chemie II, Ruhr-Universität Bochum, Germany — [2]Research Center Chemical Sciences and Sustainability, Research Alliance Ruhr, Germany — [3]Theoretische Chemie, Eidgenössische Technische Hochschule Zürich, Switzerland — [4]Institut für Theoretische Physik, Technische Universität Clausthal, Germany

Solid-liquid interfaces play a central role in many processes that are of utmost importance for a sustainable energy future. From energy storage solutions to heterogeneous catalysis, understanding interface dynamics and reactivity plays a key role in developing new materials. Molecular dynamics simulations of such interfaces rely on an accurate description of solid-liquid interactions, and density functional theory (DFT) can in principle provide reliable results. However, due to the high computational costs, time and length scales of ab initio simulations are severely limited. This can be overcome by employing machine learning potentials, which yield energies and forces several orders of magnitude faster while retaining chemical accuracy. Here, we present a high-dimensional neural network potential for lithium manganese oxide (LMO) in water as a model system for the electrocatalytic oxygen evolution reaction.

O 29: Poster: 2D Materials

Time: Tuesday 12:30–14:30 Location: Poster A

O 29.1 Tue 12:30 Poster A

Precision in Growth: Knudsen Cells and Solution-Coated Precursors Revolutionize TMDC Synthesis by CVD — •Thomas Aquinas Pobi, Osamah Kharsah, Jonah von Kuczkowski, and Marika Schleberger — Universität Duisburg-Essen, Fakultät für Physik und CENIDE, Duisburg,Germany

Chemical Vapor Deposition (CVD) stands out as a reliable technique for synthesizing transition metal dichalcogenides (TMDCs) like MoS2 and WS2 . However, the process's susceptibility to minor variations in growth parameters often hampers reproducibility. This study introduces a novel approach utilizing a combination of Knudsen cells and a solution-based, spin-coated precursor to address this challenge. By ensuring a constant and sustainable chalcone supply, this method not only overcomes reproducibility issues but also enhances the spatial distribution of the precursor. The results demonstrate a successful transition from diffusion-limited growth to attachment-limited growth, marking a significant advancement in TMDC synthesis.

O 29.2 Tue 12:30 Poster A

h-BN in the Making: The Surface Chemistry of Borazine on Rh(111) — •Eva Marie Freiberger[1], Fabian Düll[1], Phiona Bachmann[1], Johann Steinhauer[1], Hans-Peter Steinrück[1], and Christian Papp[1,2] — [1]FAU Erlangen-Nürnberg, Erlangen, Germany — [2]FU Berlin, Berlin, Germany

The intriguing properties of two-dimensional materials (2DM) gave rise to a vivid research field. One of the most-studied representatives of this class is hexagonal boron nitride (h-BN). In ultra-high vacuum (UHV), h-BN can be grown on a metal support via chemical vapor deposition (CVD) using suitable precursors, such as ammonia borane or borazine. h-BN grown on a lattice-mismatched substrate undergoes a buckling, which is, in the case of Rh(111), referred to as nanomesh.

In this UHV study, we investigated the evolution of the h-BN nanomesh from borazine on Rh(111) using synchrotron radiation-based high-resolution X-ray photoelectron spectroscopy (XPS). The adsorption of borazine at 130 K was followed in situ, showing that borazine adsorbs mainly as an intact molecule. By studying borazine on Rh(111) with NEXAFS, we were able to identify a flat-lying adsorption geometry. For a better understanding regarding the formation of h-BN, the thermally-induced reaction of borazine on Rh(111) was investigated by temperature-programmed XPS. We find that borazine multilayers already desorb below 200 K. At 300 K, dehydrogenation of the remaining monolayer occurs, and boron nitride starts to grow in a disordered fashion. Above 600 K, the formation of the nanomesh sets in and, at about 1100 K, the conversion to h-BN is complete.

O 29.3 Tue 12:30 Poster A

2D porous fantrip network on calcite (10.4) as a starting point for on-surface synthesis — •Lea Klausfering[1], Lukas Höltkemeier[1], Ralf Bechstein[1], Markus Lackinger[2], and Angelika Kühnle[1] — [1]Physical Chemistry I, Bielefeld University, 33615 Bielefeld, Germany — [2]Deutsches Museum, 80538 Munich, Germany

Triptycen anthracene derivatives have proven to be suitable molecules for two-dimensional polymers. For example the deposition on crystal surfaces under ultra-high vacuum (UHV) conditions is possible. Under UHV conditions, the fluorinated anthracene triptycene (fantrip) molecule was investigated on graphite (0001) [1] and Au (111) [2] surfaces. However, in both cases, a porous Fantrip network could only be obtained when passivating the surface prior to molecule deposition. Otherwise, the molecules did not lie perpendicular with all three anthracene blades with respect to the surface as desired, but parallel with two anthracene blades and upright with one anthracene blade. The passivation reduces the molecule-surface interaction. Here, we show that the porous fantrip network can be achieved without surface passivation when using the (10.4) surface of the bulk insulator calcite. The resulting network provides a good starting point for [4+4]-photocycloaddition on the surface to generate a two-dimensional polymer.

[1] L. Grossmann et al., Nature Chemistry, 2021, 13, 730-736.

[2] L. Grossmann et al., Angew. Chem. Int. Ed., 2022, 134, e202201044.

O 29.4 Tue 12:30 Poster A

Growth of two-dimensional hexagonal β-GeSe on Au(111) — •Dina Wilks, Veronika Blecker, Marina Hammer, Muhammad Ali Martuza, Paulus Aleksa, and Carsten Busse — Department Physik, Universität Siegen, Walter-Flex-Straße 3, 57072 Siegen

Two-dimensional group-IV monochalcogenides (general form MX with M=Sn, Ge; X=S, Se, Te) have great potential in nanotechnology due to their versatility. Theory predicts several polymorphs of these materials. For GeSe none of these have been confirmed experimentally, unlike SnSe, SnS, and SnTe.
We report the first experimental realization of β-GeSe which has a corrugated honeycomb structure. The monolayers are prepared under highly controlled conditions (molecular beam epitaxy from GeSe powder on a single crystalline Au(111) substrate under ultra-high vacuum (UHV) conditions). We find a temperature window for self-limiting monolayer growth. Low-energy electron diffraction (LEED) reveals a (5×5) superstructure with respect to Au(111). For sub-monolayer coverage, we find orientational disorder while the full layer is highly aligned with respect to the sample. Scanning tunneling microscopy (STM) shows that a superstructure unit cell contains (4×4) β-GeSe unit cells. The Au(111) herringbone reconstruction is lifted under the MX-monolayer.

O 29.5 Tue 12:30 Poster A

Facile chemical vapor deposition growth of transition metal dichalcogenide alloy lateral heterostructures for electronic and optoelectronic applications — Seung Heon Han[1], Gia Quyet Ngo[2], Moritz Quincke[3], Emad Najafidehaghani[1], •Christof Neumann[1], Uwe Hübner[4], Ute Kaiser[3], Falk Eilenberger[2,5], Antony George[1], and Andrey Turchanin[1] — [1]Institute of Physical Chemistry, Friedrich Schiller University Jena, 07743 Jena — [2]Institute of Applied Physics, Abbe Center of Photonics, Friedrich Schiller University Jena, 07745 Jena — [3]Central Facility of Electron Microscopy, Ulm University, 89081 Ulm — [4]Leibniz Institute of Photonic Technology (IPHT), 07745 Jena — [5]Fraunhofer-Institute for Applied Optics and Precision Engineering IOF, 07745 Jena

Lateral heterostructures (LH) of monolayer transition metal dichalcogenides (TMDs) such as MoS_2, WSe_2 are known for their high potential for the integration in ultrathin device technology as p-n junction diodes and photodetectors. While the properties of pure TMDs are noteworthy, the development of alloy TMDs (e.g., $Mo_xW_{(1-x)}S_2$ etc.) adds a new dimension to the research on 2D semiconductors. Alloy TMDs allow for the fine-tuning of their intrinsic electronic properties, thereby expanding the spectrum of potential applications. Here we present the growth of monolayer alloy LHs by CVD employing liquid precursors of transition metals. We characterized the synthesized material by AFM, Raman spectroscopy, HRTEM, as well as, PL spectroscopy and studied their performance in optoelectronic devices.

O 29.6 Tue 12:30 Poster A

Modifications of the Au(111) reconstruction induced by the adsorption of P_2 and its role for the growth of blue-phosphorus. — •Morris Mühlpointner and Moritz Sokolowski — Clausius Institute for Physical and Theoretical Chemistry of the University of Bonn, Wegelerstr. 12, 53115, Bonn Germany

The adsorption of phosphorus on the Au(111) surface has been studied under the aim to grow a two-dimensional “blue-phosphorus” (bP) structure, which is analogous to graphene. Furthermore, phosphorus adsorption has been shown to alter the structure of the Au(111) reconstruction from the herringbone reconstruction to a “trigon reconstruction” (TR) [1].

In the present work, we investigated the structure of the TR for increasing bP coverages using high-resolution spot-profile analyzing low-energy electron diffraction (SPA-LEED). We observed the TR prior to bP film growth with a highly-ordered structure. At higher P-coverages, patches of the bP structure and the TR coexist. With time, the structure of the TR distorts and forms disordered patches of Au(111) surface that exhibit low P-coverages. Further insight was obtained by temperature programmed desorption (TPD) spectra, which show a zero-order desorption peak, regardless of the starting coverage. We propose that this zero-order desorption peak can be explained by a phase-equilibrium.
[1] ACS Nano **14**, 3687-3695, 2020 .

This work was supported by the DFG through the research training group 2591.

O 29.7 Tue 12:30 Poster A

Electronic Fingerprints of Platinum Surface Tellurides and Pt_xTe_y Films on Pt(111) — •Isabella Stollberg, Andreas Raabgrund, and M. Alexander Schneider — Universität Erlangen-Nürnberg, 91058 Erlangen, Germany

For the synthesis of transition metal dichalcogenides, in general two approaches can be utilized: evaporating the metal and the chalcogenide on a suitable substrate [1] or directly depositing only the chalcogenide on a desired metal substrate. We investigated by STM, STS, and DFT the properties of Pt_xTe_y films on Pt(111) that are synthesized according to the latter approach. Depending on Te content, a (3×3) and (10×10) surface telluride structure are found, followed by the growth of Pt_2Te_2 and $PtTe_2$ [2].

Applying STS measurements we found characteristic features on the respective structures which can be used as fingerprints for identification. By comparison with DFT calculations we attribute the STS features to electronic states within the telluride films. Additionally, we determined the work function differences probing field emission resonances. We find that the (10×10) surface telluride has the lowest but Pt_2Te_2 and $PtTe_2$ have very similar work functions.

[1] K. Lasek et al., Nano Lett. **22**, 23 (2022)

[2] T. Kißlinger et al., Phys. Rev. B **108**, 205412 (2023)

O 29.8 Tue 12:30 Poster A

Properties and modification of antimonene on Ag(111) — •Felix Otto, Leon Bojunga, Christian Haberland, Jonas Brandhoff, Maximilian Schaal, Marco Gruenewald, and Torsten Fritz — Institute of Solid State Physics, Friedrich Schiller University Jena, Helmholtzweg 5, 07743 Jena, Germany

The search for novel two-dimensional (2D) materials with beneficial structural, electronic or optical properties is of great interest for many researchers as well as, to an increasing extend, for industry. For this reason, we investigate the properties of antimonene on Ag(111) with respect to the adsorption of organic molecules as well as atoms.

These were in a first part investigated by using the organic molecule 3,4,9,10-perylene-tetracarboxylic-dianhydride (PTCDA), which has well-known properties and serves as a sensor for the decoupling properties of the Sb-films. For this, differential reflectance spectroscopy (DRS) together with photoelectron spectroscopy (PES) was performed. This yields a classification of the PTCDA-Sb interaction in comparison to other substrates.

In a second part we modified the Sb on Ag(111) system by the deposition of phosphorous in order to grow an inorganic 2D heterolayers. However, we will show by low-energy electron diffraction (LEED) and scanning tunneling microscopy (STM) measurements that phosphorous did not form ordered films on top of Sb. Instead, our structural and electronic characterization revealed that P is able to form an ordered intermediate layer below α-antimonene.

O 29.9 Tue 12:30 Poster A

Epitaxial growth and properties of sub-monolayer to multilayer $FeBr_2$ on Au(111) — •S. E. Hadjadj[1], C. González-Orellana[2], J. Lawrence[3], D. Bikaljević[4,5], M. Peña-Díaz[2], P. Gargiani[6], L. Aballe[6], J. Naumann[7], M. Ángel Niño[6], M. Foerster[6], S. Ruiz-Gómez[8], S. Thakur[1], I. Kumberg[1], J. Taylor[9,10], J. Hayes[1], J. Torres[1], C. Luo[9,10], F. Radu[9], D. G. de Oteyza[3,11], W. Kuch[1], J. I. Pascual[4,12], C. Rogero[2,3], and M. Ilyn[2] — [1]Freie Universität Berlin, Experimentalphysik, Germany — [2]Centro de Física de Materiales, Donostia, Spain — [3]Donostia International Physics Center, Spain — [4]CIC nanoGUNE-BRTA, Donostia, Spain — [5]Institute of Physical Chemistry, University of Innsbruck, Austria — [6]ALBA Synchrotron Light Source, Spain — [7]Freie Universität Berlin, Dahlem Center for Complex Quantum Systems, Germany — [8]Max Planck Institute for Chemical Physics of Solids, Dresden, Germany — [9]HZB, Germany — [10]Physik, Technische Universität München, Germany — [11]Nanomaterials and Nanotechnology Research Center (CINN), Oviedo, Spain — [12]Ikerbasque, Basque Foundation for Science, Bilbao, Spain

We characterized the properties of the van-der-Waals material $FeBr_2$ on Au(111). The investigation via STM and LEED reveals a dominant superstructure for the first layer, which changes for the second layer. The characterization by XPS and XAS shows that the chemical composition does not change for different coverages. The magnetic characterization by XMCD reveals that the magnetization at 6 T increases during the transition from first to the second monolayer.

O 29.10 Tue 12:30 Poster A

The Effect of WS_2-Cu_2O for the Optimization of a WS_2-based LED — •Jonah von Kuczkowski[1], Osamah Kharsah[1], Leon Daniel[1], Stephan Sleziona[1], Denys Vidish[2], Kevin Musselman[2], and Marika Schleberger[1] — [1]Universität Duisburg-Essen, Fakultät für Physik und CENIDE, Duisburg, Germany — [2]University of Waterloo, Mechanical and Mechatronics Engineering, Waterloo, Canada

Scalable 2D material based LEDs have been the subject of many recent works. Tungsten disulfide (WS_2) emerges as a potential active material in such LEDs, based on its direct band gap, high stability and strong photoluminescence. Copper oxide (Cu_2O) on the other hand, a p-type semiconductor, can function as the electron-transport layer. This study investigates the interaction between WS_2 and Cu_2O within the framework of the LED. Different characterization techniques, including (low-temperature) PL, Raman spectroscopy for optoelectronic interaction and various electrical transport measurements are employed to map out the benefits and limitations of using Cu_2O in the WS_2 based LED.

O 29.11 Tue 12:30 Poster A

Photoluminescence upconversion in monolayer WSe_2 activated by plasmonic cavities through resonant excitation of dark excitons — •Niclas S. Mueller[1,2], Rakesh Arul[1], Gyeongwon Kang[1], Ashley P. Saunders[3], Amalya C. Johnson[3], Ana Sánchez-Iglesias[4], Shu Hu[1], Lukas A. Jakob[1], Jonathan Bar-David[1], Bart de Nijs[1], Luis M. Liz-Marzán[4], Fang Liu[3], and Jeremy J. Baumberg[1] — [1]NanoPhotonics Centre, University of Cambridge, UK — [2]Present address: Fritz Haber Institute of the Max Planck Society Berlin, Germany — [3]Department of Chemistry, Stanford University, USA — [4]CIC biomaGUNE, Donostia-San Sebastián, Spain

Anti-Stokes photoluminescence (PL) is light emission at a higher photon energy than the excitation, with applications in optical cooling, lasing, and the detection of infrared light. Here, we show how plasmonic nano-cavities activate anti-Stokes PL in WSe_2 monolayers through resonant excitation of a dark exciton at room temperature. The optical near-fields of the plasmonic cavities excite the out-of-plane transition dipole of the dark exciton, leading to light emission from the bright exciton at higher energy. Through statistical measurements on hundreds of plasmonic cavities, we show that coupling to the dark exciton leads to a near hundred-fold enhancement of the upconverted PL intensity. We further demonstrate a selective and reversible switching of the upconverted PL via electrochemical gating.

[1] Mueller et al. Nat. Commun. 14, 5726 (2023)

O 29.12 Tue 12:30 Poster A

Characterization of single-layer NbSe2 — •Caroline Firschke[1], Verena Caspari[1], Jean-Maxime Schlachter[2,3], Matthieu Jamet[2,3], Clemens B. Winkelmann[2,3], and Katharina J. Franke[1] — [1]Freie Universität Berlin, Germany — [2]Université Grenoble Alpes, France — [3]CEA Grenoble, France

A single layer of the superconductor NbSe2 exhibits different properties than its bulk counterpart due to its reduced dimensionality. For example, strong spin-orbit coupling combined with the lack of inversion symmetry locks electrons with opposite spin and momenta, resulting in an increased in-plane critical field.

Here, we use scanning tunneling microscopy (STM) at 1.1K to characterize single-layer NbSe2-islands on a bi-layer graphene on SiC, grown by molecular beam epitaxy. These samples had been capped with Se for transport from the growth chamber to the STM, which we remove by heating under ultra-high vacuum conditions. Although this procedure does not lead to atomically clean layers, we detect a superconducting gap and low-energy excitations outside of it, interpreted as Legget modes that are in agreement with the work by W. Wan et al. [1].

[1] Wen Wan et al. "Observation of Superconducting Collective Modes from Competing Pairing Instabilities in Single-Layer NbSe2". Adv. Mat. 34, 2206078 (2022)

O 29.13 Tue 12:30 Poster A

Machine learning angle resolved photoemission spectroscopy spectrum of transition metal dichalchogenide materials — •Ridha Eddhib and Jan Minar — New Technologies - Research Centre, University of West Bohemia, 301 00 Pilsen, Czech Republic.

Predicting material properties through machine learning necessitates the development of a specific descriptor, serving as a distinctive fingerprint for the desired properties. This descriptor becomes the input for the machine learning model, typically crafted by leveraging atomic structures as the primary source of information. Fortunately, there already exists a comprehensive repository of descriptors for encoding atomic structures. Our research focuses on the challenge of representing the mapped quantum states angularly resolved for photoemitted electrons in a solid, specifically targeting machine learning applications for state-specific electronic properties. In this context, we introduce a descriptor for Angle-Resolved Photoemission Spectroscopy (ARPES) based on the energy decomposed operator matrix elements (ENDOME) fingerprints[1]. Notably, our work sheds light on a significant limitation present in various Density Functional Theory (DFT)codes,they commonly lack the capability to directly compute ARPES spectra. Our efforts not only address this critical gap but also stand as a pioneering effort to pave the way for accurate estimates of ARPES spectra contributing significantly to the advancement of machine learning methodologies applied to materials science.[1] Knøsgaard, N. R., & Thygesen, K. S. (2022). Nature Communications, 13(1),468.

O 29.14 Tue 12:30 Poster A

Floquet Engineering in TMDs using short pulsed lasers — •Alejandro Sebastián Gómez[1], Yuriko Baba[2], Rafael A. Molina[1], and Francisco Domínguez-Adame[3] — [1]Instituto de Estructura de la Materia, IEM-CSIC, Madrid, Spain — [2]Universidad Autónoma de Madrid, Madrid, Spain — [3]GISC, Universidad Complutense de Madrid, Madrid, Spain

This work focuses on the use of pulsed lasers to manipulate certain properties of Transition Metal Dichalcogenides (TMDs), a type of two-dimensional material [1] . For this purpose, the well-known method of Floquet engineering is employed, capable of describing quantum systems with a perfectly periodic driven external field. The aim is to understand how effectively Floquet theory can be applied to non-periodic pulses such as those in pulsed lasers. To achieve this, the we employ the t-t' formalism technique [2], offering an interesting method for analyzing short pulses.

[1] Liu, G. B., Xiao, D., Yao, Y., Xu, X., & Yao, W. (2015). Electronic structures and theoretical modelling of two-dimensional group-VIB transition metal dichalcogenides. Chemical Society Reviews, 44(9), 2643-2663. [2] Ikeda, T. N., Tanaka, S., & Kayanuma, Y. (2022). Floquet-Landau-Zener interferometry: Usefulness of the Floquet theory in pulse-laser-driven systems. Physical Review Research, 4(3), 033075.

O 29.15 Tue 12:30 Poster A

Local Work Function Modulation of Hexagonal Boron Nitride on Pt(110) — •Marco Thaler, Matthias Zeilerbauer, and Laerte Patera — University of Innsbruck, Austria

Hexagonal boron nitride (h-BN) offers a wide band gap, high thermal stability and low chemical reactivity and therefore has emerged as a promising candidate to serve as an ultra thin layer for the deposition and decoupling of organic molecules on surfaces. Remarkably, on the incommensurate Pt(110) surface, h-BN can be grown as a large-area single-domain monolayer of excellent quality. The lattice mismatch between the hexagonal adlayer and the platinum substrate leads to the formation of a distinctive moiré pattern. Using field emission resonance spectroscopy (FERS), a spatial modulation of the local work function can be observed depending on which moiré region is examined. Upon room temperature deposition of 5,10,15,20-Tetrakis(4-aminophenyl)porphyrin (Co-TAPP), the porphyrins arrange into clusters occupying a variety of adsorption sites of the h-BN/Pt(110) unit cell. With the help of scanning tunneling spectroscopy a site-specific shift of the Co-TAPP states is observed, which can be associated to the local work function differences of the underlying substrate. This shows that Co-TAPP can act as an effective probe to study the work function landscape of our system.

O 29.16 Tue 12:30 Poster A

Can we disentangle the electronic contribution from the surface corrugation of the charge density wave in 2H-NbSe2? — •Nikhil Seeja Sivakumar[1], Joost Aretz[1], Sebastian Scherb[1], Marion van Midden Mavric[2], Nora Huijgen[1], Umut Kamber[3], Daniel Wegner[1], Alexander Ako Khajetoorians[1], Malte Rösner[1], and Nadine Hauptmann[1] — [1]Institute of Molecules and Materials, Radboud University, Nijmegen, The Netherlands — [2]Condensed matter physics department, Jozef Stefan Institute, Ljubljana, Slovenia — [3]Department of Physics, Princeton University, USA

A charge density wave (CDW) is a collective phenomenon present in many quantum materials. Currently, the formation mechanism of the CDW in 2H-NbSe2 is understood to originate from momentum and orbital dependent electron phonon coupling. Still, there are open questions regarding the involved bands and the role of Se atoms in the formation of the CDW. Here we utilize combined Scanning Tunnelling Microscopy (STM) and non-contact Atomic Force Microscopy (nc-AFM) to independently characterize the geometric as well as the electronic structure of the CDW in 2H-NbSe2. Distance-dependent current and frequency shift images reveal a different evolution of the relative contrast of the atomic and CDW periodicities. We utilize Scanning Tunnelling Spectroscopy to characterize the electronic structure close to the Fermi level. We compare the experimental results with ab-initio calculations of the energy and momentum-resolved charge density.

O 29.17 Tue 12:30 Poster A

Exploring Excitonic Dispersions in 2D WS_2 through Spatially Resolved Electron Energy Loss Spectroscopy — •Max Bergmann, Jürgen Belz, Oliver Massmeyer, Badrosadat Ojaghi Dogahe, Robin Günkel, Johannes Glowatzki, Andreas Beyer, Stefan Wippermann, and Kerstin Volz — Material Sciences Center and Department of Physics, Philipps-Universität Marburg, Germany

Two-dimensional transition metal dichalcogenides have attracted considerable attention in materials science, particularly in the field of valleytronics, due to their unique indirect-to-direct bandgap transition and enhanced quantum efficiency in the monolayer limit. In this study we use a monochromated scanning transmission electron microscope to measure the electron energy loss spectrum of WS_2 providing nanometer-scale spatial resolution. The measurements reveal redshifts of the excitonic excitations due to a change in the WS_2 layer number for both the excitons residing at the K-point of the band structure and excitonic features stemming from more diverse excitations at points between Γ and K. To assess the origin of these excitations, theoretical investigations, namely density functional theory with Bethe-Salpeter equations on top, were used. This comprehensive investigation provides valuable insights into the excitonic properties of WS_2 across different layer configurations, highlighting in particular the dispersion of excitons with respect to changes in layer number.

O 29.18 Tue 12:30 Poster A

Electronic properties of a magnetic semiconductor: V-doped WSe_2 — •Jana Kähler[1,2], Sinja H. Weychardt[1,2], Florian K. Diekmann[1,2], Matthias Kalläne[1,2], and Kai Rossnagel[1,2] — [1]Ruprecht Haensel Laboratory, Deutsches Elektronen-Synchrotron DESY, 22607 Hamburg, Germany — [2]Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, 24098 Kiel, Germany

Spintronics is proposed as an energy efficient alternative to conventional electronics with a wide range of applications, e.g., in the field of quantum computing. To realize efficient spin transport properties, magnetic semiconductors with a Curie temperature around room temperature and the ability to switch the magnetic order by electrical gating are promising candidates. The vanadium-doped layered transition metal dichalcogenide $2H$-WSe_2, which is characterized by strong spin-orbit coupling, a homogeneous magnetic doping distribution up to doping levels of a few percent, and the prediction of gate tunability, is a very promising candidate. However, many open questions remain regarding the electronic functionality.

Here, we present first results on the electronic transport properties and the momentum-resolved electronic structure of V-doped WSe_2 in comparison to pristine bulk samples. The samples were grown in our laboratory by chemical vapor transport and with different doping concentrations. Future magnetic structure investigations such as X-ray magnetic circular dichroism and spin- and momentum-resolved photoemission spectroscopy are discussed.

O 29.19 Tue 12:30 Poster A

Ultrafast spin dynamics and their layer-specific spin polarization of conduction band electrons in pristine and Cs-doped WSe_2 — •Sebastian Hedwig, Gregor Zinke, Benito Arnoldi, Benjamin Stadtmüller, and Martin Aeschlimann — Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau, Erwin-Schrödinger-Str. 46, 67663 Kaiserslautern, Germany

Layered 2D Van-der-Waals systems are a highly intriguing class of low-dimensional materials with promising spin functionalities for future nanoscale spintronic applications. A particularly interesting member of this class of materials are WSe$_2$ bulk crystals with their hidden spin polarization. These centrosymmetric bulk crystals exhibit an overall spin degenerate band structure while each individual layer is characterized by spin-split valence bands. We conduct time-, spin- and angle-resolved photoemission experiments to study the optically excited spin carrier dynamics at the K- and Σ-points of WSe$_2$. The high surface sensitivity of the photoemission experiments allows us to disentangle the spin polarization of the bands in the first WSe$_2$ layer. After an initial spin-selective excitation at the K-point, which depends on the pump light polarization [1], we observe a subsequent intraband scattering from the K- to the Σ-point of the WSe$_2$ conduction band. This intraband scattering leads to a time-dependent change in the spin polarization of the carriers at the Σ-point. We compare the optically generated spin-dependent carrier population at the Σ-point with that one of Cs-doped WSe$_2$ for which the conduction band valley at the Σ-point is fully occupied. [1] Phys. Rev. Lett. 117, 277201 (2016)

O 29.20 Tue 12:30 Poster A

Electronic structure of intercalated stacks of two-dimensional crystals — Franziska Rauh and •Sabine Körbel — Institute of Condensed Matter Theory and Optics, Friedrich Schiller University Jena, Germany

Layered Van-der-Waals crystals with their atomic sheets separated by several Ångström, such as stacks of two-dimensional transition-metal dichalcogenides, allow for intercalation with elements or molecules. Here we consider intercalation as a means to manipulate the electronic structure of the layered crystal. We attempt to turn a metal into an insulator, and to create a stair-case band alignment in another insulator. We determine the electronic structure of the intercalated crystals from first principles using density-functional theory.

O 29.21 Tue 12:30 Poster A

Modelling ARPES matrix elements in a 2D van-der-Waals ferromagnet — •Tania Mukherjee[1,2], Lawson Lloyd[2], Samuel Beaulieu[3], Michael Schuler[4], Jyoti Krishna[5], Túlio de Castro[2], Shuo Dong[6], Victoria Taylor[2], Yoav William Windsor[1,2], Martin Wolf[2], Laurenz Rettig[2], Ralph Ernstorfer[1,2], and Tommaso Pincelli[1,2] — [1]Technische Universität Berlin, Berlin, Germany — [2]Fritz-Haber-Institute of the Max Planck Society — [3]University of Bordeaux, France — [4]PSI, Villigen, Switzerland — [5]Freie Universität Berlin, Berlin, Germany — [6]Beijing National Laboratory for Condensed Matter Physics, Beijing, China

Recent discoveries of 2D materials with intrinsic ferromagnetic ordering show great potential for new spintronics applications. Fe$_3$GeTe$_2$ is a van der Waals ferromagnet that is particularly promising for its high and electrically tunable curie temperature. We have recently observed using ARPES that both local magnetic moments and itinerant spin excitations play a cruicial role in describing the electronic ground state and the lowest energy excitations. In this work we performed first principle DFT calculations to verify our experimental discovery, with a special attention to creating a pipeline between the DFT results and the observed ARPES signal. This is enabled by wannierization of the DFT states, followed by the construction of a tight-binding model. We demonstrate that, including spin orbit coupling, our DFT bands show good agreement with the experimental ARPES bands.The further use of chinook [Day, R.P.,et al. npj Quantum Mater. 4, 54 (2019)] allows to compare calculations and experiments in the photoemission signal.

O 29.22 Tue 12:30 Poster A

Linear and circular dichroism ARPES of the charge density wave material TiSe$_2$ — •Hibiki Orio[1], Maximilian Ünzelmann[1], Jakub Schusser[1], Kai Rossnagel[2,3], and Friedrich Reinert[1] — [1]Universität Würzburg, Experimentelle Physik VII and Würzburg-Dresden Cluster of Excellence, Würzburg, Germany — [2]Deutsches Elektronen Synchrotron DESY, Hamburg, Germany — [3]IEAP and KiNSIS, CAU Kiel, Kiel, Germany

The transition metal dichalcogenide compound TiSe$_2$ transforms into a $2 \times 2 \times 2$ charge density wave (CDW) phase below 200K. The physical process governing the buildup of the CDW has been controversial. To understand the nature of this intriguing property, the hybridized orbital character involved in the CDW gap formation is crucial information. Recent studies show that linear- and circular dichroism (LD and CD) in angle-resolved photoelectron spectroscopy (ARPES) provides insights into the orbital texture of the initial states. Here, using ARPES in the soft x-ray photon energy range, we systematically study the LD and CD of TiSe$_2$ in both the semimetallic and CDW phases.

O 29.23 Tue 12:30 Poster A

Spin polarisation of an Anderson impurity state in MoS2 mirror twin boundaries — •Tfyeche Y. Tounsi[1], Mahasweta Bagchi[1], Affan Safeer[1], Camiel van Efferen[1], Thomas Michely[1], Wouter Jolie[1], Theo A. Costi[2], and Jeison Fischer[1] — [1]II. Physikalisches Institut, Universität zu Köln, Germany — [2]Peter Grünberg Institut, Forschungszentrum Jülich, Germany

Mirror twin boundaries (MTB) are line defects found in monolayer MoS2 islands grown via molecular beam epitaxy on graphene on Ir(111). MTBs form a well-defined 1D system of finite length, hosting confined states. When the highest occupied confined state is at the Fermi level, it splits into a singly occupied and doubly unoccupied states, defining an Anderson impurity. The interaction with the substrate electron bath establishes a Kondo resonance [1].

Here, spin-polarised scanning tunnelling spectroscopy measurements on an MTB in presence of out-of-plane magnetic field at a temperature of 0.4 K show an intensity change of the impurity peaks and Kondo resonance as a function of magnetic field. The experimental data is compared to numerical renormalization group calculations, demonstrating the full spin polarisation of both Anderson impurity sates and Kondo resonance.

[1] van Efferen et al., Nat. Phys. (2023). https://doi.org/10.1038/s41567-023-02250-w.

O 29.24 Tue 12:30 Poster A

Optical characterization of sulfur vacancies in TMDCs under high vacuum — •Joel Verlande, André Maas, Leon Daniel, Lars Breuer, and Marika Schleberger — Universität Duisburg-Essen, Fakultät für Physik and CENIDE, Germany

The characterization of 2D Materials under non-ambient conditions offers the potential for a deeper analysis of various defects in the crystalline structure and their effects on the optoelectronic properties. Especially the investigation under a vacuum can provide us with crucial information about the actual influence of vacancies due to often occurring defect saturation by adsorbates, such as O$_2$ or N$_2$. These gas molecules can act as electron donors in the 2D structure that support the creation of negatively charged trions which have a higher rate of non radiating recombination. This results in a lower photoluminescent yield which could be an unfavourable property of the material.

For this study we generated sulfur vacancies in WS$_2$ and MoS$_2$ CVD-grown monolayers by irradiation with low energy Ar$^+$ ions ($E_{\text{kin}} \leq 500$ eV). Subsequently the samples were analyzed by Raman and photoluminescence spectroscopy in a vacuum cell with pressures of $p < 10^{-4}$ mbar. The high vacuum leads to a dissipation of the gas molecules whereby we were able to examine the influence of the adsorbates on the weight of trionic and excitonic photoluminescent emission.

O 29.25 Tue 12:30 Poster A

Azimuthal- and valley dependent second harmonic generation response of monolayer MoS2 — •Jin Yu, Tao Yang, R. Kramer Campen, and YuJin Tong — Universität Duisburg-Essen, Fakultät für Physik, Experimentalphysik, Lotharstr. 1, 47057 Duisburg

Transition metal dichalcogenides (TMDCs) offer advantages in terms of rapid information processing speed and low energy consumption compared to conventional electronic components. Understanding how spin-valley locking controls the interaction of TMDCs and light is crucial for optimizing TMDCs for practical applications in quantum technology. In this study, we employed elliptically and circularly polarized lasers to excite monolayers of molybdenum disulfide. Our results reveal significant changes in the disappearance of sixfold symmetry, larger azimuthally dependent second harmonic generation (SHG) intensity extremes, and the rotation of a sixfold pattern due to variations in laser polarization from linear to circular and our data aligns closely with the predictions of the steady-state model. According to the steady-state model, under low-light-intensity excitation conditions, the angle of the sixfold pattern rotation is proportional to the difference in the number of particles between the valleys. This result provides a quantitative understanding of the SHG valley polarization signal.

O 29.26 Tue 12:30 Poster A

Integrated Computational Approach for the Treatment of 2D Heterostructures — •Anastasiia Nihei[1,2], Roman Kempt[1], Tom Barnowsky[1,2], Thomas Heine[1,2], Stefano Curtarolo[3], and Rico

Friedrich[1,2,3] — [1]TU Dresden — [2]Helmholtz-Zentrum Dresden-Rossendorf, Dresden — [3]Duke University, Durham, USA

Heterostructure interfaces of two-dimensional (2D) materials enable the realization of advanced electronic functionalities at the nanoscale. The efficient computational *ab initio* modelling of these systems is, however, a challenge as it requires the proper lattice matching of the component 2D sheets with minimal strain. This often results in large structures with hundreds to thousands of atoms.

Here, we utilize the newly developed hetbuilder tool to automate the heterotructure cell construction based on coincidence lattice theory [1,2]. It is integrated with the AFLOW materials database and software [3,4] allowing for an efficient workflow for the structure generation from the bulk parent systems. We benchmark the approach by performing *ab initio* calculations on several different heterostructures of 2D materials and study their electronic properties.

[1] D. S. Koda *et al.*, J. Phys. Chem. C **120**, 10895 (2016).
[2] https://zenodo.org/record/4721346.
[3] M. Esters *et al.*, Comput. Mater. Sci. **216**, 111808 (2023).
[4] C. Oses *et al.*, Comput. Mater. Sci. **217**, 111889 (2023).

O 29.27 Tue 12:30 Poster A

Self-assembled Moiré superstructure of MXene — •Andrea Cabero del Hierro[1], Kuanysh Zhussupbekov[1,2], Samuel Berman[1], Dahnan Spurling[2], Ainur Zhussupbekova[1,2], David D. O'Regan[1], Igor V. Shvets[1], and Valeria Nicolosi[2,3] — [1]School of Physics, Trinity College Dublin, Dublin 2, D02 PN40, Ireland — [2]School of Chemistry, Trinity College Dublin, Dublin 2, D02 PN40, Ireland — [3]Centre for Research on Adaptive Nanostructures and Nanodevices (CRANN) and Advanced Materials and Bioengineering Research (AMBER), Trinity College Dublin, Dublin 2, Ireland

The atomically reconstructed moiré superlattice and its impact on the microscopic electronic structure remains absent. In this investigation, we meticulously inspect and compare the self-assembled moiré superlattices of MXene. Employing a combination of experimental scanning tunneling microscopy/spectroscopy observations and ab initio simulations, we investigated three distinct self-assembled moiré patterns characterized by wavelengths approximately around 2.32 nm, 2.17 nm, and 1.12 nm. Our results illuminate a non-monotonic behavior in the moiré potential concerning periods on the valence band side. This research not only contributes to a detailed comprehension of MXene's moiré phenomena but also establishes a fresh foundation for further exploration into unique correlated phases.

O 29.28 Tue 12:30 Poster A

Exploring the structure of extended defects in MoS_2 bilayers with a machine-learned ACE potential — •Kevin Dhamo and Bernd Meyer — Interdisciplinary Center for Molecular Materials and Computer Chemistry Center, FAU Erlangen-Nürnberg, Germany

Layered transition metal dichalcogenides (TMDs), for example MoS_2, have gathered much attention due to their unique tunable electrical, optical, thermal, and tribological properties. TMDs are usually applied as stacks of layers, which are prone to include extended defects such as in-plane dislocations or misalignments due to the rotation of subsequent layers. Density-functional theory (DFT) would be the method of choice to study the properties of such extended defects. However, the representation of extended defects often requires very large unit cells, which makes DFT calculations unfeasible.

In this work, we use DFT data to train an atomic cluster expansion (ACE) machine-learned interatomic potential for MoS_2. The capabilities of the ACE potential are benchmarked against DFT calculations of MoS_2 bilayers by comparing binding energy curves and Gamma surfaces for different bilayer stackings. Finally, the ACE potential is applied to study the structural properties of dislocations in MoS_2 bilayers and the atomic relaxations in the moiré pattern of twisted bilayer MoS_2 for different rotation angles.

O 29.29 Tue 12:30 Poster A

SNOM of lateral TMDC heterostructures — •Philipp Schwendke[1], Samuel Palato[1], and Julia Stähler[1,2] — [1]Humboldt-Universität zu Berlin — [2]Fritz-Haber-Institut der MPG

Semiconductor heterojunctions are the basis of many modern devices such as transistors, LEDs, and photovoltaics. 2D semiconductors such as TMDCs promise similar applications at an atomic level, which is why their interfaces are of particular interest. A variety of useful heterojunction properties have been realized, such as the photovoltaic effect and current rectification. While stacked van der Waals heterostructures ideally offer spatially homogeneous properties, lateral heterojunctions feature a lateral charge transfer following the band alignment across an interface region. Here, the charge carrier energy levels and lifetimes vary spatially in a way that is to date not fully understood. To explore said properties, we use scanning near-field optical microscopy (SNOM) combined with a femtosecond laser to investigate electron dynamics at ultrafast time scales with nm spatial resolution. Using previously introduced quadrature-assisted discrete (QUAD) demodulation, we are able to perform SNOM with a low repetition rate laser producing highly tunable fs laser pulses. QUAD demodulation is compatible with known methods of noise reduction, e.g. pseudo-heterodyne or balanced detection, and straightforward to extend into a pump-probe scheme for time resolution. This makes time-resolved SNOM an ideal candidate for the investigation of electronic properties of lateral heterojunctions.

O 29.30 Tue 12:30 Poster A

Compressed charges and interface dipoles persist at two-dimensional van der Waals heterojunctions — •Jae-Hyeok Ko[1], Giyeok Lee[1,3], Woosun Jang[2], and Aloysius Soon[1,3] — [1]Department of Materials Science & Engineering, Yonsei University, Seoul, Republic of Korea — [2]Integrated Science & Engineering Division, Underwood International College, Yonsei University, Incheon 21983, Republic of Korea — [3]School of Physics, University of Sydney, Sydney 2006, NSW, Australia

Heterojunctions of two-dimensional (2D) van der Waals (vdW) nanomaterials often exhibit unusual, "non-textbook" bonding mechanism that involves distinct orbital coupling within the compressed vdW gap. To unravel this atypical bonding mechanism for 2D heterointerfaces, using density-functional theory calculations, we examine the compressed charge redistribution and interface dipoles for the heterostructures of $ReSe_2$/graphene, $ReSe_2$/*h*-BN, and $ReSe_2$/*h*-BN/graphene. Here, we report the optimized atomic structures, electronic density-of-states, (integrated) planar-averaged electron density differences for these heterostructures, while focusing on the explicit contributions of the conductive graphene and insulating *h*-BN substrates to the interlayer confined charges and dipoles. Using other 2D heterosystems, we demonstrate that this charge compression effect in the vdW gap is ubiquitous and general regardless of the nature of the substrates and supports. This study highlights the importance of understanding the interlayer charges and dipoles and how this may contribute to the better designs of remote charge transfer systems in modern electronics.

O 29.31 Tue 12:30 Poster A

Unveiling the nature of atomic defects in graphene on a metal surface — •Karl Rothe, Nicolas Néel, and Jörg Kröger — TU Ilmenau, Institut für Physik

Argon ion bombardment of graphene-covered Ir(111) induces two types of atomic-scale defects. Using a scanning tunneling microscope, both defects appear as a depression without discernible interior structure suggesting the presence of vacancy sites in the graphene lattice. With an atomic force microscope, however, only one kind of defect can be identified as a site with missing carbon atoms, while the other kind reveals an intact graphene sheet. Spatially resolved spectroscopy of the differential conductance and the measurement of short-range force variations as a function of the lateral and vertical probe-defect distance corroborate the different character of the defects. The tendency of the vacancy defect to form a chemical bond with the microscope probe is reflected by the strongest attraction at the vacancy center as well as by hysteresis effects in force traces recorded for tip approach to and retraction from the Pauli repulsion range of vertical distances.

Financial support by the Deutsche Forschungsgemeinschaft through Grant No. KR 2912/17-1 is acknowledged.

O 29.32 Tue 12:30 Poster A

Characterising nitrogen defects in graphene grown by bottom-up synthesis — •Liv Warwick[1,2], Matthew A. Stoodley[2,3], Jonathan Bradford[1], Matthew Edmondson[1], Benedikt P. Klein[2,3], Fulden Eratam[2], Henry P. Hoddinott[2,4], Luke A. Rochford[5], Reinhard J. Maurer[3], David A. Duncan[2], and Alex Saywell[1] — [1]University of Nottingham, Nottingham, UK — [2]Diamond Light Source, Didcot, UK — [3]University of Warwick, Coventry, UK — [4]University of Swansea, Swansea, UK — [5]University of Cambridge, Cambridge, UK

Recently, we have pioneered a new method for the growth of defective graphene, e.g. graphitic material containing heteroatoms or vacancies, [1] by 'bottom-up' on-surface growth methodologies. In our chemical vapour deposition process [2] we use an aromatic precursor that contains the same structural element as our desired defects. Here, using a similar method, we will present a low temperature scanning tunnelling microscopy (LT-STM), X-ray photoelectron spectroscopy (XPS) and normal incidence X-ray standing waves (NIXSW) study of graphene containing nitrogen defects, grown from a nitrogen containing precursor. By transferring the samples between the different technique chambers via an ultra-high vacuum suitcase, we were able to correlate spectroscopic differences to morphological changes on the surface.

[1] Bhatt, M. D. et al. RSC Adv. 12, 21520-21547 (2022) [2] B. P. Klein. et al. Appl. Phys. Lett., 2022, 121, 191603.

O 29.33 Tue 12:30 Poster A

Intermediate diffusive-ballistic electron conduction around mesoscopic defects in graphene layers — Toni Marković[1], Wei Huang[1], William S. Huxter[2], Pietro Gambardella[1], and •Sebastian Stepanow[1] — [1]Department of Materials, ETH Zurich — [2]Department of Physics, ETH Zurich

Non-diffusive effects in charge transport become relevant as device sizes and features become comparable to the electronic mean free path. As a model system, we investigate the spatial transport around mesoscopic defects, which we refer to as pits, in graphene with scanning tunneling potentiometry. Diffusive and ballistic contributions to the transport dipole are investigated by simultaneously resolving the nanoscale topography of the pits and measuring the local electrochemical potential change in the surrounding area. We find evidence of transport in the intermediate regime between diffusive and ballistic limits, as the potential changes around pits are substantially underestimated by diffusive models. We also observe a dependence of the dipole magnitude on the shape of the pits. Our experiments and modelling are supported by lattice Boltzmann simulations, which highlights the importance of the ratio between the pit size and the mean free path in the intermediate transport regime.

O 29.34 Tue 12:30 Poster A

Imaging the local Electrochemical Potential of Graphene with Scanning Tunneling Potentiometry — Toni Marković, Wei Huang, •Mark Canavan, Jeong Ah Seo, Pietro Gambardella, and Sebastian Stepanow — Department of Materials, ETH Zurich, 8093 Zurich, Switzerland

Two-dimensional material systems and heterostructures have recently emerged as a promising class of materials for high performance electronic devices. In such systems, understanding the influence nanoscale transport effects, such as scattering at atomic defects and interfaces is crucial. Scanning Tunneling Potentiometry (STP) allows for studying charge transport on the nanoscale by relating the local electro-chemical potential to morphological features of the system. STP maintains the angstrom spatial sensitivity of conventional STM while imaging modifications to the ECP with uV resolution, offering a way to potentially investigate transport phenomena beyond the diffusive regime. Here, we present an implementation of STP in a commercial RHK Pan-Scan STM with an integrated flow cryostat capable of reaching temperatures of 12K [1]. We perform STP on epitaxial graphene to measure the sheet resistance of monolayer and bilayer grown on SiC as well resistances of interfaces between them. To investigate transport effects beyond the diffusive regime, using back-gated graphene samples would allow for STP measurements with charge carries at the Dirac point.

[1] T. Marković et al. Rev. Sci. Instrum. 92, 103707 (2021)

O 29.35 Tue 12:30 Poster A

Observation of different Li intercalation states and local doping in epitaxial mono- and bilayer graphene on SiC(0001) — Wei Huang, •Jeongah Seo, Mark Canavan, Pietro Gambardella, and Sebastian Stepanow — Department of Materials, ETH Zurich, Switzerland

Li intercalation is commonly used to enhance the carrier density in epitaxial graphene and mitigate coupling to the substrate. So far, the understanding of the intercalation process, particularly how Li penetrates different layers above the substrate, and its impact on electron transport remains incomplete. Here, we report different phases of Li intercalation and their kinetic processes in epitaxial mono- and bilayer graphene grown on SiC. The distinct doping effects of each intercalation phase are characterized using scanning tunneling spectroscopy. Furthermore, changes in the local conduction regimes are directly mapped by scanning tunneling potentiometry and attributed to different charge transfer states of the intercalated Li. The stable intercalation marked by the formation of Li-Si bonds leads to significant 56% reduction in sheet resistance of the resulting quasi-free bilayer graphene, as compared to the pristine monolayer graphene.

O 30: Poster: Proximity Effects in Epitaxial Graphene

Time: Tuesday 12:30–14:30 Location: Poster A

O 30.1 Tue 12:30 Poster A

Quasi-freestanding graphene on SiC(0001) by intercalation of silicon — •Niclas Tilgner and Thomas Seyller — TU Chemnitz Institut für Physik

To observe the extraordinary properties of freestanding graphene, the atmospheric pressure growth of epitaxial graphene has proven itself in recent years. Thereby the annealing of SiC samples in [0001] direction gives rise to a graphitisation of the surface and a carbonrich $(6\sqrt{3}\times 6\sqrt{3})R30^\circ$ reconstruction is formed. This so-called buffer layer is topological equivalent to graphene, but the π-bands are strongly deformed due to remaining bonds to the substrate. One way of decoupling is the intercalation of foreign atoms, which gives rise to a quasi-freestanding graphene layer. In the presented work the decoupling could be achieved by intercalation of silicon. The poster focuses on the discussion of three different preparation techniques in terms of their influence on the sample quality. The investigated methods are the silicon deposition at elevated temperatures, the sequential deposition/annealing and an exchange intercalation. For the latter one, the buffer layer was first decoupled with hydrogen before the process with silicon. Experiments using the diffraction of low energy electrons (LEED) reveal that only with one technique a sufficiently ordering of the silicon could be achieved. Further investigations with photoemission (ARPES, XPS) show the appearance of several surface states on the ordered samples, which are partly attributed to one of the Hubbard bands of the silicon dangling bonds.

O 30.2 Tue 12:30 Poster A

Nanoscopic transport measurements on intercalated epitaxial graphene — •Andreas Cordier, Markus Gruschwitz, Tim Güldenpfennig, and Christoph Tegenkamp — Institut für Physik, TU Chemnitz, Chemnitz, Germany

To further enhance the electronic properties of graphene, intercalation experiments are currently performed in mayn groups. Electronic transport experiments are a direct way to probe the electronic properties. However, the intercalation is often kinetically limited, thus the intercalated areas are inhomogeneous on a nm-scale. Moreover, the buffer layer/SiC(0001) surfaces also exhibit steps on a μm-scale, which further introduces anisotropy to the system. In order to account for these imperfections, we performed nanoscopic 4ppSTM transport measurements, varying the tip distances and geometry.

We realized tip configurations on single terraces as well as across many terraces, thus revealing 2D/1D/2D transport signatures, while increasing the tip distances. Moreover, using square tip configurations, the rotation of this configuration with respect to the step direction, allowed us to determine the anisotropy, which is as high as 2500. The analysis of these results allowed us to deduce the conductivity of the terraces from the conductance which amounts to $\sigma = 600$ kS/m. These experiments experimental taken at room temperature are supported by finite element simulations. Furthermore a metal-insulator transition has been found at 100K that we ascribe to the formation of a band gap in graphene, e.g. due to the induction of spin-orbit coupling in graphene by the intercalated Pb.

O 30.3 Tue 12:30 Poster A

Growth and structure of two-dimensional Pb-Layers below Epitaxial Graphene on SiC(0001) — •Peter Richter[1,2], Franziska Schölzel[1,2], Philip Schädlich[1,2], and Thomas Seyller[1,2] — [1]Technische Universität Chemnitz, Institut für Physik, Chemnitz, 09126 Chemnitz — [2]Center for Materials, Architectures, and Integration of Nanomembranes (MAIN), 09126 Chemnitz

The intercalation of epitaxial graphene (EG) on SiC with various elements has widely been utilized to tailor the electronic properties of the graphene sheet. The ability of elements with strong spin-orbit coupling or even superconducting phases to induce this behavior into the graphene, moves heavy atoms such as Pb into the spotlight [1]. Beyond that, intercalation enables the manufacturing of otherwise unstable 2D-materials and protect it against oxidation.

We investigated the intercalation of EG with Pb in real time by means of Low Energy Electron Microscopy (LEEM), unraveling a strong temperature dependence. Interestingly, intercalation at lower temperatures results in a new phase of the intercalated Pb-layer contrasting the already reported striped- and bubble-phase [2]. Angle-resolved photoemission spectroscopy (ARPES) and low energy electron diffraction (LEED) investigations suggest a strict (1×1) periodicity. Moreover, we studied the phase transition from (1×1) Pb-phase to 'bubble'-phase occurring at 600 °C by X-Ray photoelectron spectroscopy (XPS), thus connecting our intercalation recipe to earlier reports. [1] N.B. Kopnin et al., Phys. Rev. B, 064524, 84 (2011) [2] M.Gruschwitz et al., Materials, 7706, 14 (2021)

O 30.4 Tue 12:30 Poster A

Intercalation of Pb using buffer layer on 4H/SiC(0001) — •Sergii Sologub[1,2], Markus Gruschwitz[2], and Christoph Tegenkamp[2] — [1]Institute of Physics, NAS of Ukraine, Nauki avenue 46, 03028 Kyiv — [2]Institut für Physik, TU Chemnitz, Reichenhainer Str. 70, 09126 Chemnitz

Charge neutrality [1] as well as potential superconductivity [2] and induced SOC of epigraphene by Pb intercalation recently sprouted great interest. We investigated the peculiarities of the intercalation of Pb using buffer layer on 4H/SiC(0001) by SPA-LEED, SEM and STM techniques. Although proposed as an energetically unfavorable process [3], we optimize the intercalation by varying the coverage of deposited Pb, annealing temperature and duration, as well as numbers of deposition-annealing cycles. In particular, repeated cycles of 10 ML Pb-deposition at RT followed by annealing to 500°C for 5 min were effective for the formation of the stripe intercalation phase, especially with additional annealing to 700°C in every two cycles. On the other hand, the long-term annealing (about 15 hours at 350°C) of the 20 ML Pb coverage deposited on the stripe-phase substrate resulted in transformation of the stripe phase into the bubble one which was accompanied by corresponding changes in STM images and LEED patterns. Also of importance is that residual Pb clusters remaining on the surface after the intercalation processes can be mainly removed by annealing to 400°C.

[1] Adv. Mater. Interfaces 10, 2300471 (2023); [2] Nat. Phys. 6, 104 (2010); [3] Carbon 205, 336 (2023)

O 30.5 Tue 12:30 Poster A

Simulations of electronic transport in inhomogeneous intercalated graphene systems — •Tim Güldenpfennig, Markus Gruschwitz, and Christoph Tegenkamp — Institut für Physik, Technische Universität Chemnitz

Pb intercalated buffer layer on 4H-SiC(0001) creates charge neutral quasi-freestanding graphene [1]. The presence of a densely packed Pb monolayer at the interface triggers interest in transport experiments hunting for a superconductivity or proximity-induced SOC in graphene. However, nanoscale measurements sensitively depend on the local distribution of conductive phases. Multiprobe-STM reveal an 1D to 2D transition with increasing tip spacing s on a single terrace. Angle dependent measurements show a discrete anisotropy behavior. We approach the separation of contributing conductive phases by finite element simulations using COMSOL multiphysics. Our model - based on dimensions measured by SEM and XSW - consists of a series of 3.5 μm wide terraces separated by 100 nm wide barriers of alternating conductivities, respectively. Starting with the measured terrace conductivity (600 kS/m) repeated optimization provide best agreement for $R(s)$ for $\sigma_{\text{terrace}} = 500$ kS/m and $\sigma_{\text{barrier}} = 200$ S/m. Applied in a square angle dependent setup the model also recreates the discrete anisotropy behavior. On larger scale, barrier defects yield a continuous but smaller anisotropy. Finally, we explore the separation of two T-dependent phases by varying the conductivity levels in the model based on reference measurements at 30 K and RT. [1] Adv. Mater. Interfaces 10, 2300471 (2023); [2] Materials 14, 7706 (2021)

O 30.6 Tue 12:30 Poster A

Intercalation of Sn beneath the Buffer Layer on SiC(0001) studied by SPA-LEED — •Kurt Hered, Zamin Mamiyev, and Christoph Tegenkamp — Technische Universität Chemnitz, Institut für Physik, Reichenhainer Straße 70R

The intercalation of atomically flat metallic structures beneath epitaxial graphene is interesting for realizing proximity-coupled 2D systems. In this context, carbon group elements are noteworthy due to their well-known correlated electronic properties. In this work, we have investigated the intercalation of Sn beneath the buffer layer (BL) on 4H-SiC(0001) by means of high-resolution SPA-LEED. Superior to conventional LEED, SPA-LEED with its high k-space resolution and capability to investigate vertical and lateral roughness, enables control over the intercalation process. To optimize high-quality Sn-induced interface, we explore different routes, including varying Sn coverages, intercalation temperatures and time. It turns out that in all approaches, Sn intercalation progresses through intercalated disordered interface, an ordered 1×1 phase, and finally $(\sqrt{3}\times\sqrt{3})R30^\circ$ periodicity w.r.t the SiC laticce, achieved above 1000 °C. [1] Quantitative H(S) and G(S) analyses show for the clean system a mosaic two-level BL surface, with >97% BL with 2.5 Å step heights, revealing a flat SiC surface at a 0.01° inclination. After forming the Sn(1×1) interface layer, 95% transforms into QFMLG, with a slightly increased 2.64 Å step height while preserving macroscopic surface orientation. Annealing above 1000 °C results in surface roughening due to deintercalation. [1] Z. Mamiyev et al. Surf. Interfaces 34, 102304 (2022)

O 30.7 Tue 12:30 Poster A

Sn intercalated epitaxial graphene studied by scanning tunneling microscopy — •Dorothee Hennig, Chitran Ghosal, Zamin Mamiyev, and Christoph Tegenkamp — Institute of Physics, Technische Universität Chemnitz, Reichenhainer Str. 70, Germany

Electronically correlated 2D systems can be realized, for example, by adsorption of Sn on SiC(0001) [1]. The generation of such or similar phases as interfacial phases in the vicinity of epitaxial graphene (EG) promises the realization of novel quantum phases. Here we have investigated the intercalation of Sn on buffer layers (BL) on SiC(0001) using low temperature STM. We know from previous LEED investigations that a (1×1) phase forms first after deposition and subsequent heating. Further heating thins this phase further and $(\sqrt{3}\times\sqrt{3})$-reflexes appear [2]. The Sn(1×1)-phase below epitaxial graphene was found only locally with typical sizes of 2-3nm. Between these crystalline areas with distances of 5-10nm. In between, rather disordered Sn phases are formed, probably formed due to lateral strain effects or triggered by selective reactions at the interface due to the former BL. These centers also act as defects for EG, e.g. es seen by the $(\sqrt{3}\times\sqrt{3})$-reconstruction w.r.t. to EG imaged at bias voltage conditions close to Fermi energy. For the deluted phase, showing $\sqrt{3}$-reflexes, large islands (100nm) with a 6x6 periodicity similar to the BL reconstruction were found. Again, this finding suggests that the interface adsorption sites are defined by the former bonds of the BL with the SiC(0001) substrate.

[1] S. Glass et al., PRL 114, 247602 (2015). [2] Z. Mamiyev et al. Suf. Int. 34, 102304 (2022)

O 31: Poster: Organic Molecules on Inorganic Substrates

Time: Tuesday 18:00–20:00 Location: Poster C

O 31.1 Tue 18:00 Poster C

Probing Borazine on Au(111): from single molecules to clusters — •Matthias Zeilerbauer, Marco Thaler, and Laerte Patera — Department of Physical Chemistry, University Innsbruck, Austria

Borazine is the isoelectronic and isostructural inorganic analogue of benzene. However, due to the large difference in electron negativity between N and B most of the charge is drawn by the nitrogen atom, which is expected to lead to polarized B-N bonds. Here, we demonstrate borazine dosing on Au(111) exploiting a cold trap, which removes borane impurities from the gas phase [1]. Upon deposition on Au(111) at 25 K, we observed monomers as well as self-assembled clusters in various sizes, exhibiting a preferential orientation with respect to the substrate. In scanning tunneling microscopy images borazine molecules present a three-fold appearance, being attributed to the different radii of B and N. Despite the high polarity of borazine favoring dipole-dipole mediated intermolecular interactions, the molecules in the clusters are typically surrounded by hydrogen molecules. Spatially resolved tunneling spectroscopy provides maps of the inelastic electron tunneling probability, allowing for the identification of the hydrogen structures. [1] Marc G. Cuxart et al. Sci. Adv.7 2021

O 31.2 Tue 18:00 Poster C

Electrospray Deposition of Large Ring-Molecules — •Ken Kolar[1], Grant Simpson[1], Caroline Storey[2], David Leigh[2], and Leonhard Grill[1] — [1]Dept. of Physical Chemistry, University of Graz, Austria — [2]Dept. of Chemistry, University of Manchester, UK

Study of large molecules on the individual scale is of importance in biological contexts; however, their intact deposition onto a surface poses many challenges due to their large size. In this study, we present the room temperature deposition of large ring-molecules with a mass of 1124 u on an Au(100) surface within a high vacuum environment, using an optimized electrospray deposition (ESD) apparatus. The simple setup consists only of an electrospray stage [1,2] and does not contain any mass separation unit as used in more sophisticated setups [3]. Sample characterization was done by low-temperature scanning tunneling microscopy after sample transfer with a vacuum suitcase between different ultrahigh vacuum machines. The results show the successful deposition of intact molecules with very little surface contamination and thus confirm the potential of this simple technique for studying larger molecules under clean conditions at surfaces [4-6].

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O 31.3 Tue 18:00 Poster C

Orientational transition of dimolybdenum tetraacetate on Au/Cu(111) — •Alexei Nefedov[1], Thomas Strunskus[2], Maria Brzhezinskaya[3], Martin Anstett[4], Benjamin Stadtmüller[4], Martin Aeschlimann[4], and Angelika Kuehnle[5] — [1]Karlsruhe Institute of Technology, Eggenstein-Leopoldshafen, Germany — [2]University of Kiel, Kiel, Germany — [3]Helmholtz-Zentrum Berlin fuer Materialen und Energie, Berlin, Germany — [4]University of Kaiserslautern, Kaiserslautern, Germany — [5]University of Bielefeld, Bielefeld, Germany

Dimolybdenum tetraacetate (MoMo) adsorbed on metal single crystal surfaces shows a partial transition from an ordered close-packed upright standing configuration (chain phase) to a low-packed phase upon cooling (mesh phase). On the base of DFT calculations it is supposed that this phase consists from flat lying molecules. In order to confirm upright/flat orientation of the MoMo molecules in different phases NEXAFS spectroscopy was applied. The MoMo layer of different (0.1-1 ML) coverages were evaporated on the Au/Cu(111) substrate both at room and low temperatures. XPS/NEXAFS measurements were carried out at deposition temperature as well after cooling or/and warming the sample. After NEXAFS experiments it was found that a molecule coverage also plays very important for the transition. Moreover, it was established that, in addition to thermodynamics, kinetic processes may play a crucial role in this process as well.

O 31.4 Tue 18:00 Poster C

Functionalization of AsP monolayer via Amino acids adsorption — Pinar Kaya[1], Okay Tercan[1], Çağil Kaderoğlu[1], Ethem Aktürk[2], and •Handan Arkin[1] — [1]Ankara University, Faculty of Engineering, Department of Physics Engineering — [2]Aydin Adnan Menderes University, Department of Physics

2D materials attract great attention from the scientific world because of their distinct chemical and physical characteristics. Nevertheless, a few shortcomings of these materials restrict their use [1]. Biomolecules and 2D materials can be combined to address these drawbacks [2]. In this study, the single-layer honeycomb structure of the AsP, which is promising in technological applications and has a wide band gap [3,4], was used as a starting point of the hybrid system and adsorption of amino acids Glycine, Serine, which have great potential for use in industry and biomedicine [5] were investigated. Various geometric models of these amino acids on AsP single-layer pristine and vacancy were developed and their structural and electrical characteristics were examined. References [1] Zhang, Hua, et al., Bioactive Materials, 2020, 5, 1071-1086. [2] Mallineni, Sai Sunil Kumar, et al., ACS Applied Materials & Interfaces, 2016, 8,26, 16604-16611. [3] Benam, Z. H.; Arkin, H.; Aktürk, E., Comput. Mat. Sci., 2017, 140, 290-298. [4] Ozdemir, I., et al., Mat. Research Express, 2019, 6,6, 065032. [5] Gioacchinogi, Michael, et al., J. of Molecular Liquids, 2020, 301, 112407.

O 31.5 Tue 18:00 Poster C

A Detailed Look at the Self-Assembly of 1,4-Benzoquinone on Ag(111) — Hendrik Tamm[1], •Lorenz Brill[1], Christoph Wachter[2], Oliver T. Hofmann[2], Roman Forker[1], and Torsten Fritz[1] — [1]Friedrich Schiller University Jena, Institute of Solid State Physics, Helmholtzweg 5, 07743 Jena, Germany. — [2]Graz University of Technology, Institute of Solid State Physics, NAWI Graz, Petersgasse 16/II, 8010 Graz, Austria.

Since many material properties are closely linked to its structure, understanding the structure formation is imperative when trying to engineer new materials. However, many competing effects impact the final structure formation, complicating accurate predictions. Here, we investigate 1,4-benzoquinone on Ag(111) and use CO-functionalized scanning probe microscopy in conjunction with density functional theory calculations to reveal the non-intuitive, tilted adsorption of the molecules. While the self-assembly of 1,4-benzoquinone on Ag(111) has been studied previously, our study provides additional clarification and insights into its structure that were inaccessible at that time.

O 31.6 Tue 18:00 Poster C

LEEM/PEEM study of $F_{16}CuPc$/*p*-6P organic thin films on SiO_2 — •Franz Niklas Knoop, Klaus Stallberg, and Winfried Daum — Institute of Energy Research and Physical Technologies, TU Clausthal, Germany

Systematic investigations of the influence of molecular order on the charge-carrier dynamics in organic semiconductors require well-defined and controllable model systems. While single-crystalline metal surfaces are suitable substrates for the epitaxial growth of long-range ordered organic layers, strong molecule-metal interaction also modifies the carrier dynamics in the layer. In contrast, organic thin films on weakly interacting substrates such as SiO_2 exhibit only poor structural order in general.

Here, we present a low-energy electron microscopy (LEEM) study of the structure of thin copper-hexadecafluorophthalocyanine ($F_{16}CuPc$) films, which were deposited on the native oxide of Si(100) substrates after precovering of the surface with a monolayer of para-sexyphenyl (*p*-6p) molecules. The in vacuo thermally evaporated *p*-6P layer comprises crystalline domains of upright standing molecules. $F_{16}CuPc$ films deposited on *p*-6p precovered substrates exhibit μm-sized, well-ordered domains. In contrast, only nm-sized domains are formed if $F_{16}CuPc$ is deposited directly on the native oxide. Structural investigations with μLEED and AFM are combined with polarization-dependent two-photon photoemission electron microscopy (2P-PEEM) to reveal the influence of molecular order on optical excitations in the $F_{16}CuPc$ film.

O 31.7 Tue 18:00 Poster C

N-Heterocyclic carbenes and olefins on silicon — •Robert Zielinski[1,5], Sandhya Chandola[3], Maximilian Koy[4], Hazem Aldahhak[2], Mowpriya Das[4], Matthias Freitag[4], Uwe Gerstmann[2], Mike T. Nehring[1], Denise Liebig[1], Adrian K. Hoffmann[1], Canan Kosbab[1], Max Rosin[1], Simone Brozzesi[7], Ankita Das[4], Viktoria Balfanz[1], Juls Brühne[1], Wolf Gero Schmidt[2], Conor Hogan[6,7], Mario Dähne[1], Frank Glorius[4], Norbert Esser[1,5], and Martin Franz[1] — [1]Institut für Festkörperphysik, Technische Universität Berlin, Germany — [2]Lehrstuhl für Theoretische Materialphysik, Universität Paderborn, Germany — [3]Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Berlin, Germany — [4]Organisch-Chemisches Institut, Westfälische Wilhelms-Universität Münster, Germany — [5]Leibniz-Institut für Analytische Wissenschaften - ISAS e.V., Berlin, Germany — [6]Istituto di Struttura della Materia-CNR (ISM-CNR), Rome, Italy — [7]Department of Physics, Università di Roma "Tor Vergata", Italy

N-Heterocyclic carbenes and olefins are known to be excellent modifiers and anchors for the functionalization of surfaces. Here their adsorption behavior on Si(111) is investigated using scanning tunneling microscopy, X-ray photoelectron spectroscopy, and density functional theory. Covalently bound monolayers with high thermal stability and large work function reductions are found. The adsorption geometry and ordering behavior depend on the specific molecule and the domains size correlates with the defect density of the substrate.

O 31.8 Tue 18:00 Poster C

Measuring and Mapping Molecular Elongation by NC-AFM: The Case of Nonahelicene and Coronene on Ag(110) — •Max Halbauer[1], Takashi Kumagai[2], Martin Wolf[1], and Akitoshi Shiotari[1] — [1]Fritz-Haber-Institute, Faradayweg 4-6, 14195 Berlin, Germany — [2]Institute for Molecular Science, 38 NishigoNaka, Myodaiji, Okazaki 444-8585, Japan

Non-contact atomic force microscopy (NC-AFM) has become a standard tool for the investigation of individual atoms and molecules on surfaces. However, the measurement of elasticity in single molecules and its submolecular resolution have remained as a largely open question. To address this issue, a spring molecule - nonahelicene ([9]H) - was investigated and compared with its flat counterpart - coronene (cor). The molecules were imaged first by high-resolution scanning tunneling microscopy (STM) and NC-AFM on a Ag(110)-surface at cryogenic temperatures (4.8 K), ultrahigh vacuum (UHV) conditions and CO-tips. Interactions of metallic tips with [9]H and cor and the associated elongations were studied then by frequency shift- distance measurements. A novel model for the description of the NC-AFM signals considering an elastic response in the molecular spring was utilized subsequently to rationalize the behavior of both molecules. The spring constant of [9]H was thereby successfully determined to be 5.6 N/m. A position dependent systematic study of the response above the [9]H molecule was performed then to resolve the elastic component with submolecular resolution. The results demonstrate NC-AFM as a powerful tool for the detection of deformation on the atomic scale.

O 31.9 Tue 18:00 Poster C

Electronic Properties of N-Heterotriangulene Derivatives Adsorbed on Au(111) Investigated with Two-Photon Photoemission Spectroscopy — •Jakob Steidel[1], Ina Michalsky[2], Milan Kivala[2], and Petra Tegeder[1] — [1]Physikalisch-Chemisches Institut, Universität Heidelberg, Germany — [2]Organisch-Chemisches Institut, Universität Heidelberg, Germany

In many organic opto-electronic devices donor-acceptor systems (D-A systems) play a crucial role, for example as the emitting layer in organic light emitting diodes or to enhance charge separation in organic photovoltaics. Triphenylamine derivatives like the planar N-Heterotriangulene-550 (N-HTA-550) are promising electron donors. The introduction of an etheno bridge closes a seven membered ring in N-HTA-557 and strongly modifies the electronic structure of the molecule while retaining its steric demands. By oxidation of the etheno bridge and subsequent condensation with quinoxaline-2,3-diamine N-HTA-557-P, an intramolecular D-A system, is synthesized. In the present contribution we investigate the electronic structure of N-HTA-550, N-HTA-557 and N-HTA-557-P adsorbed on Au(111) with two-photon photoemission spectroscopy (2PPE). In agreement with the results of UV/Vis-absorption spectroscopy we found the optical gap of N-HTA-550 and N-HTA-557-P to be 3.7 eV and 2.6 eV respectively. For N-HTA-557 the electron affinity level and occupied molecular states were determined, yielding a fundamental gap of 2.9 eV. By means of femtosecond time-resolved 2PPE two coverage dependent exciton decay pathways in thin films of N-HTA-557-P were identified.

O 31.10 Tue 18:00 Poster C

Enhanced conductance of molecular states at interstitial sites — Jan Homberg[1], Manuel Gruber[2], Alexander Weismann[1], and •Richard Berndt[1] — [1]Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel — [2]Fakultät für Physik, Universität Duisburg-Essen

Arrays of phthalocyanine molecules on Pb(100) are investigated with scanning tunneling microscopy. Maps of the differential conductance exhibit drastic changes as the sample voltage is being varied. Maximal conductances are observed at positions between the molecules mimicking bonding states. However, the maxima are shown to result from a superposition of non-interacting states. We expect that this effect may be observed from many other molecules.

O 31.11 Tue 18:00 Poster C

functionalized platform molecules on Au and Ag (111) studied with scanning tunneling microscopy — •Behzad Mortezapour[1], Alexander Weismann[1], Sebastian Hamer[2], Rainer Herges[2], and Richard Berndt[1] — [1]Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, Germany — [2]Otto-Diels-Institut fur Organische Chemie, Christian-Albrechts-Universität zu Kiel, Germany

To prepare well-ordered molecular architectures on solid surfaces we use the triangular molecular platform trioxatriangulenium. Its central carbon atom enables vertical attachment of various ligands. We present low-temperature scanning tunneling microscopy results for platforms with the ligands phenyl, 3-pyridyl and sec-butyl. The molecules were sublimated onto Au (111) and Ag (111) surfaces at ambient temperature with 99 % of the molecules remaining intact. On the reconstructed Au substrate, the molecules arrange into hexagonal and honeycomb arrays, preferentially in fcc areas. On Ag, the molecules solely adopt closed-packed hexagonal patterns. Single molecules were manipulated with the scanning tunneling microscope tip. We show first spectroscopic results and present current-distance measurements that cover the transition from tunneling to a single molecule contact.

O 31.12 Tue 18:00 Poster C

Charge transfer at organic-metal interfaces mediated by strong external electric fields — •Nina Kainbacher[1,2] and Peter Puschnig[1] — [1]Institute of Physics, University of Graz, Austria — [2]Institute of Solid State Physics, Graz University of Technology, Austria

Charge transfer at organic-metal interfaces plays a key role in the charge injection properties of organic-based devices. In this work, we investigate how external electric fields affect the interfacial charge transfer. This offers a new window into deepening our fundamental understanding of charge transfer processes at hybrid interfaces. To investigate this with low computational effort, we perform a theoretical study on prototypical systems using ground-state density functional theory under static external fields applied perpendicular to the substrate plane. The chosen systems comprise the π-conjugated molecules, p-sexiphenyl, PTCDA and pentacenetetrone on three kinds of metal surfaces, Cu(110), Ag(110) and Ag(111), respectively. This choice enables us to compare molecules with different electron affinities as well as substrates with varying interaction strength. Charge transfer at the interface is induced by the vibration of the molecule perpendicular to the substrate plane and by the polarization in response to the electric field. For the former effect, we analyze the density of states as well as the bond lengths as a function of the adsorption distance. Finally, we show how the electric fields affect charge transfer at the interface by studying charge density differences and projections of the density of states.

O 31.13 Tue 18:00 Poster C

Benchmarking theoretical electronic structure methods in a wide binding energy range with photoemission orbital tomography — •Anja Haags[1,2,3], Xiaosheng Yang[1,2,3], Larissa Egger[4], Dominik Brandstetter[4], Hans Kirschner[5], Alexander Gottwald[5], Mathias Richter[5], Georg Koller[4], Michael G. Ramsey[4], François C. Bocquet[1], Serguei Soubatch[1], F. Stefan Tautz[1], and Peter Puschnig[4] — [1]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, 52425 Jülich, Germany — [2]Jülich Aachen Research Alliance (JARA), Fundamentals of Future Information Technology, 52425 Jülich, Germany — [3]Experimental Physics IV A, RWTH Aachen University, 52074 Aachen, Germany — [4]Institute of Physics, University of Graz, NAWI Graz, 8010 Graz, Austria — [5]Physikalisch-Technische Bundesanstalt (PTB), 10587 Berlin, Germany

Photoemission orbital tomography (POT) has become a powerful tool to investigate the electronic structure of organic molecules on surfaces. In this combined experimental and theoretical technique, the measured photoemission intensity distributions at particular binding energies can be understood using calculated momentum-space signatures of molecular orbitals. For bisanthene adsorbed on Cu(110), we apply POT in a wide binding energy range to obtain an experimentally-derived molecular orbital projected density of states. We not only identify 13 π and 22 σ orbitals, but also access the respective binding energies of each orbital. This allows us to benchmark the performance of density functional theory calculations in order to account for a precise orbital energy alignment at the molecule/metal interface.

O 31.14 Tue 18:00 Poster C

Quantum transport across single-molecule junctions bridging adlayer-modified electrodes — •Mong-Wen Gu[1,3], Chi-Ta Lai[1], I-Chih Ni[2], Chih-I Wu[2], and Chun-hsien Chen[1] — [1]Department of Chemistry, National Taiwan University, Taipei, Taiwan — [2]Graduate Institute of Photonics and Optoelectronics, National Taiwan University, Taipei, Taiwan — [3]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, Jülich, Germany

To explore the complexity of quantum transport across single-molecule junctions, it is essential to develop new types of electrode materials beyond gold electrodes which are overwhelmingly popular due to their reliability and inertness in the fabrication of molecule junctions. Herein, two electrochemical techniques, underpotential deposition and surface-limited redox replacement, were used to prepare gold electrodes which were modified by an atomic layer of platinum and palladium. The single-molecule conductance of α,ω-hexanes (X-$(CH_2)_n$-X, X = -SCH_3 and -CN, n = 4,6,8) on the bimetallic electrodes was found to be 2*30 times higher than that on bare gold electrodes. This enhanced conductance is ascribed to the partially filled d-shells on the bimetallic electrodes. Such a feature leads to a significantly increased SDOS at the Fermi energy and thus to an improved efficiency of interfacial transport. This work demonstrates a rational strategy for engineering the molecule-metal surface to develop emergent single-molecule testbeds.

O 31.15 Tue 18:00 Poster C

LT-STM investigation of 9-Anthrol on Au (111) — •Franz Plate[1], Suchetana Sarkar[1], Diego Peña[2], and Francesca Moresco[1] — [1]Center for Advancing Electronics Dresden, TU Dresden, 01062 Dresden, Germany — [2]Centro de Investigación en Química Biolóxica e Materiais Moleculares (CiQUS), Universidade de Santiago de Compostela, Santiago de Compostela 15782, Spain

We present a low temperature scanning tunneling microscope (STM) investigation of 9-Anthrol adsorbed on the Au (111) surface. This chemically unstable tautomer of Anthrone, is studied at the single molecule level, showing its structural and electronic properties. Anthrone was deposited on Au(111) by direct contact deposition followed by annealing at 200°C. High resolution STM images in constant height mode using a CO functionalized tip are presented, as well as scanning tunneling spectroscopy spectra and differential conductance maps of the single molecules.

O 31.16 Tue 18:00 Poster C

Towards k-space imaging molecular orbitals on ferromagnetic surfaces — •Martin Anstett, Gregor Zinke, Lu Lyu, Benjamin Stadtmüller, and Martin Aeschlimann — Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau, 67663 Kaiserslautern, Germany.

Interfaces between organic molecules and ferromagnets are the key functional units of all molecular spintronic applications. The device functionalities are determined by the details of the strong chemical interactions at the interfaces, which typically lead to strong distortions of the wave functions of the molecular orbitals on the ferromagnetic surfaces.

In this contribution, we present our recent progress in imaging the orbitals of molecules on ferromagnetic surfaces at room temperature by combining spin- and momentum-resolved photoemission spectroscopy and photoemission orbital tomography. The main challenge in this endeavour is the lack of ordered or even aligned molecular films on magnetic surfaces. Using ultrathin cobalt films on Au(111) as a ferromagnetic substrate, we are able to realise long-range self-assembled molecules on this surface. These films show distinct emission patterns for the different molecular orbitals. We will discuss signatures of the strong molecule-surface interaction in the molecular orbital emission pattern.

O 31.17 Tue 18:00 Poster C

Collective States in 2D Molecular Monolayers — •Sabrina Juergensen[1], Moritz Kessens[1], Charlotte Berrezueta-Palacios[1], Nikolai Severin[2], Sumaya Ifland[2], Jürgen P. Rabe[2], Niclas S. Mueller[3], and Stephanie Reich[1] — [1]Freie Universität Berlin, Berlin, Germany — [2]Humboldt Universität zu Berlin, Berlin, Germany — [3]NanoPhotonics Centre - University of Cambridge, Cambridge, UK

Organic monolayer crystals have emerged as a new material to replace conventional semiconductors like silicon or TMDs and have become a wide research area in recent years. Mostly these 2D materials are grown out of dye molecules that tend to form J-aggregates leading to outstanding optical properties. In J-aggregates the dipole moments of the molecules are aligned resulting in a strong coupling of the molecules, forming a collective state. The photonic excitation of the collective states to higher electronic levels will result in a very narrow and strong emission also known as superradiance.

To study the collective state in an 2D material in dependency of the dielectric environment, we grew monolayers of a perylene derivative on two different van der Waals materials with different electronic properties, providing a perfect platform to study the fundamental mechanism of the collective state. High resolution AFM was used to determine the packing density of the molecules in a monolayer. Different optical methods were used to characterize the collective state of the molecular monolayer. The experimental data were further compared to real space simulations.

O 32: Poster: Solid-Liquid Interfaces

Time: Tuesday 18:00–20:00 Location: Poster C

O 32.1 Tue 18:00 Poster C

Molecular dynamics simulations of dicalcium silicate - water interfaces by High-Dimensional Neural Network Potentials — •Bernadeta Prus[1,2] and Jörg Behler[1,2] — [1]Theoretische Chemie II, Ruhr-Universität Bochum, Germany — [2]Research Center Chemical Sciences and Sustainability, Research Alliance Ruhr, Germany

In recent years, there has been a growing utilization of High-Dimensional Neural Network Potentials (HDNNP) based on Density Functional Theory (DFT) calculations to enable high-quality molecular dynamics simulations of water interactions with a variety of solid minerals. This study explores the case of dicalcium silicate (Ca_2SiO_4), which is important in many fields. This chemical compound exhibits five polymorphic states, the low-temperature polymorph, denoted as γ, is naturally occurring in the Calcio-olivine mineral. The primary focus of this research is to compare the reactivity in contact with water of different terminations along the [010] surfaces of the γ polymorph of dicalcium silicate. The chosen computational approach allows the development of a single HDNNP suitable for molecular dynamics simulations for all distinct interfaces significantly reducing the computational time.

O 32.2 Tue 18:00 Poster C

Constructing High-Dimensional Neural Network Potentials for Oxide-Water Interfaces — •Jan Elsner and Jörg Behler — Theoretische Chemie II, Ruhr-Universität Bochum, Germany, and Research Center Chemical Sciences and Sustainability, Research Alliance Ruhr, Germany

Oxide-water interfaces are promising systems for catalytic water splitting, a process of significant interest due to its potential in sustainable hydrogen production. However, the complex nature of such interfaces and long time scales associated with dynamical processes presents a substantial theoretical challenge. High-Dimensional Neural Network Potentials (HDNNPs) provide a solution to these challenges by enabling atomistic simulations with DFT-level accuracy at only a fraction of the computational expense. We present the construction of a HDNNP suitable for studying oxide-water interfaces with the overarching goal to utilize these HDNNPs for atomistic simulations of these interfaces.

O 32.3 Tue 18:00 Poster C

Development of high-dimensional neural network potentials for solid-liquid interfaces — •Daniel Trzewik[1,2], Moritz R. Schäfer[1,2], Alexander L. Knoll[1,2], and Jörg Behler[1,2] — [1]Theoretische Chemie II, Ruhr-Universität Bochum, Germany — [2]Research Center Chemical Sciences and Sustainability, Research Alliance Ruhr, Germany

Solid-liquid interfaces play an essential role for chemical processes involving catalysis, electrochemistry and materials science. Modelling of these interfaces with first-principles methods remains computationally demanding due to the required system size. Machine learning potentials offer an efficient alternative at similar level of accuracy. The utilized high-dimensional neural network potentials (HDNNPs) in this project allow for a detailed investigation of solid-water interfaces. Molecular dynamics simulations reveal the structural arrangement and properties of the interface water as well as the interaction with the surface.

O 32.4 Tue 18:00 Poster C

Detection of surface intermediates for oxygen evolution on hematite using operando sum frequency spectroscopy — •Yuke Yang, Yujin Tong, and R. Kramer Campen — Faculty of Physics, University of Dusiburg-Essen, 47057 Duisburg, Germany

Hematite (α-Fe_2O_3) is a photoanode material that is widely studied for photoelectrochemical oxygen evolution (OER) due to its abundance, stability, non-toxicity, etc. However, the mechanism of OER on hematite that limits its performance is still undetermined. There is still controversy over single- and multi-hole mechanisms depending on the sample and electrolyte. Methods to probe the OER mechanism are indirect and thus possible control of the mechanism by the change in the sample is difficult to infer. Vibrational sum frequency spectroscopy (VSFS) is an interface-specific technique for systems containing bulk phases with inversion symmetry, which allows the direct observation of intermediates. Our VSFS results reveal the bias-dependent (dis)appearance of a succession of intermediates such as Fe-OH, Fe=O, and Fe-OOH. The results unambiguously show that the OER on our sample proceeds via a single-hole mechanism at pH 12.8. Additionally, we observe a bias-dependent surface phonon mode at around 650cm-1 due to lattice distortion, which may be related to the polaron formation and transport. Because of femtosecond pulses in the VSFS measurement, this work also suggests a path toward resolving chemical dynamics on ultrafast timescales.

O 32.5 Tue 18:00 Poster C

Influence of Cholic Acid and 1-Methylbenzimidazole on Cell Performance in Water-Based Dye-Sensitized Solar Cells — •Leon Rosenbecker, Daniel Holzhacker, and Derck Schlettwein — Institut für Angewandte Physik, Justus-Liebig-Universität Gießen

Dye-Sensitized Solar Cells (DSSCs) present an environmentally friendly low-cost photovoltaic concept, interesting under conditions of low light intensity and, in particular, for indoor applications. However, commonly used materials such as platinum at the counter electrode, acetonitrile as the solvent, and cobalt complexes as the redox mediator can be considered problematic. In this work, DSSCs with aqueous solutions of readily available 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO) as the redox mediator were prepared. To suppress undesired recombination, we studied cholic acid (CA) as a co-adsorbate, 1-methylbenzimidazole (MBI) as an electrolyte additive, or a combination of both. CA as a co-adsorbate improved both the short-circuit current density (jsc) and fill factor (FF) while not affecting the open-circuit voltage (Voc) up to a concentration of 7.5 mM in the dye bath. In case MBI was used as the additive in the electrolyte, also the Voc was improved, while jsc and FF were found to be higher than for CA. Increasing the MBI concentration from 0.1 M up to 0.4 M steadily increased the performance. A combination of both additives further increased the short-circuit current density while not sacrificing on the other cell characteristics.

O 32.6 Tue 18:00 Poster C

Comparison of PBE-, RPBE-, and RPBE-D3-approximation for diffusion energy barriers of S_{ad} and CH_3S_{ad} on clean and halogen-covered Cu(100) surfaces — •Falk Wendorff and Eckhard Pehlke — Institut für Theoretische Physik und Astrophysik, Christian-Albrechts-Universität zu Kiel, 24098 Kiel, Germany

In density functional calculations often different approximations to the exchange-correlation energy functional are appropriate depending on the particular type of physical system. While the approximation by Perdew, Burke and Ernzerhof, PBE-GGA [1], is widely applied in surface calculations, water is known to be overstructured when using PBE-GGA and other functionals are preferable [2]. Thus, as a prerequisite for future studies of diffusion at electrochemical interfaces (motivated by [3]), we have investigated how much the diffusion energy barriers of S_{ad} and CH_3S_{ad} on clean and halogen-covered Cu(100) surfaces versus vacuum change, when using RPBE [4], without and with additional empirical correction for van der Waals interactions [5] instead of PBE. The calculations have been carried out with PWscf and PWneb from Quantum ESPRESSO [6].

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O 32.7 Tue 18:00 Poster C

DFT study of S subsurface diffusion on c(2×2)-Br covered Ag(100) — •Sönke Buttenschön, Jannik Thomas, and Eckhard Pehlke — Institut für Theoretische Physik und Astrophysik, Christian-Albrechts-Universität zu Kiel, 24098 Kiel, Germany

In their video-STM study of the diffusion of sulfur adatoms on c(2×2)-Br covered Ag(100), Rahn and Magnussen [1] have observed an astounding subsurface diffusion mechanism different from the S_{ad} diffusion within the Br adlayer. They conclude that the S atoms can move within the top Ag layer underneath the c(2×2) halogen adsorbate layer. We have carried through density functional total-energy calculations with PWscf and PWneb from the Quantum ESPRESSO package [2] to examine the diffusion mechanism. The vacancy formation energy in the top Ag layer underneath the halogen away from, and close to the sulfur as well as energy barriers for various relevant elementary diffusion hops are presented.

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O 32.8 Tue 18:00 Poster C

Electrochemical Setup for High-Resolution Hydration Layer Mapping under Potential Control — •Antonia Köhler, Ralf Bechstein, and Angelika Kühnle — Physical Chemistry I, Bielefeld University, Germany

Electrochemical processes are of importance in a wide range of fields such as energy storage, sensors and electrocatalysis. However, many fundamental processes at the electrode-electrolyte interface are still poorly understood. It is widely known that the presence of an applied electric field has a significant influence on the structure at the interface, including the hydration structure. An ideal method for investigating the hydration structure at the molecular level is three-dimensional atomic force microscopy (3D-AFM). Here, we present 3D-AFM data unravelling the hydration structure at the Au(111) surface in the absence of an applied electric field. Furthermore, an electrochemical setup that allows for high-resolution hydration layer mapping under potential control is described. It is based on the 3D-AFM setup, additionally equipped with a three-electrode arrangement connected to a potentiostat. In future, this setup should be capable to map the electrode-electrolyte interface under potential control.

O 32.9 Tue 18:00 Poster C

In-liquid plasma modified Ni expanded metals as hydrogen evolution reaction and oxygen evolution reaction catalyst — •Justus Leist, Lukas Forschner, Timo Jacob, and Albert K. Engstfeld — Ulm University, Institute of Electrochemistry, Ulm, Germany

Applying a sufficiently high voltage to a gas-evolving electrode in an aqueous electrolyte can lead to the formation of a water vapor sheath around the electrode, in which a plasma can be ignited. With this so-called Contact Glow Discharge Electrolysis (CGDE), it is possible to modify the surface structure of an electrode. [1] This in turn can be used to tune the catalytic activity of an electrode, as shown recently for the oxygen evolution reaction (OER) on CGDE modified Ni foams.[2]

In this work, we show the effect of CGDE treated Ni expanded metals on the hydrogen and oxygen evolution reaction. These kinds of electrodes are less prone to bubble inclusion compared to foams. The structural properties of the electrodes are studied by scanning electron microscopy (SEM) and electrochemical methods, such as cyclic voltammetry (CV). The catalytic activity for the OER and HER is investigated using alkaline flow electrolysis cells to mimic industrial operating conditions at different temperatures. The observed trends in reactivity are discussed with respect to the structural changes obtained due to the CGDE and their relationship with oxidation states before and the temperature during HER and OER.

[1] Artmann et al. *ChemPhysChem* **22** (2021) 242. [2] Hausmann et al. *Advanced Energy Materials* **12** (2022) 38.

O 32.10 Tue 18:00 Poster C

Classical and ab initio simulations of the gold - water interface including instantaneous polarisation effects — •Elspeth Smith and Marialore Sulpizi — Ruhr Universität Bochum, Bochum, Germany

Electronic polarisation plays a huge role in the structure and dynamics of many systems, including metal-electrolyte interfaces, and its inclusion requires careful consideration in the setup of any molecular simulation. Typically, classical force fields are not designed to include polarisation effects, however recent advancements in their design have began to accurately simulate instantaneous polarisation effects. In this work, I study gold-water interfaces, employing a recently developed classical model which includes polarisation effects by means of a harmonically coupled core-shell pair for every gold atom. I compare results obtained from this potential with those from ab initio density functional theory simulations to clarify the extent and applicability of the classical polarizable force field. In particular molecular distribution and orientation at the gold/liquid interface is addressed. These techniques are applied to a number of systems, including pure water and water-electrolyte solutions, as well as different geometries of the gold surface, including the (111) and (100) surfaces.

O 32.11 Tue 18:00 Poster C

Benchmarking various ab initio and empirical models to describe electrified water-metal interfaces — •Samuel Mattoso, Mira Todorova, Stefan Wippermann, and Jörg Neugebauer — Max-Planck-Institut für Eisenforschung , Max-Planck-Straße 1, 40237 Düsseldorf

Modeling of electrified water-metal interfaces provides valuable insights into processes at solid/liquid interfaces and suggests routes towards improving clean energy storage and production, corrosion protection and sustainable electrocatalysis. Density functional theory (DFT) is often the tool of choice. A faithful description of metal-water interfaces requires an equally accurate description of both the solid and the liquid phase, in addition to the interface between them. To this end, we explore the impact of different exchange-correlation functionals on relevant bulk properties, such as the water radial distribution function and metal lattice constant. We then use H adsorption to probe our accuracy in describing the Au(111) surface, before employing DFT-based molecular dynamics (MD) simulations to explore the behavior of H at the Au(111)/water interface. In addition, we discuss the employed workflows, encompassing both classical and quantum mechanical, static and dynamic calculations, also under applied bias.

O 32.12 Tue 18:00 Poster C

Temperature-dependent structure formation in the wetting layer of the ionic liquid [C_2C_1Im][OTf] on Au(111) — •Jonas Hauner[1], Hanna Bühlmeyer[1], Simon Trzeciak[2], Julien Steffen[3], Andreas Görling[3], Dirk Zahn[2], and Jörg Libuda[1] — [1]Interface Research and Catalysis, Friedrich-Alexander-Universität Erlangen-Nürnberg, Egerlandstraße 3, 91058 Erlangen, Germany — [2]Computer Chemistry Center, Department of Chemistry and Pharmacy, Friedrich-Alexander-Universität Erlangen-Nürnberg, Nägelsbachstraße 25, 91052 Erlangen, Germany — [3]Chair of Theoretical Chemistry, Friedrich-Alexander-Universität Erlangen-Nürnberg, Egerlandstraße 3, 91058 Erlangen, Germany

We report on in-situ studies on the structure of ultrathin films of the ionic liquid (IL) 1-ethyl-3-methylimidazolium trifluoromethanesulfonate ([EMIm][OTf]) on Au(111). The IL thin films were prepared by physical vapor deposition and investigated by scanning tunneling microscopy (STM). We carried out measurements at sample temperatures between 100 K and 350 K on submonolayers and monolayers of [EMIm][OTf]. In this temperature range, we identified three different surface structures. At a submonolayer coverage, [EMIm][OTf] tends to exhibit a 2D-glass structure growing close to gold terraces and the elbows of the herringbone structure of Au(111). Oblique and hexagonal structures of the IL are visible at higher coverage. We investigated the phase transitions and the melting behavior of these wetting layers by temperature dependent STM measurements. Ordered domains are visible above the bulk melting temperature up to 350 K.

O 32.13 Tue 18:00 Poster C

In Situ Study of Ferrocenylundecanethiol Conformations — •Nelli Kremer, Tim Lämmerzahl, and Eckart Hasselbrink — Universität Duisburg-Essen, Essen, Deutschland

Self-assembled monolayers (SAMs) of alkanethiols have been extensively studied as model systems. Many research groups have investigated the structures of the monolayer under ambient conditions. However, structural changes of monolayers on electrode surfaces in electrolyte solutions in comparison to those in air are expected, because of the interaction between the adsorbed molecules and the species in solution. Therefore SAMs with ferrocene-terminated alkanethiols are of great interest since the orientation of the monolayer can be controlled by the redox-active functional group. By applying a potential, the ferrocene group in the monolayer on a gold surface readily exchanges an electron with gold. Previous studies have shown that the angle between the alkyl chain and the normal to the electrode surface changes upon oxidation, while structural reorientation associated with the rotation of the ferrocene group occurs.

Our vibrational sum frequency spectroscopy setup, which provides a resonant narrowband infrared laser pulse for pumping and a broadband femtosecond IR visible pulse pair, enables us to suppress the nonresonant background, which arises due to the metallic surface, and study the intramolecular vibrational energy redistribution (IVR) of 11-Ferrocenylundecanethiol (FcC11) on gold as well as the change of IVR due to the potential-dependent reorientation.

O 32.14 Tue 18:00 Poster C

Tip Classification of High Resolution AFM Imaging in Liquids — •Farzin Irandoost[1], Fillippo Federici Canova[2], Takeshi Fukuma[3], Tobias Dickbreder[4], Franziska Sabath[4], Ralf Bechstein[4], Angelika Kühnle[4], and Adam S. Foster[1,3] — [1]Department of Applied Physics, Aalto University; Helsinki, Finland — [2]Nanolayers Research Computing Ltd., London, England — [3]Nano Life Science Institute (WPI-NanoLSI), Kanazawa University; Kanazawa, Japan — [4]Physical Chemistry I, Bielefeld University, Germany

AFM imaging in liquids is profoundly influenced by scanning height and tip, leading to different 2D maps for the same crystal surface. This study introduces a workflow to discern diverse contrast patterns arising from AFM scanning height and tip dependency. Our workflow, firstly, clusters images in large experimental datasets based on contrast pattern similarities, then links the clusters to relevant free energy simulations based on the tip characteristics.

Success relies on selecting a sensitive latent vector and implementing a robust clustering benchmark. While statistical analysis of Fourier transforms peaks as image descriptors, aided by tSNE and K-means clustering shows promise in qualitative evaluation, developing a quantitative method for large dataset evaluation remains a priority. To this end, we are developing a method for lattice vector extraction out of experimental data, which makes quantitative benchmarks available for periodic crystal images. Also, the latter can be used as an alternative way of describing images containing periodic patterns.

O 32.15 Tue 18:00 Poster C

Development of Novel Substrates for In Situ Time-resolved Infrared Spectroscopy of Catalytically Active Interfaces — •Moritz Thun[1,2], Johannes Dittloff[1,2], and Ian D. Sharp[1,2] — [1]Walter Schottky Institute, Technische Universität München, Germany — [2]Physics Department, TUM School of Natural Sciences, Technische Universität München, Germany

Electrocatalytic reduction of CO_2 to carbon-based fuels can help to reduce atmospheric greenhouse gas concentrations and provide chemical storage capacities for renewable energies. Copper-based electrocatalysts uniquely offer a tunable range of diverse products. However, understanding and controlling the underlying reaction mechanisms requires in situ probes of the catalyst interface under working conditions. To this end, surface enhanced infrared reflection absorption spectroscopy (SEIRAS) can be used to detect the absorption and desorption of molecules on catalytic interfaces in real time.

Here, we demonstrate a stable substrate with SEIRAS-active surface morphology comprising evaporated Ti/Au metal film on nanostructured black silicon, as confirmed by monitoring the monolayer adsorption of 4-methoxypyridine. Next, Ti/Cu on black silicon, stabilized against surface reconstruction by a thin Nafion layer, allowed us to detect adsorbed CO during CO_2 reduction. We aim to detect additional products of the CO_2 reduction on copper by improving material design, enabling its extension to time-resolved SEIRAS measurements. Furthermore, we are exploring the replacement of Au layers with sputtered metal nitrides on black silicon to achieve versatile SEIRAS activity.

O 32.16 Tue 18:00 Poster C

Interfacial behavior of Perchlorate and Sodium Perchlorate in Air/Water Interface at varying concentrations — •Christina Susan Abraham and Marialore Sulpizi — Department of Physics, Ruhr Universität Bochum, 44780 Bochum, Germany

The presence of polarizable anions at aqueous interfaces has important implications that are relevant to both technological and environmental processes. Consequently, there has been a growing interest in understanding the propensity of anions at the air/water interface. While the behavior of the anions at interfaces has been studied, the effect of increased anionic concentrations at the air/water interface is still not very clear. This becomes important particularly in the field of aqueous based electrolytes where higher concentrations would directly affect the voltage window. In this study we have investigated the interfacial behavior of HClO4 and NaClO4 salt solutions employing AIMD simulations. The concentrations explored in this investigation span a spectrum from low (1M) to high (12M). As the salt concentration increases, the ion*water interactions show significant changes, not only in comparison with the lower concentrations but also with respect to the bulk HClO4/NaClO4-water systems.

O 32.17 Tue 18:00 Poster C

Fourth-Generation High-Dimensional Neural Network Potentials for Molecular Chemistry in Solution. — •Djamil Abdelkader Adel Maouene[1,2], Moritz Richard Schäffer[1,2], Moritz Gubler[3], Stefan Goedecker[3], and Jörg Behler[1,2] — [1]Theoretische Chemie II, Ruhr-Universität Bochum, Germany — [2]Research Center Chemical Sciences and Sustainability, Research Alliance Ruhr, Germany — [3]Department Physik, Universität Basel, Switzerland

Machine learning has found many applications in the fields of chemistry and materials science, and high-dimensional neural network potentials (HDNNPs) have become an accurate tool to represent the multi-dimensional potential energy surface in atomistic simulations.

Here, we compare the performance of two types of HDNNPs, i.e., 2G-HDNNPs and 4G-HDNNPs, in the description of organic molecules in aqueous solution. While it has been shown for many systems that 2G-HDNNPs are well suited to represent local bonding as a function of the atomic environments, they are not applicable to systems in which long-range charge transfer is important. Such systems can be addressed by 4G-HDNNPs, in which the atomic charges depend on structural or electronic changes even very far away in the system. Using typical organic molecules, the performance of both approaches is illustrated.

O 33: Poster: Supported Nanoclusters and Catalysis

Time: Tuesday 18:00–20:00 Location: Poster C

O 33.1 Tue 18:00 Poster C

HR-XPS Study on the Adsorption of CO on Graphene-Supported Co Clusters — •Natalie Waleska-Wellnhofer[1], Fabian Düll[1], Udo Bauer[1], Phillip Bachmann[1], Johann Steinhauer[1], and Christian Papp[1,2] — [1]Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany — [2]Freie Universität Berlin, Germany

Cobalt plays an important role as catalytic active material for the industrial production of hydrocarbons by Fischer-Tropsch synthesis. To overcome the material gap, we investigated graphene-supported Co clusters as model catalyst by HR-XPS to gather more information about the catalysts' properties. Using CO as a probe molecule, we were able to determine the available adsorption sites on the as-prepared clusters which are edge/top and bridge/hollow sites. The desorption temperatures of CO from these sites were determined by temperature-programmed XPS to be 360 and 240 K, respectively. Additionally, CO dissociation was observed. The dissociation products were used to study the influence of C and O on the reactivity of the clusters. For the precovered clusters, no CO adsorption at edge sites was observed due to site blocking. Furthermore, no CO dissociation was found. Thus, the edge sites are determined to be the most active sites. Moreover, the desorption temperature of CO decreased as a result of a change in the adsorption energy on the precovered clusters.

We thank Helmholtz-Zentrum Berlin for allocation of beamtime and BESSY II staff for support during beamtime. This work was funded by the DFG within SFB 953.

O 33.2 Tue 18:00 Poster C

Photocatalytic Alcohol Reforming on Platinum Cluster Co-Catalyst Loaded Titania(110) — •Anna Lemperle, Philip Petzoldt, Lucia Mengel, Martin Tschurl, and Ueli Heiz — Chair of Physical Chemistry, School of Natural Sciences & Catalysis Research Center, Technische Universität München, Lichtenbergstr. 4, 85748 Garching, Germany

Sunlight is a widely abundant resource, whose potential applications have come into focus in the face of environmental challenges. The field of photocatalysis aims to use light to power chemical reactions, thereby storing its energy inside chemical bonds. However, state of the art catalysts are limited in their performance. To systematically optimise the efficiency of photocatalysts, fundamental understanding of reaction mechanisms is essential. Only well-defined model systems allow to trace back catalytic results to distinct catalyst features and elucidate individual reaction steps. Alcohol reforming on metal loaded semiconductor single crystals under ultra-high vacuum conditions represents an ideal model reaction to enable such mechanistic studies. Encapsulation of clusters with a metal oxide overlayer caused by strong metal-support interactions are of particular interest, as it affects both turnover-frequency and selectivity of the catalyst.

In this poster, mechanistic insights gained from photocatalytic conversion of methanol over platinum cluster loaded $TiO_2(110)$ single crystals will be presented. In particular, we will focus on the effects of reductive heating on the co-catalyst and support demonstrating the importance of a profound understanding of their interplay.

O 33.3 Tue 18:00 Poster C

Investigation of highly efficient black titania nanotube photocatalyst by soft X-ray spectroscopy — •Hesham Hamad[1] and Giuliana Aquilanti[2] — [1]Fabrication Technology Research Department, Advanced Technology and New Materials Research Institute (ATNMRI), City of Scientific Research and Technological Applications (SRTA-City), New Borg El-Arab City, 21934, Alexandria, Egypt — [2]Elettra – Sincrotrone Trieste, s.s. 14 - km 163,5 in AREA Science Park, 34149 Basovizza, Trieste, Italy

The recent discovery of black TiO2 nanoparticles with enhancing solar absorption will trigger an explosion of interest; but black TiO2 nanoparticles remain a mystery. Here we elucidate more properties and try to understand the inner workings of black TiO2 nanotubes with hydrogenated disorders in a surface layer surrounding a crystalline core. In this work, TiO2 nanotubes (TNT) and its black hydrogenated TiO2 nanotubes (BTNT) were synthesized and investigated by characterization techniques. The merit of this project is to investigate the prepared synchrotron-based X-ray absorption fine structures (XAFS). It probed the unoccupied and occupied molecular orbitals of densities of states for O 2p and Ti 3d hybrid orbital characteristics, respectively. The reduction behavior and electronic and crystalline structure of a series of TNT and BTNT as photocatalysts will be investigated by using synchrotron-based XAFS. These techniques provide novel opportunities for tackling the structure and the dynamics of chemical and physical systems in solution.

O 33.4 Tue 18:00 Poster C

Fluorescence Carbon dots (CQDs) produced via hydrothermal carbonization incorporated in Graphene Oxide layers — •Daniel Saavedra[1], Marcelo A. Cisternas[2], Donovan E. Diaz-Droguett[1], and Ulrich G. Volkmann[1] — [1]Instituto de Física, Pontificia Universidad Católica de Chile, Santiago, Chile — [2]Escuela de Ingeniería Industrial, Universidad de Valparaíso, Santiago, Chile

Carbon quantum dots (CQDs) have received increasing attention due to their excellent photostability, low toxicity, tunable fluorescence performance [1], high extinction coefficient and brightness. In this study, carbon dots were prepared by carbonization of chitosan via hydrothermal carbonization method [2]. To improve the stability, graphene oxide synthesized by the Hummer method was incorporated. The obtained CQDs were characterized by various techniques including FTIR, AFM, TEM, UV-Vis and fluorescence spectroscopy. The results obtained are aimed at incorporating carbon dots into nanostructured surfaces to enhance hydrogen production by photocatalytic water splitting. Acknowledgements: ANID Ph.D. Fellowship (DS). References: [1] Wu, Q, et al. (2023), Nano Research, 16(2), 1835-1845. [2] Zattar, A. P. P., et al. (2021), Nanotubes and Carbon Nanostructures, 29(6), 414-422.

O 33.5 Tue 18:00 Poster C

In situ observation of Pt encapsulation in powder and single crystalline model catalysts — •Mohammad Salehi[1], Florian Kraushofer[1], Sebastian Kaiser[1], Matthias Krinninger[1], Johanna Reich[1], Virginia Pérez-Dieste[2], Friedrich Esch[1], and Barbara A. J. Lechner[1] — [1]Technical University of Munich, Garching, Germany — [2]ALBA Synchrotron Light Source, Barcelona, Spain

The activity of catalysts supported on reducible oxides is frequently altered significantly at high temperatures due to the strong metal support interaction (SMSI), which leads to an encapsulation layer surrounding the noble metal particles, changing the available active sites. Nevertheless, the effects of oxidizing and reducing treatments on the encapsulation layer under high pressures remain controversial, as ultrahigh vacuum (UHV) analyses often fail to fully understand the intricate nature of these systems. To comprehensively apprehend the atomic-scale structural dynamics and the process of active site formation, it is imperative to employ in situ or operando measurements. In this study, we utilize near-ambient pressure X-ray photoelectron spectroscopy (NAP-XPS) and near-edge X-ray absorption fine structure (NEXAFS) techniques to conduct a comparative analysis between rutile TiO_2 powder catalysts and rutile $TiO_2(110)$ single crystal samples concerning Pt encapsulation in oxygen. We find that on single crystals, Pt encapsulation dynamics depend both on the reduction state of the sample and on the Pt loading. Powder samples behave more similarly to near-stoichiometric crystals than strongly reduced ones but also show qualitative distinctions

O 33.6 Tue 18:00 Poster C

Operando ambient pressure HAXPES studies of Cu/ZnO(10-14) and Cu/ZnO(000-1) model catalysts for methanol synthesis — •Heshmat Noei[1], Robert Gleissner[1], Pauline Schütt[2], Michael Wagstaffe[1], Christopher Goodwin[3], Markus Soldemo[3], Mikhail Shipilin[3], Patrick Lömker[3], Christoph Schlueter[1], Peter Amann[3], Gregor Feldbauer[4], Anders Nilsson[3], and Andreas Stierle[1] — [1]Deutsches Elektronen-Synchrotron (DESY), Hamburg, Germany — [2]Universität Hamburg Department of Physics, Hamburg, Germany — [3]Stockholm Universitet, Department of Physics, Sweden — [4]Institute of Advanced Ceramics, TU Hamburg, Germany

The vicinal ZnO(10-14) facet recently gained scientific interest for its high density of surface steps and outstanding stability [1]. It has the lowest formation energy in comparison with other mixed-terminated facets such as (10-10) or (11-20) and therefor can be expected to play a distinct role in Cu/ZnO/Al2O3 powder catalysts [2]. We investigate the growth and morphology of the Cu nano particles on the vicinal ZnO surface under UHV conditions. Ambient Pressure XPS studies reveal the higher abundance of reaction intermediates for Cu/ZnO(10-14) in comparison to the Cu/ZnO(000-1) system on the surface of these model catalysts. Switching from CO/H2 over CO/CO2/H2 to CO2/H2 conditions and vice versa revealed complete reversibility. References [1] H. Zheng, Phys. Rev. Lett. 2013, 111, 086101. [2] K.S. Chan, Appl. Phys. Lett. 2015, 106, 212102.

O 33.7 Tue 18:00 Poster C

Advancing Single-Atom Catalysis: Developing a New Apparatus for Near-Ambient Pressure Applications — •Johannes Filzmoser, Adam Lagin, Jiri Pavelec, Ulrike Diebold, Michael Schmid, and Gareth S. Parkinson — TU Wien - Institute of Applied Physics - Wiedner Hauptstraße 8-10, 1040 Vienna, Austria

Single-atom catalysts (SAC) are much-studied in surface science due to their great potential in heterogeneous catalysis. Model systems consisting of metal adatoms on single-crystal metal-oxide surfaces are being investigated in ultra-high vacuum (UHV) to understand the fundamentals of SAC. A step towards industrial SAC systems is an examination of idealized model systems under more realistic conditions, i.e., at elevated pressures and temperatures. This work aims to develop a UHV-compatible reaction cell, enabling sample exposure to gases at about 1 mbar pressure and temperatures up to 300°C. The composition of the exhaust gas from the reactor is quantitatively analyzed by mass spectrometry. Due to the low number of active sites on model SAC, measuring turnover frequencies presents a significant challenge. Therefore, we are investigating methods to optimize detection techniques.

O 33.8 Tue 18:00 Poster C

Computational Modeling of Metal-Organic Frameworks as Heterogeneous Catalysts for Biomass Conversion — •Manuel A Ortuño[1], Thanh-Hiep T. Le[1], David Ferro-Costas[1,2], and Antonio Fernandez-Ramos[1,2] — [1]Centro Singular de Investigación en Química Biolóxica e Materiais Moleculares (CIQUS), Universidade de Santiago de Compostela, 15782 Santiago de Compostela, Spain — [2]Departamento de Química Física, Facultade de Química, Universidade de Santiago de Compostela, 15782 Santiago de Compostela, Spain

Metal-organic frameworks (MOFs) have emerged as promising heterogeneous catalysts due to well-defined structure, surface area, and tunable pore size. In particular, MOFs with Zr-based nodes are stable and therefore used as heterogeneous catalysts for a myriad of reactions.

Here we focus on UiO-66, formed by Zr-oxide nodes and 1,4-benzenedicarboxylate linkers, for the catalytic conversion of methyl levulinate into γ-valerolactone. We computed cluster and periodic models at DFT level to unravel the reaction mechanism. The simulations supported a defective node as active site, where the reaction takes place via three main steps: hydrogen transfer, nucleophilic attack, and elimination. Further studies suggested that the presence of Ce in the node might improve catalytic performance. Later, we found that both cluster and periodic models behave similarly, and the choice of density functional does not affect the rate-determining step but it does significantly change the Gibbs energy barriers.

O 33.9 Tue 18:00 Poster C

In situ XPS and SXAS study on CO adsorption on a FeNC catalyst — •Benedikt P. Klein[1,3], Beomgyun Jeong[1], Hafiz Ghulam Abbas[2], Geunsu Bae[2], Adith R. Velmurugan[2], Chang Hyuck Choi[4], Geonhwa Kim[5], Dong Woo Kim[5], Ki-jeong Kim[5], Byeong Jun Cha[6], Young Dok Kim[6], Frederic Jaouen[7], Reinhard J. Maurer[3], and Stefan Ringe[2] — [1]Korea Basic Science Institute, Daejeon, ROK — [2]Korea University, Seoul, ROK — [3]University of Warwick, Coventry, UK — [4]POSTECH, Pohang, ROK — [5]PAL, Pohang, ROK — [6]Sungkyunkwan University, Suwon, ROK — [7]University of Montpellier, France

A reliable way to quantify the number of active sites in a catalyst is crucial to evaluate its performance. One option to achieve this is to utilize strongly adsorbing gas molecules as a probe to identify the catalytically active sites. Once the molecules are adsorbed on the active sites, their surface density can be determined by using spectroscopic techniques. For this approach it is necessary to identify the probe molecules according to their spectroscopic features, a task much helped by the computational simulation of spectra. Here, we present the *in situ* XPS and SXAS study for the adsorption of CO on an iron-nitrogen-carbon (FeNC) catalyst under near-ambient gas pressure. Using the experimental spectroscopic data combined with state-of-the-art DFT based spectroscopy simulations, we determine the active site density for the electrochemical oxygen reduction reaction of this catalyst.

O 33.10 Tue 18:00 Poster C

Oxide formation and oxide/metal interaction in CeO_x/Ni(111) — •Dominic Guttmann, Björn Riedel, Raquel Sánchez-Barquilla, Carlos Morales, and Jan Ingo Flege — Applied Physics and Semiconductor Spectroscopy, Brandenburg University of Technology Cottbus-Senftenberg, Cottbus 03046, Germany

Ni/ceria catalysts exhibit a high activity for methane to methanol conversion, making them very promising for applications within a sustainable economy. Possibly, their activity may be strongly enhanced due to the facile exchange between Ce^{4+} and Ce^{3+} states, with the latter likely responsible for activating O-H and C-H bonds. Here, we aim to unravel the complex metal-oxide interactions in the inverse CeO_x/Ni(111) system under oxidizing and reducing environments. Using low-energy electron diffraction (LEED) we find that the CeO_x(111) grown by reactive molecular beam epitaxy preferentially aligns with the main directions of the Ni(111) substrate or is azimuthally rotated by ±10°. By using X-ray photoelectron spectroscopy (XPS) and ultraviolet photoelectron spectroscopy (UPS), we find that less NiO is formed during deposition of CeO_x than when it is held without ceria at the same conditions (O_2 partial pressure and temperature). Finally, we observe a complex behavior of the cerium and nickel oxidation states when exposing the system to O_2 or H_2 atmospheres.

O 34: Poster: Surface Dynamics & Electron-Driven Processes

Time: Tuesday 18:00–20:00 Location: Poster C

O 34.1 Tue 18:00 Poster C

Targeted growth of metastable interface polymorphs — •Simon B. Hollweger, Anna Werkovits, and Oliver T. Hofmann — Institute of Solid State Physics, TU Graz, Austria

Growing a specific interface polymorph of organic molecules on inorganic substrates is far from trivial. The target structure is usually not the one that is thermodynamically most favorable, but rather an energetically higher-lying, metastable structure. It is primarily unclear how the growth conditions need to be tuned to maximize the yield of a higher-energy polymorph. These conditions are usually explored experimentally by a tedious trial-and-error routine. To avoid this obstacle we theoretically investigate the growth behavior of such systems. We discuss a class of systems where we can start from a lower-energy polymorph and through systematic changes of pressure and temperature induce a structural change to the desired target structure. For that two requirements need to be fulfilled, (a) the metastable target structure has a similar packing density as the initial structure and (b) an 'elevator' polymorph exists that has a higher packing density and is thermodynamically accessible at lower temperatures. To explore in which range of substrate-molecule and molecule-molecule interactions the necessary conditions are fulfilled we employ a combination of kinetic Monte Carlo simulations and ab-initio thermodynamics. This allows us to formulate structure-to-property relationships. For actually determining 'growth recipes' for a specific target structure we utilize Deep Reinforcement Learning combined with kinetic Monte Carlo simulations to obtain optimal temperature and pressure curves.

O 34.2 Tue 18:00 Poster C

Exploring the Commensurate Charge Density Wave in Tantalum Disulfide: Insights from Scanning Tunneling Microscopy and Spectroscopy — •Georg A. Traeger[1], Jan-Niclas Schmidt[1], Kai Rossnagel[3,4], Murat Sivis[1,2], Claus Ropers[1,2], and Martin Wenderoth[1] — [1]University of Göttingen, IV. Physik, Fredrich-Hund-Platz 1, 37077 Göttingen, Germany — [2]Department of Ultrafast Dynamics, Max Planck Institute for Multidisciplinary Sciences, 37077 Göttingen, Germany — [3]Institute for Experimental and Applied Physics, University of Kiel, 24098 Kiel, Germany — [4]Ruprecht Haensel Laboratory, Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany

In recent years, transition-metal dichalcogenides have attracted great interest, owing to a wide range of correlated and tunable physical phenomena. 1T-tantalum disulfide is a widely-studied model system, which exhibits a series of charge-density wave states and a strong influence of stacking on its electronic properties. Here, we present a study of the low-temperature commensurate charge density wave and its metastable states by means of Scanning Tunneling Microscopy (STM) and Spectroscopy (STS). We investigate the defect- and laser-induced creation and manipulation of metastable states, emphasizing the roles of translational phase boundaries and heterochirality.

O 34.3 Tue 18:00 Poster C

Invariant embedding approach to secondary electron emission from surfaces — •Franz Xaver Bronold and Felix Willert — Institut für Physik, Universität Greifswald, 17489 Greifswald, Germany

Secondary electron emission from the walls confining a gas discharge is an important surface process in low-temperature plasma physics. It affects, for instance, the operation modii of barrier discharges, Hall thrusters, and divertor plasmas in fusion devices. Little is however known quantitatively about the process because it typically occurs at energies below 50 eV which are hard to access experimentally. In this contribution, we present a theoretical scheme for calculating the secondary electron emission yield in this energy range. It is based on applying the invariant embedding principle, originally developed for calculating the albedo of planetary atmospheres, to the electron surface scattering kernel characterizing the interaction of a primary electron with a surface [1]. To illustrate the approach we apply it to polycrystalline silicon and germanium surfaces using a semiempirical randium-jellium model containing the Schottky barrier, impact ionization across the band gap as well as scattering on phonons, defects, and ion cores. The emission yields we obtain by solving the full nonlinear embedding equation numerically without an approximate decoupling of angle and energy variables are in satisfactory agreement with measured data to also support the use of the scattering kernel in the boundary condition of the electron Boltzmann equation of a plasma simulation.
[1] F. X. Bronold and F. Willert, arXiv:2309.00534.

O 34.4 Tue 18:00 Poster C

Power discontinuity and shift of the energy onset of a molecular de-bromination reaction induced by hot-electron tunneling — Ana Barragan[1], Roberto Robles[2], Nicolas Lorente[2,4], and •Lucia Vitali[1,2,3,4] — [1]Advanced Polymers and Materials: Physics, Chemistry and Technology, Chemistry Faculty (UPV/EHU) — [2]Centro de Física de Materiales CFM/MPC(CSIC-UPV/EHU), — [3]Ikerbasque Research Foundation for Science, Bilbao 48009, Spain; — [4]Donostia International Physics Center (DIPC), 20018 San Sebastián,

Understanding the mechanism of molecular dissociation under applied bias is a fundamental requirement to progress in (electro)- catalysis as well as in (opto)-electronics. The working conditions of a molecular-based device and the stability of chemical bonds can be addressed in metal-organic junctions by injecting electrons in tunneling conditions. Here, we have correlated the energy of debromination of an aryl group with its density of states in a self-assembled dimeric structure of 4'-bromo-4-mercaptobiphenyl adsorbed on a Au(111) surface. We have observed that the electron-energy range where the molecule is chemically stable can be extended, shifting the bias threshold for the rupture of the -C-Br bond continuously from about 2.4 to 4.4 V by changing the electron current. Correspondingly, the power needed for the dissociation drops sharply at 3.6 V, identifying different reaction regimes and the contribution of different molecular resonance states.

O 34.5 Tue 18:00 Poster C

Investigation of Ballistic Transport on Black Phosphorus with MONA — •Manuel Seitz, Andreas Christ, Emilian Eisermann, Patrick Härtl, Markus Leisegang, and Matthias Bode — Physikalisches Institut, Experimentelle Physik II, Julius-Maximilians-Universität Würzburg, Am Hubland, 97074 Würzburg, Germany

Due to the ever-increasing miniaturization of electronic components, we are approaching a frontier where 2D-materials are promising for nanoscale electronic devices. One auspicious material is the semiconductor black phosphorus (BP) [1]. BP exhibits an anisotropic surface structure and shows a band gap that is influenced by the number of layers [1, 2]. The strong anisotropy as well as the possibility to tune the band gap are properties which may strongly impact the charge transport.
In this project, we aim to investigate surface transport in the ballistic regime of BP using the molecular nanoprobe (MONA) technique[4]. We thoroughly evaluated various cleaving techniques and investigated the resulting surfaces. Furthermore, single adatoms with bistable binding configurations are characterized [5]. In a proof-of-principle experiment we show that these bistable atoms can be used as detectors for MONA measurements.
[1] X. Ling *et al.*, Proc. Natl. Acad. Sci. **112**, 4523 (2015)
[2] J. Kim *et al.*, Science **349**, 723 (2015)
[3] Y. Du *et al.*, J. Appl. Phys. **107**, 093718 (2010)
[4] M. Leisegang *et al.*, Nano Lett. **18**, 2165 (2018)
[5] B. Kiraly *et al.*, Nat. Commun. **9**, 3904 (2018)

O 35: Poster: Surface Reactions

Time: Tuesday 18:00–20:00 Location: Poster C

O 35.1 Tue 18:00 Poster C

Surface Science Investigations of Oxanorbornadiene/ Oxaquadricyclane Ester Derivatives as MOST Systems on Pt(111) — •Felix Hemauer[1], Valentin Schwaab[1], Eva Marie Freiberger[1], Natalie J. Waleska-Wellnhofer[1], Daniel Krappmann[1], Hans-Peter Steinrück[1], and Christian Papp[2] — [1]Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany — [2]Freie Universität Berlin, Germany

The harvesting and storage of solar power is feasible in a chemical manner with so-called molecular solar thermal (MOST) systems. In one-photon one-molecule processes, the conversion of the energy-lean norbornadiene (NBD) into its strained quadricyclane (QC) isomer takes place upon irradiation. On demand, the energy-releasing back reaction is triggered. For a sustainable storage and release cycle, the catalytic release needs to occur without loss of the MOST molecules. Since the methylene bridgehead group was found to be the weakest link, the surface chemistry of derivatized hetero-NBD/QC pairs was assessed on Pt(111). Specifically, synchrotron radiation-based XPS experiments were performed on 2,3 bis(methylester)-oxa-NBD/QC derivatives and its extended 2,3-bis(benzylester) substitution. Next to qualitative information on adsorption motifs, respective reaction pathways were deduced by means of temperature-programmed measurements. That is, the energy release in the cycloreversion reaction from the energy-rich isomers was studied, as well as competing desorption and decomposition steps determined. The DFG (392607742) supported this work. We thank HZB for allocation of synchrotron radiation beamtime.

O 35.2 Tue 18:00 Poster C

Non-thermal plasma modification of unsized carbon fibers — •Torben Noah Gärtig, Viktor Udachin, and Wolfgang Maus-Friedrichs — Clausthal Center of Material Technology, Clausthal University of Technology, Clausthal-Zellerfeld, Germany

Bonding of carbon fibers to the resin matrix for carbon-fiber-reinforced polymers is mostly achieved with sizing materials. An alternative treatment by oxidizing fibers with a non-thermal dielectric barrier discharge (DBD) plasma in air or oxygen atmospheres may provide a more environmentally friendly approach, which can simplify recycling of fibers and exclude the usage of sizing substances. In the present study, unsized carbon fibers were plasma treated in air and oxygen atmospheres at 1000 hPa and 25 °C with treatment times up to 10 minutes. The fiber surface before and after treatment was analyzed with X-Ray Photoelectron spectroscopy as well as microscopic methods like atomic force microscopy (AFM) and confocal laser scanning microscopy (CLSM). The plasma was characterized with optical emission spectroscopy (OES). It was found that air DBD plasma under ideal power parameters can oxidize fibers better than oxygen DBD plasma due to nitrogen reactive species aiding ozone generation. The observed oxidation does not result in significant morphology changes.

O 35.3 Tue 18:00 Poster C

Model Catalytic Studies on the Thermal Dehydrogenation of Alcohol-Based Liquid Organic Hydrogen Carrier Systems — •Valentin Schwaab[1], Felix Hemauer[1], Natalie J. Waleska-Wellnhofer[1], Eva Marie Freiberger[1], Hans-Peter Steinrück[1], and Christian Papp[2] — [1]Friedrich-Alexander-Universität Erlangen-Nürnberg — [2]Freie Universität Berlin

The transition to a sustainable, renewable-based energy system requires novel energy storage technologies. Hydrogen production through electrolysis is a promising approach, however, under standard conditions the gas has a low volumetric energy density and is difficult to handle. So-called liquid organic hydrogen carriers (LOHCs) enable safe storage of hydrogen at high volumetric energy densities through the reversible hydrogenation of organic compounds. Alcohols are particularly interesting systems, as they often exhibit exceptionally low dehydrogenation temperatures in their reaction to the respective hydrogen-lean carbonyl compounds.

We investigated the dehydrogenation reactions of two alcohol-based LOHC pairs, namely, benzaldehyde/cyclohexylmethanol and acetophenone/1-cyclohexylethanol, on a Pt(111) model catalyst. The adsorption and the thermal evolution of the compounds was monitored in situ in synchrotron radiation photoelectron spectroscopy (SRPES) experiments at BESSY II, Helmholtz-Zentrum Berlin (HZB).

We acknowledge financial support by the Bavarian Ministry of Economic Affairs, Regional Development and Energy, and by the DFG (419654270). We thank HZB for the allocation of beamtime.

O 35.4 Tue 18:00 Poster C

Grazing incident extended X-ray absorption fine structure on liquids at Beamline P64 at Petra III — •Lukas Voss, Dirk Lützenkirchen-Hecht, Frederic Braun, Franz Eckelt, and Carlo Schneider — Bergische Universität Wuppertal, NRW

The existing setup for grazing incidence X-ray absorption spectroscopy at Beamline P64, PETRA III, has been expanded to accommodate liquid samples. This enhancement involved the integration of a mirror device in front of the reflectometer, redirecting the X-ray beam downward onto the liquid sample surface. The adjustable mirror facilitates variations in both height and angle, enabling diverse incident angles on the surface. Leveraging this improved configuration, our investigation focused on the formation of germanium nanoparticles on the surface of deionized water. The high-speed scanning monochromator at P64 allowed for the rapid acquisition of complete EXAFS spectra within fractions of a second. Consequently, we successfully captured the exceedingly swift reaction between germanium(IV)-isopropoxide and water in situ. To further advance our capabilities, a specialized liquid sample chamber is currently under construction. This chamber aims to facilitate the study of surface reactions in liquids at controlled gas interfaces and temperatures. Its versatile design enables the detection of the reflected X-ray beam for Refl-EXAFS, while also facilitating fluorescence detection using a PIPS detector. This integrated approach enhances our ability to comprehensively investigate and analyze diverse liquid systems, such as e.g. Langmuir-Blodgett-films, providing valuable insights into surface interactions.

O 35.5 Tue 18:00 Poster C

Oxide reduction mechanisms on Cu and Fe surfaces under non-thermal plasma — •Viktor Udachin, Fabien Krebs, Oliver Höfft, Lienhard Wegewitz, and Wolfgang Maus-Friedrichs — Clausthal University of Technology, Clausthal-Zellerfeld, Germany

Oxide layers on metal surfaces usually limit the performance of materials in different industrial applications like coating or bonding. Therefore, the ability of rapid reduction of such oxides is of a big interest. Whereas methods as chemical reduction usually require complex procedures to obtain metallic surfaces, non-thermal plasmas have been shown as a quick and environmental-friendly method for surface deoxidation without notable morphological changes. Nevertheless, no detailed information on mechanisms of reduction on different metal systems is accessible. In the current work, the mechanisms of surface oxide reduction under non-thermal dielectric barrier discharge (DBD) Ar/H2 plasma were investigated. We performed depth profile characterization of oxidized Cu and Fe surfaces before and after treatment via angle resolved X-ray photoelectron spectroscopy (AR-XPS). Surface topography of Cu was studied in vacuum with atomic force microscopy (AFM), scanning tunneling microscopy (STM) as well as spectroscopy (STS) directly after oxide reduction, showing individual steps of surface deoxidation. Funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) - Project-ID 394563137 - SFB 1368

O 35.6 Tue 18:00 Poster C

CO$_2$ reduction by trapped electrons at the NH$_3$/Cu(111) interface. — •Maya Heine, Lukas Gierster, and Julia Stähler — Humboldt Universität zu Berlin

When electrons are photoinjected from a metal substrate into D$_2$O or NH$_3$ adlayers they can become trapped and exhibit lifetimes up to minutes. The energetics and dynamics of such trapped electrons have been studied across picoseconds to minute timescales using time-resolved two-photon photoelectron spectroscopy and are well understood [1]. An exciting next step is to investigate the interactions of such electrons with molecules. For example, trapped electrons have already been shown to participate in the water splitting reaction at the amorphous solid water interface [2]. We investigate the interaction of CO$_2$ with the crystalline NH$_3$/Cu(111) interface, as much remains to be understood regarding the timescales and energetics of possible reactions of this important molecule. We show first results where we observe quenching of the trapped electron signal and work function modifications upon CO$_2$ addition, suggesting activation of CO$_2$ by the trapped electrons.

[1] Stähler et al. *Chemical Science 2, no. 5, 907-16* (2011)

[2] King et al. *The Journal of Physical Chemistry C 121, no. 13, 7379-86* (2017)

O 35.7 Tue 18:00 Poster C

Experimental investigation of GaInP(100) surfaces exposed to H2O — •David Ostheimer[1], Christian Dressler[2], Mohammad Amin Zare Pour[1], Sahar Shekarabi[1], Max Grossmann[3], Agnieszka Paszuk[1], and Thomas Hannappel[1] — [1]TU Ilmenau, Grundlagen von Energiematerialien, Ilmenau, Deutschland — [2]TU Ilmenau, Theoretische Festkörperphysik, Ilmenau, Deutschland — [3]TU Ilmenau, Theoretische Physik I, Ilmenau, Deutschland

Despite GaInP being employed in photoelectrochemical devices with world-record efficiencies, stability remains a significant challenge. A detailed understanding of the reactions at the semiconductor/electrolyte heterointerface is crucial to tailor the semiconductor surface appropriately to avoid losses of the photogenerated charge carriers and to reduce corrosion. As a first step, we investigate the interaction of phosphorous-rich (P-rich) and group-III-rich-GaInP(100) surfaces, with water vapor. Our analysis reveals that the initial surface reconstruction of GaInP(100) has a significant effect on the interaction with water. The P-rich GaInP(100) surface showed minimal alteration following exposure to 300 kL of water vapor. In contrast, the group-III-rich surface exhibits no RAS anisotropy after exposure. XPS measurements indicate a reduction of In-In- bonds, and that the initial pathway toward oxidation mainly involves the hydroxylation of these dimers, which subsequently leads to a formation of InOx species. Both findings could be further confirmed by ab-initio Born-Oppenheimer molecular dynamic simulations with 0.5 fs timesteps and 30 ps total simulation time for each system.

O 36: Poster: Ultrafast Electron Dynamics at Surfaces and Interfaces

Time: Tuesday 18:00–20:00 Location: Poster C

O 36.1 Tue 18:00 Poster C

Differences in the electron dynamics of inequivalent (001) surfaces of *Td*-WTe$_2$: top and bottom or topological trivial and topological non-trivial? — •Klaas Opitz[1], Hauke Beyer[1], Eduard Moos[1], Kai Rossnagel[1,2,3], and Michael Bauer[1,2] — [1]Institute of Experimental and Applied Physics, Kiel University, 24098 Kiel, Germany — [2]Kiel Nano, Surface and Interface Science KiNSIS, Kiel University, 24118 Kiel, Germany — [3]Deutsches Elektronen-Synchrotron DESY, Ruprecht Haensel Lab, 22607 Hamburg, Germany

ARPES spectra of [100]-oriented *Td*-WTe$_2$ show clear differences, which in the past have been associated either with different topological phases [1] or alternatively interpreted as a signature for the small but finite structural differences of the (001) and (00$\bar{1}$) surface resulting from the broken inversion symmetry of the material [2]. In this contribution we will present time-resolved ARPES data of *Td*-WTe$_2$ that clearly hint to distinct differences also in the electron dynamics for these two surfaces. We will discuss our results under consideration of the different interpretations of the ARPES data.

[1] Y. Wu *et al.*, Phys. Rev. B. **94**, 121113(R) (2016).

[2] Y. Wan *et al.*, Phys. Rev. B. **105**, 085421 (2022).

O 36.2 Tue 18:00 Poster C

Second harmonic spectroscopy of Fe-porphyrin/Cu(001) interfaces — •Newsha Vesalimahmoud, Mahendra Kabbinahithlu, Ping Zhou, Uwe Bovensiepen, and Andrea Eschenlohr — University of Duisburg-Essen, Faculty of Physics and CENIDE, Lotharstr. 1, 47057 Duisburg, Germany

Hybrid systems of metallo-porphyrin/metal interfaces, which are known as spinterfaces, are promising candidates to control the spin-dependent transport in spintronic devices [1]. This study focuses on the iron porphyrin molecules (FeOEP)/Cu(001) interface using interface-sensitive time-resolved second harmonic generation (SHG) spectroscopy. We analyze the polarization and wavelength dependent SHG for different molecular adsorbate thicknesses on Cu(001) using a fundamental beam in the range of 500 - 700 nm. Polarization dependent measurements show a higher p-P SHG yield than s-P SHG, as the intrinsic absolute value of $|\chi^{(2)}_{zzz}|$ is much larger than $|\chi^{(2)}_{zxx}|$. Moreover, SHG spectra show an increasing intensity up to a maximum at 2.3 eV for Cu(001) due to one ω resonance. After adsorption of 2 monolayers (ML) and 5ML FeOEP/Cu(001), the spectral dependence remains similar to the Cu(001) surface except for an enhancement at 2.25 eV for 5ML FeOEP/Cu(001). We attribute this enhancement to the energetic position of the lowest unoccupied molecular orbital (LUMO) at the FeOEP/Cu(001) interface. Moreover, we discuss the electron dynamics analyzed through pump-probe SHG at on- and off-resonant photon energies.

[1] Wende, H., et al., Nat. Mater, **6**, 516 (2007).

O 36.3 Tue 18:00 Poster C

Transient Absorption Spectroscopy of NiO — •Mahendra Kabbinahithlu[1], Tobias Lojewski[1], Sergey Kovalenko[2], Nico Rothenbach[1], Katharina Ollefs[1], Heiko Wende[1], Uwe Bovensiepen[1], Julia Stähler[2], and Andrea Eschenlohr[1] — [1]Universität Duisburg-Essen, Fakultät für Physik und Center for Nanointegration (CENIDE), Lotharstraße 1, 47057 Duisburg, Germany — [2]Humboldt-Universität zu Berlin, Institut für Chemie, Brook-Taylor-Straße 2, 12489 Berlin, Germany

In nickel oxide (NiO), a correlated transition-metal oxide, the strong electron-electron repulsion splits the d-bands into an occupied lower Hubbard band (LHB) and an unoccupied upper Hubbard band (UHB). The presence of additional oxygen p-bands from adjacent sites, located between the Hubbard bands makes NiO a charge-transfer insulator. We present excited state dynamics in NiO thin films via time-resolved optical spectroscopy. NiO is pumped above the band gap with 3.98 eV photons and the transient absorption spectrum is probed using a time-delayed supercontinuum. The time-resolved absorption spectrum shows within the first 500 fs pump-induced changes at 2.2 eV, 3.2 eV and 4.3 eV. The negative change represents the reduced transitions from the bleached ground state and the positive changes represent the increased transitions from the photo-excited state. Around 2 ps, relaxation from the photo-excited state leads to emergence of a fluence dependent positive feature. We discuss this feature as transitions from polarons and shallow defect states lying below the UHB edge.

O 36.4 Tue 18:00 Poster C

Laser-induced ultrafast electron dynamics based on a two-band model — •Stephanie Roden, Tobias Held, Sebastian T. Weber, and Bärbel Rethfeld — Department of Physics and Research Center OPTIMAS, RPTU, Kaiserslautern-Landau

After an ultrashort laser excitation, the energy of an optical laser pulse is absorbed by the electrons of a solid. The thermalization of the excited electron system and the relaxation with the phonons to a joint temperature can be calculated in a kinetic manner by coupled Boltzmann collision integrals.

Previous implementations are based on an effective one-band model for the electron system [1]. This approximation does not allow a distinction between intra- and interband relaxation within the electron system. However, a separate consideration of these relaxation processes can strongly influence calculated optical parameters [2] as well as the electron-phonon coupling [3]. In this work, we extend the existing one-band model to an energy-resolved two-band model for a thin gold film that distinguishes between free sp- and more localized d-electrons. We show selected results of the intertwined relaxation dynamics following an ultra-short laser pulse.

[1] B. Y. Mueller and B. Rethfeld, PRB 87, 035139 (2013)

[2] P. D. Ndione, S. T. Weber, D. O. Gericke and B. Rethfeld, Sci. Rep. 12, 4693 (2022)

[3] T. Held, S. T. Weber, and B. Rethfeld, arXiv:2308.01067 (2023)

O 36.5 Tue 18:00 Poster C

Coherent and incoherent ultrafast lattice dynamics in WTe_2 — •Hanqian Lu[1,2], Victoria Taylor[2], Hyein Jung[1,2], Jannik Malter[2], Ralph Ernstorfer[1,2], and William Windsor[1,2] — [1]Technische Universität Berlin — [2]Fritz Haber Institute of the Max Planck Society

WTe$_2$ is a layered transition metal dichalogenide, which has attracted significant attention in recent years, in particular due to its potential as a topological material and a polar metal. Here we study its response to ultrafast photoexcitation using femtosecond electron diffraction (FED). We observe both coherent and incoherent photoinduced lattice dynamics, in particular the long-lived 0.23 THz shear mode. We quantify these responses along different crystal axes, and present a microscopic model of atomic vibrations of all atoms in the unit cell.

O 36.6 Tue 18:00 Poster C

Time-resolved photoelectron spectroscopy of charge separation at the PTCDA/TiOPc heterointerface on Ag(111) — •Marcel Theilen, Alexa Adamkiewicz, Alexander Lerch, Robert Wallauer, and Ulrich Höfer — Philipps-University Marburg, Germany

A key element for charge separation in organic donor/acceptor heterostructures is the formation of charge transfer (CT) states at the interface between the donor and acceptor molecules. A promising model system for studying such excitonic states is the well-defined heterosystem of PTCDA/TiOPc/Ag(111), which we examine by means of time-resolved two-photon photoemission (2PPE). Based on previous 2PPE measurements, the charge carrier dynamics upon selective excitation of excitons in the molecular layers are dominated by transfer processes into the interface state (IS) between molecule and metal. Increasing the molecule-metal distance results in a more effective decoupling to the IS and a possible formation of CT excitons at the donor/acceptor interface. However, the previous 2PPE measurements couldn't access the population of the excited molecular states, which is possible by means of time-resolved photoemission orbital tomography (tr-POT), as it provides a direct insight into the momentum-space dynamics of the excited molecular states. Here we demonstrate our first measurements of the charge carrier dynamics it the 2 ML PTCDA/2 ML TiOPc heterosystem using tr-POT. We follow the momentum-space dynamics of the excited molecular states and see, according to the 2PPE measurements, a strong coupling to the IS between molecule and metal.

O 36.7 Tue 18:00 Poster C

Towards femtosecond momentum microscopy of field-effect gated transition metal dichalcogenides — •Bent van Wingerden, Jan Philipp Bange, Jonas Pöhls, Wiebke Bennecke, David Schmitt, Daniel Steil, Gijsbert Simon Matthijs Jansen, Thomas Weitz, Marcel Reutzel, and Stefan Mathias — 1. Physikalisches Institut, Georg-August Universität Göttingen, Göttingen, Germany

2D-materials have received great attention due to their astounding optical and electronic properties. Several degrees of freedom (DoF) result in extensive phase diagrams and reveal manifold possibilities for applications in spin-, valley- and optoelectronics. To yield applications in technology, control over these DoF has to be established. Our goal is to study the effect of charge carrier doping by field-effect gating on the band structure and ultrafast exciton dynamics of TMDs in time-resolved momentum microscopy. Previous experimental work visualized the impact of field-effect doping on the electronic band structure for graphene and WSe$_2$, utilizing static μARPES endstations at synchrotrons [Nguyen *et al.* Nature 572 (2019)]. Here, we will discuss our efforts to study gated 2D-materials with our laboratory based experiment that combines a time-of-flight momentum microscope with a high-repetition rate HHG-beamline [Keunecke *et al.*, Rev. Sci. Ins. 91, 063905 (2020)]. This approach will facilitate the study of ultrafast exciton dynamics and may lead to the discovery of novel relaxation dynamics [Schmitt *et al.*, Nature 608, 499 (2022)].

O 36.8 Tue 18:00 Poster C

Coherent Electron Dynamics Probed by Interferometrically Time- and Angle-Resolved Nonlinear Plasmoemission Spectroscopy with a Birefringent Delay-Line — •Pascal Dreher, Alexander Neuhaus, Michael Horn-von Hoegen, and Frank-J. Meyer zu Heringdorf — Faculty of Physics and Center for Nanointegration, Duisburg-Essen (CENIDE), University of Duisburg-Essen, 47048 Duisburg, Germany

The early optical response of a solid to an intense light field is governed by single-particle and collective excitations within the electron system. Due to the perturbation, changes in the electron populations occur, and coherent electronic polarizations are excited. Interferometrically time-resolved two-photon photoemission (ITR-2PPE) has been highly successful at disentangling the population and polarization dynamics in various material systems. If instead multi-photon transitions are utilized (ITR-mPPE) the multitude of involved electronic states and their possible interactions complicate the extraction of lifetimes and dephasing times. We combined a birefringent delay-line with nanofocusing of surface plasmon polaritons (SPPs) on flat metal surfaces to acquire phase-locked pulses with transient electric field strengths sufficient to coherently drive highly nonlinear electronic transitions. The excited electron dynamics is observed using time- and angle-resolved photoemission spectroscopy. We discuss modelling of the dynamics with Lindblad equations and comment on the extraction of lifetimes and dephasing times in this highly nonlinear setting.

O 36.9 Tue 18:00 Poster C

100 kHz HHG at 18 eV for time-resolved ARPES — •Sunil Dahiya[1], Hermann Erk[1], Aaron Diercks[1], Stephan Jauernik[1], and Michael Bauer[1,2] — [1]Institute of Experimental and Applied Physics, Kiel University, 24098 Kiel, Germany — [2]Kiel Nano, Surface and Interface Science KiNSIS, Kiel University, 24118 Kiel, Germany

Conventional time- and angle-resolved photoemission (trARPES) employs probe pulses of approximately 6 eV, providing only limited access to momentum space. To encompass a broader momentum space, covering the entire Brillouin zone in solids, higher photon energies are required. This objective is achieved through high harmonic generation (HHG) [1], generating photon pulses with energies in the range of 10 to several hundred electron volts.

In this presentation, we introduce a 100 kHz high-harmonic generation source for trARPES. The concept, including the laser system used, is based on a recently presented setup that produces vacuum ultraviolet (VUV) pulses at a photon energy of 11 eV [2]. In our experiment, we select the next higher harmonic (5th harmonic) of the spectrum at 18 eV photon energy. The HHG source is driven by the 3rd harmonic (350 nm, 0.02 mJ pulse energy) of a Yb:KGW amplifier (1038 nm, 190 fs) and is operated with Argon or Krypton. We demonstrate the performance of the source in initial ARPES measurements of various solid targets.

[1] T. Rohwer, et al., Nature 471, 490 (2011).
[2] C. Lee, et al, Rev. Sci. Instrum. 91, 043102 (2020).

O 36.10 Tue 18:00 Poster C

Dynamics of energy-resolved electron densities at the non-thermal stage in gold — •Markus Uehlein, Christopher Seibel, Tobias Held, Sebastian T. Weber, and Baerbel Rethfeld — Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau

During femtosecond laser excitation, the electron distribution in a metal is disturbed to a state far from equilibrium, i.e. far from a Fermi distribution. The resulting high-energy electrons (hot electrons), e.g., drive chemical reactions at surfaces and are utilized in biosensing methods. An important step for further studies is to understand how many electrons exist in a specific energy range and on what timescale. This is measurable for example with photoemission experiments.

We apply a kinetic model based on full Boltzmann collision integrals to trace dynamics of the electronic distribution [1]. Thereby, the different processes contributing to excitation and thermalization can be investigated independently. We study the temporal evolution of the electron densities in various energy ranges, pointing out the influence of primary and secondary electron generation [2]. Furthermore, we compare the microscopic description of the electron-electron scattering with a relaxation time approach and an extended temperature-based description.

[1] B.Y. Mueller, B. Rethfeld, Phys. Rev. B 87, 035139 (2013)
[2] C. Seibel, M. Uehlein *et al.*, J. Phys. Chem. C (2023)
DOI: 10.1021/acs.jpcc.3c04581

O 36.11 Tue 18:00 Poster C

Nonequilibrium optical properties in laser-excited noble metals — •Pascal D. Ndione[1], Tobias Held[1], Sebastian T. Weber[1], Dirk O. Gericke[2], and Baerbel Rethfeld[1] — [1]Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau — [2]CFSA, Department of Physics, University of Warwick

We study the electron density and optical responses in gold following excitation with XUV and visible light. We develop multiband rate equations that track the density response in each active electron band [1]. The rate equations also trace the energy content of the *sp*- and *d*-electrons and couple them to the phonons. Moreover, we use a modified Drude-Lorentz approach to calculate the transient dielectric function [2]. Our results show that visible light excitation leads to an overpopulation of the *sp*-band, primarily driven by photo-excitation, while XUV irradiation results in an underpopulation of the *sp*-band, dominated by subsequent impact ionization. Comparison of our simulation for the optical response with pump-probe experiments [1, 3] shows excellent agreement, suggesting a strongly improved understanding of the relevant physical processes and their timescales.

References:
[1] P.D. Ndione et al., Sci. Rep., 12, 4693 (2022)
[2] P.D. Ndione et al., ArXiv, 2307.11874 (2023)
[3] Z. Chen et al., Nat. Commun., 12, 1638 (2021)

O 36.12 Tue 18:00 Poster C

Towards time- and angle-resolved photoemission spectroscopy of plasmon enhanced van-der-Waals heterostructures — •Mattis Langendorf, Marco Merboldt, Jan Philipp Bange, Wiebke Bennecke, Paul Werner, David Schmitt, Jonas Pöhls, Anna Seiler, Thomas R. Weitz, Marcel Reutzel, and Stefan Mathias — Georg-August-Universität Göttingen, I. Physikalisches Institut, Germany

Light matter interaction in metals is mostly determined by a collective excitation of the charge carrier density, a quasiparticle called plasmon. For the case of surface plasmon polaritons (SPPs) that are localized at the surface-vacuum interface, the SPPs can be used to excite excitons in two-dimensional transition metal dichalcogenides (TMDs) that have been exfoliated onto the metal substrate. With this contribution, we work towards the characterization of the ultrafast exciton dynamics in TMDs that have been excited by SPPs. So far, femtosecond momentum microscopy had been used to study the optically excited exciton dynamics in TMDs [1]. Here, our approach is to image the propagating SPPs with photoemission electron microscopy and then to visualize the formation of excitons in the TMD with dark-field imaging techniques [2].

[1] Schmitt, Bange et al., Nature 608, 499-503 (2022).
[2] Schmitt et al. arXiv.2305.18908 (2023).

O 36.13 Tue 18:00 Poster C

Ultrafast Low Energy Electron Diffraction of Layered Materials — •Alp Akbiyik[1], Felix Kurtz[1], Monica Kolek Martinez de Azagra[2], Lukas Jehn[1], Hannes Böckmann[1], Dennis Epp[1], Thomas Weitz[2], and Claus Ropers[1,3] — [1]Max Planck Institute for Multidisciplinary Sciences, Göttingen, Germany — [2]1st Physical Institute, University of Göttingen, Germany — [3]4th Physical Institute, University of Göttingen, Germany

Enhanced by reduced dimensionality and the formation of heterostructures, van-der-Waals materials including few-layer graphene and transition metal dichalcogenides exhibit a large variety of tunable and correlated phenomena. Studying the non-equilibrium response of such materials, ultrafast surface-sensitive measurement techniques such as angle-resolved photoemission spectroscopy is instrumental in revealing couplings and correlations in the time domain. As a complementary method for surface-sensitive structural dynamics, we employ ultrafast low-energy electron diffraction (ULEED) [1]. In this contribution, we present recent progress in the development and application of this technique for the study of structural phase transformations and lattice thermalization. Moreover, we will discuss progress towards higher versatility of the approach in studying exfoliated samples prepared and transferred in an argon atmosphere.
[1] G. Storeck et al., Structural Dynamics 7, 034304 (2020).

O 36.14 Tue 18:00 Poster C

Photoemission electron microscopy of exciton dynamics in thinfilm TMD materials — •Lina Hansen, Kerstin Harland, Katrin Meier, Arvid Klösgen, and Jan Vogelsang — Institut für Physik, Carl von Ossietzky Universität Oldenburg, 26129 Oldenburg, Germany

Atomically thin layers of transition metal dichalcogenide (TMD) materials offer unique optical properties through their excitonic effects in the reduced dimensions. The resulting enhanced light-matter interactions are based on the arising excitons with long lifetimes being the main mechanism for light emission and recombination processes.

Using a photoemission electron microscope (PEEM) with optical excitation in the visible, near-infrared and extreme ultraviolet spectral region, thinfilm TMD materials are studied. In a first step, our investigations focus on exciton diffusion dynamics at structural interfaces. In particular, we utilize the unique tight focusing capabilities of our set-up to locally excite excitons in the laterally heterogenous TMD material and reveal the spatial behavior of the exciton decay.

O 36.15 Tue 18:00 Poster C

Coherent oscillations in the 4p$_{x,y}$ valence band of 1T-TiSe$_2$ — •Jan Böhnke, Mehul Jotshi, Stephan Schmutzler, Cornelius Gahl, and Martin Weinelt — Fachbereich Physik, Freie Universität Berlin, Germany

Upon cooling below T $\sim$ 200 K, 1T-TiSe$_2$ undergoes a structural phase transition which is characterized by a charge redistribution and a periodic lattice distortion. By time-resolved ARPES it is possible to investigate the dynamics of both, electronic band structure and phonon response. Upon excitation with a fs laser pulse with 1.55 eV photon energy we observe fluence-dependent coherent oscillations in the Se 4p$_{x,y}$ valence band of TiSe$_2$ which have been previously reported in a time-resolved photoemission experiment [1]. In contrast, no signature of coherent oscillations is found in the backfolded Ti 3d conduction band, suggesting that it is not a sensitive probe for the phase transition. Interestingly, we find image-potential states (n = 1, 2, 3) with lifetimes of up to 300 fs, indicating a low defect density of the surface after in-vacuum cleavage.
[1] S. Duan et al., Nature 595, 239-244 (2021)

O 36.16 Tue 18:00 Poster C

Few electron correlations from nanometric needle tips triggered by femtosecond laser pulses — •Felix Lopez Hoffmann, Jonas Heimerl, Stefan Meier, and Peter Hommelhoff — Department Physik, Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU), 91058 Erlangen

Metal needle tips are a standard source of free electrons for electron microscopes. When many electrons are emitted from such tips, space charge effects due to Coulomb repulsion appear as intuitively expected [1]. Here we demonstrate that already two electrons repel each other if confined tightly in space and time. In specific, we use femtosecond laser pulses to trigger multiphoton photoemission from metallic needle tips with a few nanometer in radius. The emitted electron pairs show an energy anti-correlation with a visibility of 56%, a mean energy splitting of 3.3 eV and a correlation decay time of 82 fs [2]. Similar results have been obtained in a TEM [3]. By energy filtering the emitted beam the number distribution of electrons per laser pulse can be tuned from Poissonian to sub/super-Poissonian statistics. Sub-Poissonian emission, in our case with a second order correlation $g(2)$ as low as 0.34, allows for shot-noise reduced imaging. Using such Coulomb-induced energy splitting, also heralding of electrons becomes possible. Furthermore, we show first results on spatial correlations stemming from transverse momentum exchange.
[1] Kuwahara et al., Appl. Phys. Lett. 109(1), 013108 (2016)
[2] Meier et al., Nat. Phys. 1-8 (2023)
[3] Haindl et al., Nat. Phys. 19, 1410-1417 (2023)

O 36.17 Tue 18:00 Poster C

An XUV time- and angle-resolved photoemission spectroscopy setup for complementary experiments with high temporal and high energy resolution — •Mohamed Amine Wahada, Tommaso Pincelli, Lawson Lloyd, Alessandro de Vita, Tulio de Castro, Martin Wolf, Ralph Ernstorfer, and Laurenz Rettig — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany

Time and angle resolved photoemission spectroscopy (trARPES) is a powerful technique to investigate the transient electronic band structure of materials on a femtosecond timescale. However, due to the Heisenberg's uncertainty principle, the achievable energy resolution is limited by the pulse bandwidth required for a given temporal resolution. Here, we present a new scheme to benefit separately from a high temporal resolution or a high energy resolution within one single experiment operating in the extreme ultraviolet (XUV) regime at 500 kHz. In the first case, 40 fs, 400 nm laser pulses generated by optical parametric chirped pulse amplification are used to generate the XUV. In the second case, 200 fs laser pulses at 515 nm are used. Both beams are guided to a harmonics generation chamber, where a single harmonic at ~21.7 eV is isolated and guided into the experimental trARPES chamber. The first setup allows high ~20 fs temporal resolution measurements while the second setup achieves an energy resolution of <40 meV. In conjunction with a wavelength tunable pump, this tool allows to perform pump probe ARPES experiment on the same sample granting access to complementary observables.

O 36.18 Tue 18:00 Poster C

A high repetition rate XUV source for time-resolved momentum space mapping of photoelectrons — •Yu Zhang, Adam S. Wyatt, James O. F. Thomson, Richard T. Chapman, Charlotte E. Sanders, and Emma Springate — Central Laser Facility, STFC Rutherford Appleton Laboratory, Research Complex at Harwell, Harwell, United Kingdom

With an ultrashort pulsed extreme ultraviolet (XUV) laser as the photon source, angle-resolved photoemission spectroscopy (ARPES) has extended the study of electronic structure beyond the 3D momentum space and into the temporal domain, with femtosecond resolution. Recently a new high repetition rate (100 kHz) XUV source has been developed in the Artemis facility of Central Laser Facility, UK, to provide an XUV beam source at energies from 20 to 45 eV, with a maximum flux of about 10^{10} photons/second. Together with a tunable laser source, time-resolved ARPES measurements are offered at Artemis with much higher data collection efficiency compared to the previous setup based on a 1 kHz system.

In this poster, the detailed beamline setup will be demonstrated. Besides a pulse-duration preserving XUV monochromator, a demagnification unit has been developed to focus the XUV spot size down to about 20 micrometers at the sample. This new capability allows users to perform experiments on small samples and distinct structural domains of inhomogeneous samples.

O 36.19 Tue 18:00 Poster C

Strong-Field Electron Emission in a Photoemession Electron Microscope (PEEM) — •Kerstin Harland, Lina Hansen, Katrin Meier, Arvid Klösgen, and Jan Vogelsang — Institut für Physik, Carl von Ossietzky Universität, 26129 Oldenburg, Germany

Time-resolved photoemission electron microscopy (PEEM) is well suited for investigating electric fields at nanostructures. Beyond imaging, exerting control on charge carriers with strong optical electric fields on the nanoscale is an ongoing challenge. Space charge constraints so far limit the applicable optical field strengths.

To overcome this problem, we investigate electron emission from a thin, monocrystalline gold flake with our photoemission electron microscope. We write an antenna in the gold flake so that surface plasmons can be excited. To reach the needed high intensity, we focus a 2000-nm wavelength laser beam with few-cycle pulses from our home-built laser system through the backside of the sample onto the gold flake antenna. Compared to conventional PEEM setups, this allows us to obtain a smaller focus size and address individual antennas without space charge problems. The antenna in the gold flake concentrates the optical energy from the laser in a small region, where electrons are emitted. We investigate the emission point and energy spectrum from these electrons with varying laser parameters.

O 36.20 Tue 18:00 Poster C

Impact of the CEP of the driving pulses on High-order Harmonic Generation — •Katrin Meier, Lina Hansen, Arvid Kloesgen, Kerstin Harland, and Jan Vogelsang — Institut für Physik, Carl von Ossietzky Universität Oldenburg, 26129 Oldenburg, Germany

The nonlinear process of High-order Harmonic Generation (HHG) is a method of generating ultrashort pulses in the extreme ultraviolet spectral range. They are of great interest to researchers because they can be used, for example, to probe ultrafast electron dynamics in various structures.

In this work, a home-built laser system is used that delivers ultrashort laser pulses centred around 2000 nm. The intense few-cycle near-infrared pulses are focused into a specially designed gas target where high harmonics of the driving laser field are generated. The carrier-envelope phase (CEP) of the driving pulses is measured with an f-to-2f interferometer. The focus of the work is on the influence of the CEP on the HHG spectrum and the corresponding temporal pulse structure.

O 36.21 Tue 18:00 Poster C

Few-cycle 2 μm light source for high-order harmonic generation at 200 kHz repetition rate — •Arvid Klösgen, Katrin Meier, Niels Cordes, Julia Altenburg, Lina Hansen, and Jan Vogelsang — Institut für Physik, Carl von Ossietzky Universität Oldenburg, 26129 Oldenburg, Germany

Electron microscopy with femtosecond time resolution requires laser sources with high repetition rates beyond 100 kHz to collect sufficient statistics without detrimental space charge effects. Investigations on the attosecond time scale additionally require high pulse energies to permit the generation of attosecond light pulses in a gas jet.

We report on a home-built high repetition rate laser system that includes an ytterbium-based 1030-nm, turn-key pump laser, white light generation, non-collinear optical parametric amplification in the visible range, difference frequency generation and additional optical parametric amplification. The system delivers broadband, few-cycle light pulses at a central wavelength of 2 μm with a pulse energy of 25 μJ and a passively stable carrier-envelope phase. Short- and long-term stability, beam quality, the temporal pulse structure and tunable parameters such as phase matching, time delay and pulse compression are subject of this study.

O 36.22 Tue 18:00 Poster C

Time-of-flight momentum microscopy for subcycle time-resolved photoemission — •Sarah Zajusch, Suguru Ito, Lasse Münster, Jens Güdde, Robert Wallauer, and Ulrich Höfer — Philipps-Universität Marburg

Time-of-flight momentum microscopy enables the simultaneous acquisition of energy and momentum distribution within the complete photoemission horizon. Thus, this technique paves the way to trace for example all exciton formations in TMD materials [1] and orbital images on ultrafast timescales [2]. In order to also investigate subcycle dynamics such as lightwave-driven currents in topological surface bands [3], we have to face additional challenges like photoelectron streaking and multi-hit detection.

Here, we present how photoelectron streaking affects time-of-flight experiments and how it can be compensated in the post processing. In addition, depending on pump frequency and field strength, it can be necessary to include a real-time streaking correction during the running experiment. We also show how multi-hit detection can be realized while maintaining good measurement performance.

[1] R. Wallauer *et al.*, Nano Lett. 21, 13, 5867-5873 (2021).
[2] R. Wallauer *et al.*, Science **371**, 1056-1059 (2021).
[3] S. Ito *et al.*, Nature **616**, 696 (2023).

O 36.23 Tue 18:00 Poster C

EUV pump-probe spectroscopic imaging — •Hannah Strauch[1], Fengling Zhang[2], Bent van Wingerden[1], Daniel Steil[1], Stefan Witte[2], Thorsten Hohage[3], Stefan Mathias[1], and G. S. Matthijs Jansen[1] — [1]University of Göttingen, I. Physical Institute, Göttingen, Germany — [2]Advanced Research Center for Nanolithography, Amsterdam, The Netherlands — [3]University of Göttingen, Institute of Numerical and Applied Mathematics, Göttingen, Germany

Extreme ultraviolet (EUV) spectroscopy accesses element-specific core levels, and thereby enables an element-resolved view of electronic, structural and magnetic dynamics. Likewise, EUV diffractive imaging allows nanoscale imaging of complex structures. The combination of both strengths in femtosecond spectromicroscopy is highly promising, but typically requires long measurement runs as well as a rigorous mathematical treatment of the data.

Here, we address these challenges by combining Fourier-transform spectroscopy and Fourier-transform holography (FTH) in a table-top interferometric high-harmonic generation experiment. From the far-field diffraction pattern of two phase-locked EUV pulses, we reconstruct spectromicroscopic images using an iteratively regularized Gauss-Newton algorithm. This approach implements prior knowledge of the spectral domain and thereby reduces the sampling requirements and measurement time by an order of magnitude. We will report on progress towards time-resolved spectroscopy of light-induced dynamics in exfoliated 2D material nanostructures based on spectroscopic FTH.

O 36.24 Tue 18:00 Poster C

Characterization of THz electric field transients by time-resolved photoelectron emission spectroscopy — •Hamed Abbasi[1], Ping Zhou[1], Florian Diekmann[2], Kai Rossnagel[2], Martin Mittendorff[1], and Uwe Bovensiepen[1] — [1]University of Duisburg - Essen, Germany. — [2]Kiel University, Germany

Energy- and momentum-dependent analysis of THz electric field induced charge carrier dynamics can be achieved on the surface of materials in THz-pump photoemission-probe experiments [1]. Here we report such an experimental setup in which the THz field is generated at 250 kHz repetition rate using a photoconductive antenna [2], with which we obtained a field with 1 THz center frequency and 0.5 ps duration. We have synchronized the THz pulse with an ultrashort

UV pulse for time-resolved angle-resolved photoemission spectroscopy. 45meV energy streaking of photoelectrons on the sample surface could be achieved at a cleaved surface of $1T$-TaS$_2$ sample, which is correspond to 6 kV/cm THz electric field. The separation of field induced dynamics in the material and acceleration of the emitted photoelectron in vacuum will be discussed.
We thank the DFG for funding through CRC 1242.

[1] Reimann et al. Nature **562**, 396 (2018).
[2] Dreyhaupt et al. Appl. Phys. Lett. **86**, 121114 (2005).

O 36.25 Tue 18:00 Poster C

Singlet fission and triplet dynamics in pentacene embedded in a surface-anchored metal-organic framework — •Martin Richter[1], Zhiyun Xu[2], Philipp Ludwig[3], Pavel Kolesnichenko[1], Uwe Bunz[3], Christof Wöll[2], and Petra Tegeder[1] — [1]Physikalisch-Chemisches Institut, Universität Heidelberg — [2]Institut für Funktionelle Grenzflächen, Karlsruher Institut für Technologie — [3]Organisch-Chemisches Institut, Universität Heidelberg

It has been observed, that the rate of SF and the lifetime of the generated triplets strongly depend on the molecular arrangement.[1]

Here, a cofacial orientation of pentacene molecules is achieved by embedding them in a surface anchored metal-organic framework (SURMOF). Transient absorption spectroscopy has been used to analyze the ultrafast dynamics as well as long lived states after photoexcitation. The observed difference absorption spectra indicate, that after the initial excitation a singlet excited state generates a correlated triplet pair within a few picoseconds that retains singlet character. Subsequent dynamics show the formation of a long-lived species (40us) with triplet character. This exceeds by far the observed lifetime of triplets generated in pentacene thin films (10ns) and may enhance triplet harvesting capabilities in photovoltaic devices.[2]

1. Lubert-Perquel, Nat Commun. 2018, 9, 4222
2. Poletayev, A.D., Adv. Mater. 2014, 26, 919-924

O 37: Poster: Electronic Structure of Surfaces and Spectroscopy of Surface States

Time: Tuesday 18:00–20:00 Location: Poster D

O 37.1 Tue 18:00 Poster D

iMott - imaging spin detector based on Mott scattering for SARPES — •Dmitry A. Usanov[1], J. Hugo Dil[1,2], and Vladimir N. Strocov[1] — [1]Spectroscopy of Quantum Materials group, Paul Scherrer Institut, Villigen CH-5232, Switzerland — [2]Institut de Physique, École Polytechnique Fédérale de Lausanne, Lausanne CH-1015, Switzerland

The electron spin is the crucial parameter of modern spintronics and therefore its determination in energy structures of solids is highly important. In order to boost spin-resolved ARPES' efficiency and accessibility, a prototype of a new imaging-type multichannel spin detector for electrons based on Mott scattering (nicknamed iMott) is being developed. The presentation will be focused on two main aspects: first, high stability and excellent image transfer will be demonstrated; second, the algorithms of signal acquisition and processing will be discussed. Two possible operation regimes, namely, the accumulating and single-electron counting modes, will be compared in conjunction with intensity and signal-to-noise ratio requirements, which determine reliable detection of spin asymmetry. The energy resolution of the multichannel detectors and the whole polarimeter will be shown and discussed. Further development directions will be outlined.

O 37.2 Tue 18:00 Poster D

A New Setup for the Measurement of the Angular Correlation of Annihilation Radiation — •Kilian Brenner, Francesco Guatieri, and Christoph Hugenschmidt — Lichtenbergstr. 1 85748 Garching

The measurement of the Angular Correlation of positron Annihilation Radiation (ACAR) provides an elegant solution to investigate Fermi-surfaces. The present ACAR spectrometer at TUM uses Anger cameras basically comprising large NaI(TL) crystals read out by an array of 61 photomultiplier tubes. In our new setup, we employ pixelated LYSO scintillation crystals in combination with state-of-the-art Multi-Photon-Pixel-Counters (MPPCs), drastically improving momentum resolution and count rate by capitalizing on the smaller detector-element size and faster readout. We present the new setup, its capabilities and preliminary measurements.

O 37.3 Tue 18:00 Poster D

Two-photon photoemission on Ag(111) with time-of-flight momentum microscope — •Yu-Chan Lin[1], Chieh-I Chen[1,2,3], Mukesh Singh[1], Prabesh Bista[1,2,4], and Cheng-Tien Chiang[1,2,5] — [1]Institute of Atomic and Molecular Sciences, Academia Sinica, Taipei, Taiwan — [2]Molecular Science and Technology Program, Taiwan International Graduate Program, Academia Sinica, Taipei, Taiwan — [3]Department of Chemistry, National Tsing Hua University — [4]Department of Physics, National Central University — [5]Department of Physics, National Taiwan University, Taipei, Taiwan

This study explores two-photon resonances in the electronic structure of Ag(111) through the application of two-photon photoemission. The light source is based on third harmonic generation via nonlinear optical crystals, transforming 1.2 eV, 300 fs laser pulses to 3.6 eV. By employing time-of-flight photoelectron momentum microscope, the electronic states of both occupied and unoccupied regions of the Ag sp bands can be observed via resonant two-photon absorption. By measuring the angular and time-of-flight distributions of the photoelectrons, the energy-momentum dispersion relation of these bands can be retrieved. These results are analyzed in detail and compared with earlier literature at different photon energies.

O 37.4 Tue 18:00 Poster D

Efficient Data Acquisition in Multi-Dimensional Photoemission Spectroscopy using denoising — •Muhammad Zain Sohail[1,2], Dmytro Kutnyakhov[2,3], and Kai Rossnagel[2,3] — [1]RWTH Aachen University, 52066 Aachen, Germany — [2]Deutsches Elektronen-Synchrotron DESY, 22607 Hamburg, Germany — [3]Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, 24098 Kiel, Germany

In the realm of photoemission spectroscopy, the exploration of large multi-dimensional phase spaces necessitates time-intensive data acquisition to ensure statistical robustness. Despite the unparalleled capabilities of free-electron lasers (FELs), in peak brightness and ultrashort pulsed X-rays, the limitations of low repetition rates prolong the data acquisition process. This impedes the agility of decision making that could otherwise enhance experimental results in the limited and valuable beamtime. By employing denoising strategies to mitigate noise while preserving intrinsic information, our proposed approach aims to streamline the data acquisition process, and effectively manage the escalating size and complexity of multi-dimensional photoemission data.

O 37.5 Tue 18:00 Poster D

Soft X-ray momentum-resolved photoelectron spectroscopy with a momentum microscope at Diamond Light Source — •Deepnarayan Biswas[1], Matthias Schmitt[1,2], Jieyi Liu[1], Olena Tkach[3], Ralph Claessen[2], Gerd Schönhense[3], and Tien-Lin Lee[1] — [1]Diamond Light Source Ltd., UK — [2]University of Würzburg, Germany — [3]University of Mainz, Germany

Angle-resolved photoelectron spectroscopy (ARPES) experiments are usually performed with excitation energies below ~100 eV to harness the higher photoionisation cross-sections and better energy and (inplane) momentum resolutions. However, the very short electron mean free paths at low energies severely limit the bulk sensitivity. It is therefore desirable to extend ARPES to the soft x-ray (SX) range, which opens up the opportunity for probing unconventional electronic structures at buried interfaces, studying 3D electronic systems etc.

We are currently commissioning a SX-ARPES end-station at Beamline I09 at DLS, equipped with a state-of-the-art momentum microscope (MM). MMs preserve the momentum view and resolution irrespective of the excitation energy, which is advantageous to ARPES performed at high energies. In addition, MMs enable selective studies of micron/sub-micron-sized sample areas using field apertures. Our MM is uniquely designed to use a single hemispherical analyser followed by a time-of-flight section fitted with a fast delay-line detector as a combined energy filter, resulting in highly efficient data collection. In this poster, we will present current progress and the potential applications of this new facility to quantum materials research.

O 37.6 Tue 18:00 Poster D

Influence of optical orbital angular momentum on the circular dichroism of Rashba system surface states — •Jannis Lessmeister[1], Tobias Eul[2], Benjamin Stadtmüller[1], and Martin Aeschlimann[1] — [1]Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau, Germany — [2]Institute of Experimental and Applied Physics, University of Kiel, Germany

Since its discovery in 1992 [1], twisted light has found its way into a wide variety of research areas and applications. Recently the interaction between its intrinsic orbital angular momentum (OAM) and condensed matter has become a steadily growing field of research [2].

In our research we employ momentum microscopy to gain new insights into the interaction of twisted light with metallic surfaces. Using a circularly polarized light beam carrying OAM we investigate the circular dichroism of the Au(111) Shockley surface state. We particularly focus on the influence of the photonic OAM on the dichroism signal of materials with different atomic spin-orbit strengths.

[1] Allen et al., Phys. Rev. A 45 (1992)
[2] Quinterio Rosen et al., Rev. Mod. Phys 94 (2022)

O 37.7 Tue 18:00 Poster D

Substrate doping and defect influence on P-rich InP(001):H surface properties — Rachele Sciotto, •Isaac Azahel Ruiz Alvarado, and Wolf Gero Schmidt — Lehrstuhl für Theoretische Materialphysik, Universität Paderborn, Paderborn, Germany

Density-functional theory calculations on P-rich InP(001):H surfaces are presented. Depending on temperature, pressure and substrate doping, hydrogen desorption will occur and influence the surface electronic properties. For p-doped samples, the charge transition levels of the P dangling bond defects resulting from H desorption will lead to Fermi level pinning in the lower half of the band gap. This explains recent experimental data. For n- doped substrates, H-deficient surfaces are the ground-state structure. This will lead to Fermi level pinning below the bulk conduction band minimum. Surface defects resulting from the adsorption of additional hydrogen can be expected as well but affect the surface electronic properties less than H desorption.

O 37.8 Tue 18:00 Poster D

Implementation of a fiber-based cathodoluminescence detector system for an ultrafast scanning electron microscope — •Filip Majstorovic[1], Paul H. Bittorf[1], and Nahid Talebi[1,2] — [1]Institute for Experimental and Applied Physics, Kiel University, Leibnizstraße 19, D-24118 Kiel, Germany — [2]Kiel Nano, Surface and Interface Science KiNSIS, Kiel University, Christian-Albrechts-Platz 4, D-24118 Kiel, Germany

When high-energy electron beams interact with materials, light can be emitted by the excited sample. This radiation is known as coherent or incoherent cathodoluminescence (CL), depending on the underlying interaction mechanism, and gained a major interest in the study of minerals, semiconductors and plasmonic resonances in nanoparticles, where both spectral and temporal statistics can be unraveled. Due to the high spatial resolution and large spectral excitation bandwidth of the electron beams inside an SEM, electrons locally probe photonic modes, resulting in the capability to image their spatial near-field distribution via CL spectroscopy. Here, we report on the construction of a fiber-based CL detector for an SEM. We present first measurements to prove the ability for raster scanning the samples CL emission using fibers. Additionally, this detector is implemented inside an ultrafast SEM (USEM) setup for realizing a time-resolved electron-light pump-probe measurement configuration, where the luminescence of a sample, coming from the excitation either with electron or laser pulses, will be spatially resolved. This will be used to improve the USEM setup and analyze the dynamics of electron-light-matter interaction.

O 37.9 Tue 18:00 Poster D

Electronic structure study of intercalated transition metal dichalcogenide — •Jyoti Kaswan[1], Laurent Nicolaï[1], Michal Procházka[1], Sarath Sasi[1], Sunil W Dsouza[1], Veronica Vavruňková[1], Rostislav Medlín[1], Zdeněk Sofer[2], Stefanie Gärtner[3], and Jan Minár[1] — [1]University of West Bohemia, Plzeň, Czech Rep. — [2]University of chemistry and technology Prague, Czech Rep. — [3]University of Regensburg, Germany

Intercalated transition metal dichalcogenide (TMDs) has recently attracted attention of condensed matter community owing to the exhibition of the exotic phenomena depending upon the intercalate transition metal [1]. We explore the electronic structure of V-intercalated TMDs V1/3NbS2 using spin and angle resolved photoemission spectroscopy (SARPES) in combination with one step model of photoemission as implemented in the SPR-KKR package [2], our calculations can combine temperature- and phonon energy-dependent effects via inclusion of both bulk and surface phonons. Raman spectroscopy has been employed to determine the spin-phonon interaction along with the inplane metal-metal distance and out of plane interlayer distance. For determining bulk as well as surface lattice structure of the V1/3NbS2 monocrystals, we have utilized multiple characterizations techniques Low electron energy diffraction (LEED), Transmission electron microscopy (TEM) and single crystal X-ray diffraction. [1] B. Edwards et al, Nature Materials 22, 459 (2023) [2] H. Ebert, D. Ködderitzsch and J. Minár, Rep. on Prog. in Phys. 74, 096501 (2011)

O 37.10 Tue 18:00 Poster D

Model for Linear and Circular Dichroism in Angle-Resolved Photoemission Spectroscopy — •Maximilian Ünzelmann[1], Noah Endres[1], Begmuhammet Geldiyev[1], Jakub Schusser[1], Hendrik Bentmann[2], and Friedrich Reinert[1] — [1]Experimentelle Physik 7 and Würzburg-DresdenCluster of Excellence ct.qmat, Universität Würzburg — [2]Center for Quantum Spintronics, Department of Physics, NTNU, Norway

Angle-resolved photoemission spectroscopy (ARPES) in combination with linear and circular dichroism (LD and CD) has evolved as a powerful tool to study spin-split electronic states in solids including, e.g., Rashba systems [1,2] or Weyl semimetals [3]. In particular, the combination of spin-orbit coupling (SOC) and inversion symmetry breaking (ISB) leads to the formation of spin- and orbital angular momentum (OAM) in the electronic wave functions. Here, we show – using a simple model for the photoemission matrix element – that both LD and CD are sensitive to the OAM in the initial state wave function. We apply our model to the eigenstates obtained from tight-binding band structure calculations. On this basis, the influence of (i) experimental parameters and (ii) properties of the initial state on the dichroic signal will be discussed. Finally, we show that those model calculations compare nicely with real experimental data on various types of material systems.

[1] H. Bentmann et al., Phys. Rev. Lett. 119, 106401 (2017)
[2] M. Ünzelmann et al., Phys. Rev. Lett. 124, 176401 (2020)
[3] M. Ünzelmann et al., Nat. Commun., 12, 3650 (2021)

O 37.11 Tue 18:00 Poster D

Surface preparation of topological insulator Bi_2Te_3 and ferromagnet $La_{0.7}Sr_{0.3}MnO_3$ for band structure measurements — •Øyvind Finnseth, Damian Brzozowski, Ingrid Hallsteinsen, and Hendrik Bentmann — Norwegian University of Science and Technology

The proximity coupling of topological insulators with magnetically ordered systems in heterostructure systems, wherein the magnetic order serves to break the time reversal symmetry of the topological insulator, have attracted much attention in later years. As components in such a system, we consider thin films of the topological insulator Bi_2Te_3 and ferromagnetic $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) grown by pulsed laser deposition. The signature of the topological state of Bi_2Te_3 may be found in the electronic band structure of the surface state. In order to understand the effect of proximity coupling on the surface state in a heterostructure, a full understanding of the properties of its constituents is required. However, the surfaces of both Bi_2Te_3 and LSMO degrade after exposure to air, rendering band structure measurements of surface states challenging. We therefore study surface preparation methods necessary for ARPES measurements on these materials. The studied methods include annealing with and without an oxygen atmosphere for LSMO, along with argon ion sputtering at different energies and duration for both samples. The suitability of each method is evaluated by XPS, yielding information on the presence of contaminants as well as the stoichiometry of the samples, while LEED serves to inspect the crystalline quality of the sample surface.

O 37.12 Tue 18:00 Poster D

Sum-Frequency Generation Spectro-Microscopy in the Reststrahlen Band of Wurtzite-type Aluminum Nitride — •Dorothée Mader, Richarda Niemann, Martin Wolf, Sebastian Maehrlein, and Alexander Paarmann — Fritz Haber Institute of the Max Planck Society, Berlin, Germany

Non-centrosymmetric materials including ferroics are commonly studied by second-harmonic imaging.[1] This method is highly sensitive to the material's symmetry, but is commonly employed non-resonantly. In contrast, infrared-visible (IR-VIS) sum-frequency generation (SFG) spectroscopy is employed resonantly to reveal vibrations, local structure and bonding of the material.[2] In SFG, the sample is excited at the IR-resonance and mixed with a non-resonant VIS beam leading to generation of sum-frequency light that is detected in the VIS. SFG benefits from the resonance but typically lacks spatial resolution. In this work, we combine the benefits of nonlinear optical imaging and SFG spectroscopy by employing SFG spectro-microscopy using the free electron laser of the Fritz Haber institute.[3] Specifically, we test the feasibility of our instrumentation for spectroscopy with m-plane wurtzite-type aluminum nitride as a model system. We find the experimental spectra to agree with our theoretical calculations. In the future, this method will provide spatially resolved spectroscopic information in inhomogeneous systems such as ferroics and their domains.

[1] M. Fiebig et al., APL 66, 2906, (1995).
[2] W. Faust et al., PRL 17, 1265, (1966).
[3] R. Niemann et al., APL 120, 131102, (2022).

O 38: Poster: Electronic Structure Theory

Time: Tuesday 18:00–20:00 Location: Poster D

O 38.1 Tue 18:00 Poster D

Pythonizing VASP: unleashing the power of collaboration in electronic structure theory — •Martin Schlipf[1], Sudarshan Vijay[1], and Georg Kresse[1,2] — [1]VASP Software GmbH, Berggasse 21, 1090 Vienna, Austria — [2]Faculty of Physics and Center for Computational Materials Science, University of Vienna, Kolingasse 14-16, 1090 Vienna, Austria

The Vienna Ab-initio Simulation Package (VASP) provides powerful capabilities for materials simulations. In this presentation, we describe our new interface between VASP and the Python ecosystem that enhances data handling and empowers users to tailor VASP's behavior through Python scripting.

Currently, the diverse tools for VASP require text-based parsing of the output of the calculations. To avoid future Python packages reimplementing this fragile parsing, we introduce a structured output in HDF5 format and implement a parsing tool—*py4vasp*. With py4vasp, we obtain the data in Python dictionaries and provide postprocessing functionality like plotting and visualization.

In addition, we recognize the desire of many users to modify VASP's behavior. We discuss a novel interface that executes Python code from within a VASP calculation. By providing this entry point, users can change, for example, the potential, the energies, or the forces and extend VASP functionalities according to their specific research needs.

O 38.2 Tue 18:00 Poster D

Accelerating plane-wave-based *ab initio* molecular dynamics by optimization of Fast-Fourier transforms for modern HPC architectures — •Christian Ritterhoff, Tobias Klöffel, Sagarmoy Mandal, and Bernd Meyer — Interdisciplinary Center for Molecular Materials and Computer Chemistry Center, FAU Erlangen-Nürnberg, Germany

The most important advantage of plane-wave basis sets is that wave functions can be transformed efficiently from reciprocal to real space and back by using the Fast-Fourier transform (FFT) algorithm. This allows to evaluate the kinetic and potential energy in reciprocal and real space, respectively, where both operators are diagonal. This reduces the computational cost for applying the Hamilton operator from N^2 to $N \log N$. However, the scalability of current FFT libraries is rather limited on today's HPC systems, which offer large numbers of compute nodes, each of them with many cores. Here we present our optimization of the FFT routines in the Car-Parrinello molecular dynamics code CPMD (www.cpmd.org). Data distribution and communication patterns have been revised to make optimal use of combined MPI and OpenMP parallelization. Scalability is further increased by combining FFTs into batches and by introducing overlapping computation and communication. The improved performance of the new FFT routines will be demonstrated by a series of benchmark simulations with our optimized version of the CPMD code [1].

[1] T. Klöffel, G. Mathias, B. Meyer, Comput. Phys. Commun. **260** (2021) 107745

O 38.3 Tue 18:00 Poster D

Simulation of time-resolved RIXS spectra of laser-excited La2CuO4 based on multiplet ligand-field theory using Wannier orbitals — •Nils Brouwer[1], Maurits W. Haverkort[2], Sina Shokri[2], and Alexander I. Lichtenstein[1,3] — [1]European X-Ray Free-Electron Laser Facility, Holzkoppel 4, 22869 Schenefeld, Germany — [2]Institute for Theoretical Physics, Heidelberg University, Philosophenweg 19, 69120 Heidelberg, Germany — [3]Institute of Theoretical Physics, University of Hamburg, Notkestraße 9-11, 22607 Hamburg, Germany

Recent advancements in X-ray free electron laser science allow for the measurement of resonant inelastic X-ray scattering (RIXS) spectra of optically excited materials on the femtosecond time scale. Naturally, these advancements pose increasing challenges for theory to follow up with reasonable accuracy. This is especially true for highly correlated materials like La2CuO4, which often display very interesting functional properties.

In this work, following the method presented in reference [1], we apply multiplet ligand-field theory on a single site copper cluster and a multi-site copper cluster. Both clusters are parametrized by all electron DFT calculations of La2CuO4 using Wannier orbitals. Then, we calculate the time evolution of the laser-excited state of the cluster using the Krylov basis. Finally, we present RIXS spectra based on the time evolution of the laser-excited state.

[1]Phys. Rev. B 85, 165113 (2012)

O 39: Poster: Metal Substrates

Time: Tuesday 18:00–20:00 Location: Poster D

O 39.1 Tue 18:00 Poster D

Chemisorption and diffusion of fluorocarbon compounds on Ag(110) — •Shilpa Panchami Raj, Matthew Timm, and Leonhard Grill — University of Graz

The chemisorption and diffusion of molecules adsorbing at metal surfaces are key processes in heterogeneous catalysis [1]. In this work, we present a scanning tunnelling microscopy (STM) study under ultrahigh vacuum (UHV) of the deposition of CF3I on the anisotropic Ag(110) surface. Experiments were done at cryogenic temperatures of 5 K, which allows imaging single molecules in a stable fashion and also to manipulate them with the tip of the microscope in a controlled way. Dissociative chemisorption of CF3I on Ag (110) was observed and studied in dependence on the sample temperature. Moreover, first attempts will be presented of how diffusion of individual molecules can be induced in a controlled way.

[1] G. Ertl, Angew. Chem. Int. Ed. 47, 3524 (2008)

O 39.2 Tue 18:00 Poster D

S on Ir(111): transition between two superstructures observed by LEED and Auger electron spectroscopy — •Alice Bremerich, Paulus Aleksa, Robin Ohmann, and Carsten Busse — Department Physik, Universität Siegen, Deutschland

Detailed knowledge of the adsorption of chalcogens on metal surfaces is of high interest to control the preparation of complex atomically thin materials like transition metal dichalcogenides (TMDC) or even their heterostructures. Furthermore, chalcogens are also useful intercalants, which can modify the interaction of 2D materials with their substrates. We investigated the adsorption of elemental sulfur on pristine Ir(111), which was proven to be an excellent substrate for TMDCs on a graphene buffer layer. The resulting superstructures are determined by low energy electron diffraction (LEED), and with Auger electron spectroscopy (AES) we could quantify the S coverage Θ_S for each deposition to calibrate the sulfur source in terms of sulfur pressure and deposition time.

For S coverages $\Theta_\mathrm{S} < 0.3$ ML we observe the well-defined superstructure $\left(\sqrt{3}\times\sqrt{3}\right)R30^\circ$. For 0.3 ML $< \Theta_\mathrm{S} <$ 0.5 ML we found a transition between this and the $c\,(4\times 2)$ superstructure by the formation of domain walls. Here the LEED spots of the $\left(\sqrt{3}\times\sqrt{3}\right)R30^\circ$ structure split into three spots where the distance between them increases with increasing coverage until it forms the $c\,(4\times 2)$ structure for $\Theta_\mathrm{S} > 0.5$ ML.

O 39.3 Tue 18:00 Poster D

Optimizing a Surfactant-Free Silver Substrate for Surface-Enhanced Raman Spectroscopy — •Georgiana Ion, Stefania D Iancu, and Nicolae Leopold — Faculty of Physics, Babes-Bolyai University, 400084, Cluj-Napoca, Romania

Surface-Enhanced Raman Spectroscopy (SERS) relies on the adsorption of analytes to metallic nanoparticles, such as silver, a process often impeded by surfactants. This study introduces a straightforward and efficient method to obtain a surfactant-free SERS-active substrate, facilitating prompt analyte adsorption.

Citrate-capped silver nanoparticles (AgNPs) were concentrated, dried on a glass slide, and exposed to an ozone-rich environment (UV Ozone Cleaner) to eliminate the citrate surfactant. However, exposure of AgNPs to O_3 resulted in metal oxidation, rendering the substrate SERS inactive. To remove AgO from the surface, we tested various pH and time combinations and found that the most effective method involved immersing the glass slide with the silver substrate in a pH 4-5 H_2O_2 solution for 70 seconds. After removing the AgO, SERS analysis revealed the disappearance of citrate from the AgNPs surface present before the cleaning process. Subsequently, immersion in a fumaric acid solution (5x10^{-5}M) was employed to assess SERS enhancement. The SERS signal suggested spontaneous adsorption to the metallic surface, affirming the creation of a SERS-active substrate.

In conclusion, a rapid method for producing a surfactant-free SERS substrate was developed and tested, based on surface O_3 cleaning followed by H_2O_2 reduction.

O 39.4 Tue 18:00 Poster D

Probing the chemical bond between Ni and a CO-terminated AFM tip — •Maximilian Krüger, Fabian Stilp, Marco Weiss, and Franz J. Giessibl — Institute of Experimental and Applied Physics, University of Regensburg, 93040 Regensburg, Germany

AFM with a CO-terminated tip usually images the total charge density of atoms in a sample. Huber *et al.* [1] discovered exceptions to the chemically inert behaviour of the CO tip. For single Fe adatoms on Cu(111) a subatomic toroidal configuration was found, which maxima are aligned with the threefold symmetry of the underlying Cu lattice. This is resulting from the hybridisation of electronic states of CO and the adatom.

DFT calculations [2] suggest a similar, by 60 degrees rotated signature for Ni adatoms on the Cu(111) surface. Motivated by this, the same AFM measurement processes are used to examine Ni.

It is revealed that the interaction between Ni and CO also transitions from physisorption to chemisorption when approaching. At small tip-sample distances Ni appears as an attractive center with a surrounding repulsive ring as it is the case for Fe, but with different ratios between attractive and repulsive forces. Ultimately the three maxima could not be detected.

[1]: F. Huber, et al. 'Chemical Bond Formation Showing a Transition from Physisorption to Chemisorption'. Science **366**, 6462 (2019): 235–238.

[2]: S. Polesya, S. Mankovsky, H. Ebert. unpublished calculations.

O 39.5 Tue 18:00 Poster D

On-top hydrogen adsorption on Pt(111): A complementary studies by LEED, TPD and sum-frequency generation spectroscopy — •Zhipeng Huang, Tobias Roos, Yujin Tong, and R. Kramer Campen — Faculty of Physics and Center of Nanointegration (CENIDE), University of Duisburg-Essen

Studying the hydrogen (H) adsorption on a metal surface is important for understanding the hydrogen evolution and oxidation reactions occurring at catalytic metal surfaces. However, there remain controversies on whether hydrogen can be adsorbed on-top sites stably even for the most-stable and close-packed (111) surface of platinum (Pt) under well-defined UHV conditions.

Here we present our studies on hydrogen adsorption on Pt(111) by the surface sensitive thermal desorption spectroscopy (TDS), low-energy electron diffraction (LEED) and vibrational-resonant sum-frequency generation spectroscopy (VSFG). We found that the stable adsorption sites of hydrogen on Pt(111) depend strongly on the substrate temperature when dosing hydrogen. The on-top adsorbed hydrogen is observed by both LEED and VSFG when dosing hydrogen with the substrate temperature at 210 K. We systematically studied the on-top adsorbed hydrogen on Pt(111) under different coverages using VSFG and revealed how the lateral interaction affects the on-top hydrogen-Pt vibrational frequency. Our results indicate that the prepared on-top adsorbed hydrogen might be accessed through a local equilibrium and different than that predicted by current generations of theory.

O 39.6 Tue 18:00 Poster D

***Ab initio*-based Chemisorption and Thermodynamic Stability of Atomic H and O on Pt-Ir Alloy Surfaces** — •Tobias Wittemann, Thorsten Klüner, and Halil Ibrahim Sözen — Institute of Chemistry, Carl-von-Ossietzky University of Oldenburg, 26129 Oldenburg, Germany

Fuel cell electric vehicles (FCEVs) powered by proton-exchange membrane fuel cells (PEMFCs) are a CO_2 emission-free alternative to conventional fossil fuel vehicles. By preventing the phenomenon of anodic carbon corrosion, bifunctional Pt-Ir anode catalysts could help enhancing the long-term durability of the PEMFCs applied in such vehicles.

In our study, we employed density functional theory (DFT) to investigate the chemisorption of atomic hydrogen (H) and oxygen (O) on bimetallic $Pt_3Ir(111)$ and PtIr(111) surfaces in comparison to the monometallic Pt(111) and Ir(111) surfaces. Our results show that the chemisorption on the alloys becomes more exothermic with increasing number of Ir atoms at the adsorption site. Using the *ab initio* thermodynamics approach, we calculated phase diagrams for the H and O adsorption to quantitatively describe the interplay of the pressure, temperature and surface coverage under typical operation conditions of PEMFCs.

Our findings provide a fundamental insight into the H and O adsorption on bimetallic Pt-Ir surfaces and can serve as a starting point for subsequent studies of the hydrogen oxidation reaction (HOR) and oxygen evolution reaction (OER), which are crucial for the rational design of novel anode catalyst materials.

O 39.7 Tue 18:00 Poster D

On-surface reactions of triazido-s-heptazine — •Ilias Gazizullin[1], Christophe Nacci[1], Friedrich Esch[2], and Leonhard Grill[1] — [1]Physical Chemistry Department, University of Graz, Austria — [2]Faculty of Chemistry, TU München, Germany

2,5,8-Triazido-s-heptazine is an aromatic molecule containing only carbon and nitrogen atoms. Heptazine core functionalized with reactive azido-groups makes it an interesting candidate as a precursor for on-surface synthesis of graphitic carbon nitride, an important 2D material [1]. The challenge in this approach is that there are several alternative reaction pathways, which are difficult to control, due to the high reactivity of the aromatic azides [2].

Here, we study these molecules by low temperature scanning tunneling microscopy (STM) under ultra-high vacuum conditions, which allows to study changes in single molecules induced by different impacts. Molecular transformations are induced either by voltage pulses from the STM tip or by illumination with UV light. In each case, we observe two types of processes. Short illumination or short voltage pulse induce dissociation of the azido-group into a molecular nitrogen and a nitrene molecule. On the other hand, longer illumination times lead to drastic increase of the apparent height, which will be discussed in view of other options, for instance the formation of another product or different adsorption configurations.

[1] Krinninger et al., Chem. Mater., 35, 17, 6762 (2023)

[2] Hellerstedt et al., Angewandte Chemie, 58, 2266 (2019)

O 39.8 Tue 18:00 Poster D

Design of a simple and highly reliable evaporator for rare-earth metals — •Tobias Edelmann, Patrick Härtl, Markus Leisegang, and Matthias Bode — Physikalisches Institut, Experimentelle Physik II, Julius-Maximilians-Universität Würzburg, Am Hubland, 97074 Würzburg, Germany

Rare-earth metals exhibit versatile structural, electronic, optical, and magnetic properties. In our technology-based society, there is a significant demand for these metals in various applications, both in applied and basic research. Since the high chemical reactivity of rare-earth metals essentially impedes the preparation of clean surfaces from bulk crystals, their properties are usually studied as thin films which are epitaxially grown on highly pure mono-crystalline substrates. To guarantee the chemical purity of rare earth metal films, we have developed a simple and highly reliable evaporator design. Mounted on a DN35CF-sized electrical feedthrough, this home-built evaporator does not require any external cooling but still remains stable over extended periods of times, facilitating even the deposition of thick layers [1]. The design details will be presented in-depth in this poster, showcasing its application by the characterization of one specific evaporant by means of spin-polarized scanning tunneling microscopy.

[1] P. Härtl *et al.*, Phys. Rev. B **105**, 174431 (2022).

O 39.9 Tue 18:00 Poster D

P2O and P4O on Ag(111): Homogeneous layers, heterostructures, and mixed layers — Ravi Priya, Keda Jin, and •Peter Jakob — Department of Physics, Philipps-Universität Marburg, Germany

A refined structural analysis of 6,13-pentacenequinone (P2O) and 5,7,12,14-pentacenetetrone (P4O) phases on Ag(111) has been conducted using SPA-LEED. In addition, FT-IR absorption spectroscopy and TDS have been employed to characterize the various phases and associated phase transformations. For both species, lateral ordering is governed by orientation-dependent intermolecular interactions including steric hindrance and in particular hydrogen bonding. P2O as well as P4O display characteristic reflexes in LEED, signalling long-range ordering. These patterns are largely retained up to bilayer coverages. When heterolayers are grown, the reflexes of the ordered contact layers are preserved. Layer exchange is found to occur for the 'P4O on P2O' stacking at $T > 200K$; no such processes are found for the reversed stacking sequence up to the desorption temperatures of second layer P2O. Different ordered P2O-P4O mixed phases have been identified depending on their relative concentrations in the monolayer and their structure determined. In our study different initial layers have been used to create the various P2O-P4O mixed phases: (i) well-defined 'P4O on P2O' heterolayer stacks plus thermal annealing, (ii) co-deposition of P2O and P4O at 83K, followed by annealing, and (iii) incremental increase of P4O concentration within P2O/Ag(111) monolayer by stepwise addition of P4O followed by annealing.

O 39.10 Tue 18:00 Poster D

Structure of tellurium phases on Fe/Ir(111) — •Alexander Wegerich, Tilman Kisslinger, Lutz Hammer, and M. Alexander Schneider — Universität Erlangen-Nürnberg, 91058 Erlangen, Germany

Within the group of transition metal chalcogenides (TMCs), iron tellurides hold great promise in terms of their magnetic and electronic properties [1-2]. A way to prepare such systems in a scalable way accessible for surface science methods is to induce reactions between Te and Fe at the surface of a chosen substrate in ultra-high vacuum. Ir(111) was chosen, since no (surface) telluride formation was observed on Ir(111) enhancing the chance of iron telluride growth upon deposition of Fe and Te.

By quantitative LEED-IV analyses, STM and DFT, we determine the occurring surface structures as function of Te and Fe content and preparation temperatures. No iron telluride phases but the formation of Te adphases on pseudomorphic Fe layers on Ir(111) were observed. At $\Theta_{\mathrm{Te}} = 0.25\,\mathrm{ML}$ a stable Fe/Ir(111)-(2×2)-Te occurs that has no counterpart on the bare Ir(111) surface, whereas increasing Θ_{Te} to $0.33\,\mathrm{ML}$ and beyond produces a $\left(\sqrt{3} \times \sqrt{3}\right)\mathrm{R}30^\circ$ adsorbate phase that can be further compressed to accommodate more Te, exactly as for Te on Ir(111).

[1] Z. Zhang et al., Phys. Rev. Mater. **4**, 125003 (2020)

[2] D. Fobes et al., Phys. Rev. Lett. **112**, 187202 (2014)

O 39.11 Tue 18:00 Poster D

Re-calibrating the experimentally derived structure of the metastable surface oxide on copper via machine learning-accelerated in silico global optimization — •Hyun Jun Kim[1], Giyeok Lee[1,2], Seung-Hyun Victor Oh[1], Catherine Stampfl[2,3], and Aloysius Soon[1,2] — [1]Department of Materials Science and Engineering, Yonsei University, Seoul, Republic of Korea — [2]School of Physics, The University of Sydney, NSW, Australia — [3]The University of Sydney Nano Institute, The University of Sydney, NSW, Australia

The oxidation of copper and its surface oxides are gaining increasing attention due to the enhanced CO_2 reduction reaction (CO2RR) activity exhibited by partially oxidized copper among the copper-based catalysts. The "8" surface oxide on Cu(111) is seen as a promising structure for further study due to its resemblance to the highly active $Cu_2O(110)$ in the C-C coupling of CO2RR, setting it apart from other O/Cu(111) surface oxides resembling $Cu_2O(111)$. However, recent X-ray photoelectron spectroscopy experiments analysis challenges the currently accepted atomic structure of the "8", prompting a need for reevaluation. In this study, we highlight the limitations of conventional methods when addressing such challenges, leading us to adopt global optimization search techniques. After a rigorous process to ensure robustness of our computational workflow, the unbiased global minimum of the "8" structure is identified. Interestingly, this new configuration differs significantly from other surface oxides and also from previous "8" models while retaining similarities with the $Cu_2O(110)$ surface.

O 40: Poster: Topological Materials

Time: Tuesday 18:00–20:00 Location: Poster D

O 40.1 Tue 18:00 Poster D

Topological phases of quasi-one-dimensional systems — •Nemanja Madić, Tatjana Vuković, and Saša Dmitrović — University of Belgrade Faculty of Physics, Belgrade, Serbia

Obstructed atomic limits (OALs) of crystalline insulators are topological phases characterized by the Wannier orbitals centers unmovable to ion-occupied positions. Complete information on the orbital's movability from the Wyckoff position (WP) is encoded within the symmetry group of the system. The symmetry of a quasi-one-dimensional (Q1D) system is described by one of the line groups gathered within thirteen infinite families. Their electronic structures are always Wannierizable; consequently, all topological insulating phases of such systems correspond to OALs. As inequivalent topological insulating phases are separated by closing the band gap or breaking the crystalline symmetry, the complete set of such phases can be deduced by analyzing the Q1D systems symmetry group. Here, we explore the OALs of spinless and spinful systems whose symmetry groups belong to the 11th and 13th families. To this end, firstly, the real space invariants, needed to test the movability of Wannier centers allocated at the maximal WP of the line groups, are calculated. Next, these results and recently obtained sets of elementary band representations of ordinary and double line groups are applied to obtain the set of distinct Wannier homotopy classes. Finally, district ion positions of the several systems, with equal symmetry groups, are analyzed to identify all possible phases corresponding to the OAL. Results are illustrated on models created within the PythTB package.

O 40.2 Tue 18:00 Poster D

Topological edge states in quasi-one-dimensional systems — •Bogdan Stanojević and Saša Dmitrović — University of Belgrade Faculty of Physics, Belgrade, Serbia

Robust topological edge states (TESs) of crystalline materials are of great interest, both from the fundamental and application-driven point of view. These states, protected by the lattice symmetry, have been theoretically predicted in 1D inversion symmetric chain [1], as well as in 2D crystals with symmetries p2, p3, and pmm [2]. Here, the analysis is generalized to all quasi-1D (Q1D) systems. These materials are translationally or helically periodic along only one axis, with their symmetry group belonging to one of the thirteen infinite families of line groups (LGs) [3]. Here we consider only time reversal symmetrical Q1D systems. As the full symmetry group of such a system is described by one of the grey LGs, recently derived complete sets of elementary band co-representations (co-EBRs) of grey LG [4], enable the search for TESs: for the system of chosen symmetry, the analytical continuation of distinct elementary band structures at the edges of the irreducible domain singles out possible robust TESs. The results are illustrated on several tight-binding models developed within PythTB package [5].

[1] J. Zak, Phys. Rev. B 32, 2218 (1985). [2] A. Silva, J. van Wezel, SciPost Phys. 10, 137 (2021). [3] M. Damnjanović and I. Milošević, Line Groups in Physics: Theory and Applications to Nanotubes and Polymers (Lecture Notes in Physics vol 801), (Berlin: Springer) (2010). [4] S. Dmitrović S et al., J. Phys. A: Math. Theor. 55 385201 (2022). [5] https://www.physics.rutgers.edu/pythtb/

O 40.3 Tue 18:00 Poster D

On the Edge State of Finite 2D Bi_2Se_3 Crystals — •Auke Vlasblom, Victor Wesselingh, Jesper Moes, Jara Vliem, Daniel Vanmaekelbergh, and Ingmar Swart — Utrecht University, Utrecht, The Netherlands

Topological insulators in two or three dimensions exhibit an insulating bulk, but possess gapless conducting edge or surface states that are protected by time-reversal symmetry. One property of the surface/edge state is spin momentum locking, which results in two types of surface currents, each with opposite spin (up or down) and opposite momentum (direction). In the 2D regime, top and bottom states hybridize and the interior becomes insulating. The two remaining types of edge currents are fully protected from backscattering by non-magnetic impurities.

Here, we study thin (3–6 quintuple layers) finite sized Bi_2Se_3 crystals, prepared via wet-chemical synthesis, using scanning tunneling microscopy and spectroscopy. Measurements are performed on Bi_2Se_3 nanoplatelets with varying thickness to study the transition between 3D and 2D topological insulators. Additionally, the robustness of the edge states is investigated with respect to structural defects and deposition of magnetic impurities.

O 40.4 Tue 18:00 Poster D

Stability of Majorana Fermions in Coulomb-Disordered Topological Insulator Nanowires — •Leonard Kaufhold — Institute for theoretical physics, University of Cologne, Germany

A topological insulator (TI) nanowire, proximity-coupled to an s-wave superconductor (SC) and subject to a longitudinal magnetic field can, in principle, be brought into an effectively spinless superconducting phase under a relatively large region of realistically accessible parameter space and therefore host Majorana Bound States (MBS) at its ends. Due to their non-Abelian statistics, devices that can manifest MBS are a promising candidate for fault-tolerant quantum computing. A TI however, is typically realized as a heavily doped semiconductor with a donor density in the order of $*10^{19}$cm-3. The random potential arising from these impurities by themselves is much larger than the electronic band gap of the surface states due to dielectric confinement, easily destroying their one-dimensional nature necessary exhibit a topologically superconducting phase along the entire wire. Although this potential is screened by the metallic surface states as well as the superconductor, the ultimate fate of the MBS, i.e. whether they can "survive" the screened potential, is not at all obvious. On this poster, we will provide a general review of the TI-SC hybrid nanowire and numerically investigate its phase space based on Andreev spectra, edge state localization and topological invariants.

O 40.5 Tue 18:00 Poster D

A new Bi_4Br_4 surface structure — •Jonathan K. Hofmann[1,2], Hoyeon Jeon[3], Saban M. Hus[3], An-Ping Li[3], Mingqian Zheng[4], Jin-Jian Zhou[5], Zhiwei Wang[4], Yugui Yao[4], F. Stefan Tautz[1,2], Bert Voigtländer[1,2], and Felix Lüpke[1] — [1]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, Germany — [2]Lehrstuhl für Experimentalphysik IV A, RWTH Aachen University, Germany — [3]Center for Nanophase Materials Science, Oak Ridge National Laboratory, Tennesse, USA — [4]Beijing Key Lab of Nanophotonics and Ultrafine Optoelectronic Systems, Beijing Institute of Technology, China — [5]Institute of Physics, Chinese Academy of Sciences and Beijing National Laboratory for Condensed Matter Physics, China

Bi_4Br_4 is a higher-order topological insulator with a quasi-one dimensional crystal structure [1]. We present a new Bi_4Br_4(001) surface structure analysed with Scanning Tunneling Microscopy/Spectroscopy (STS/STS) at 5 K. STM topography shows an oblique surface structure with characteristic parallel chains shifted by $\sim \frac{b}{3}$ with respect to each other, as opposed to the shift of $\frac{b}{2}$ on α-Bi_4Br_4(001) [1]. We also observe AB-layer stacking, similar to α-Bi_4Br_4 [1]. STS reveals the presence of metallic edge states at surface steps, indicating that this new Bi_4Br_4 structure is also a topological insulator. We propose a crystallographic model for this surface and support our results by density-functional theory calculations.

[1] Shumiya, N.; Hossain, M. S.; Yin, J.-X.; Wang, Z.; Litskevich, M.; Yoon, C.; *et al.*, Nat. Mater. 2022, **21**, 1111*1115.

O 40.6 Tue 18:00 Poster D

The Coarse Geometric Origin of Topological Phases — •Christoph S. Setescak[1] and Matthias Ludewig[2] — [1]Institute of Experimental and Applied Physics, University of Regensburg, Universitätstraße 31, 93080 Regensburg, Germany — [2]Faculty of Mathematics, University of Regensburg, Universitätstraße 31, 93080 Regensburg, Germany

Topological phases of matter rely on the concept that the ensemble of occupied bulk energy bands of a translationally invariant Hamiltonian can be classified by topological invariants by making use of internal electronic symmetries. Non-trivial invariants give rise to exceptional electronic states at the boundary. This approach falls short when dealing with disorder induced by prevalent crystal defects. We propose that one should work in a coarse geometric framework, where the invariant can be defined in the presence of disorder. This construction is physically motivated, provides a natural setting for the bulk-boundary correspondence and furthermore provides a numerical efficient way to calculate the invariants. We apply this approach to a low energy tight-binding model of a three dimensional topological insulator with time reversal symmetry. We discuss the phase diagram in the disorder-free case and analyze the evolution of the topological phase upon the introduction of disorder.

O 41: Overview Talk Javier Aizpurua

Time: Wednesday 9:30–10:15 Location: HE 101

Invited Talk O 41.1 Wed 9:30 HE 101

Extreme interaction of photons and electrons in metallic nanocavities and STM tunnelling gaps — •Javier Aizpurua — Donostia International Physics Center, San Sebastian, Spain — Ikerbasque, Basque Country, Spain — University of the Basque Country (UPV/EHU), Spain

A meta-insulator-metal cavity is an ideal configuration to explore light-matter interaction in extreme spatiotemporal regimes. The interplay between optical and transport properties in metallic nanogaps provides new capabilities to produce photocurrents induced by ultrashort photon pulses, for instance, or to obtain light emission from single molecules driven by tunneling DC currents. We adopt a combination of theoretical approaches to address aspects of these dynamical processes. A description of the electron dynamics in nanogaps based on time-dependent density functional theory serves to unveil the origin of photoinduced currents and asses their behavior with photon intensity and carrier envelope phase. Furthermore, a quantum description of excitons in single organic molecules, interacting with a picocavity formed in a tunneling gap, provides an adequate framework to understand intramolecular resolution of molecular light emission in STM. The rich interaction between photons and electrons at the surface of metallic nanocavities brings optoelectronics to the realm of the atomic scale.

O 42: Focus Session: Frontiers of Electronic-Structure Theory – Advances in Time-Dependent and Nonequilibrium Ab Initio Methods III

Time: Wednesday 10:30–12:30 Location: HE 101

O 42.1 Wed 10:30 HE 101

Pauli blocking and core-hole screening in X-ray transient absorption spectroscopy at the Zn K-edge in ZnO — •Lu Qiao[1], Thomas C. Rossi[2], Keith Gilmore[1], Renske M. Van der Veen[2], and Claudia Draxl[1] — [1]Physics Department and IRIS Adlershof, Humboldt-Universität zu Berlin, 12489 Berlin, Germany — [2]Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, 14109 Berlin, Germany

X-ray transient absorption (XTA) spectroscopy can provide valuable insight into the atomic structure, the electronic properties, and the dynamics of materials. Zinc oxide (ZnO) is a prototypical wide band gap band insulator with many applications in optoelectronics and photovoltaics. Combining state-of-the-art first principle calculations and picosecond XTA spectroscopy, we reveal XTA spectra at the Zn K-edge of ZnO thin films are composed of overlapping contributions from lattice heating and core-hole screening. In particular, we analyze the computed results obtained from the Bethe-Salpeter equation and time-dependent density functional theory to disentangle the contributions from Pauli blocking and core-hole screening. Our combined experimental and theoretical approach reveals that core-hole screening enhances with increasing excitation densities; its contribution to the XTA spectra dominates over Pauli blocking.

O 42.2 Wed 10:45 HE 101

Photoemission tomography of excitons in periodic systems through Wannier interpolation — •Christian Simon Kern and Peter Puschnig — Institute of Physics, NAWI Graz, University of Graz, Austria

Excitons, bound optical excitations below the band gap of a material, are theoretically described as correlated electron-hole states. Their wave functions can be obtained by many-body perturbation theory in the framework of the Bethe-Salpeter equation or within time-dependent density functional theory. Experimentally, time- and angular-resolved photoemission spectroscopy (t-ARPES) is arguably the most direct method to measure the excitons' momentum-space signatures, which is also boosted by the recent developments in ultra-fast laser physics.

For molecules in the gas-phase, a formal connection of the theoretical exciton wave function with measured t-ARPES momentum maps in the spirit of photoemission orbital tomography was recently established [1] and has lead to interesting consequences for t-ARPES: the photoemission signature of an exciton is measured at all kinetic energies that are in accordance with the energy conservation from its hole contributions, while the respective momentum maps result from a coherent sum over all electronic contributions. Here, we extend this formalism to periodic systems and momentum-dark excitons, which allows for the simulation of photoemission from excitons in crystals, van der Waals hetero-structures or metal-organic interfaces.

[1] C. S. Kern et. al., Phys. Rev. B 108, 085132 (2023).

O 42.3 Wed 11:00 HE 101

Unveiling electronic correlation and many-body effects in the CrMnFeCoNi high-entropy alloy — •David Redka[1,2], Marco Caputo[5], Trpimir Ivsic[3], Heinz Huber[2], Hubert Ebert[4], J. Hugo Dil[3,5], László Forró[3,6], and Ján Minár[1] — [1]University of West Bohemia, Czech Republic — [2]Hochschule München, Germany — [3]Institute of Physics, EPFL, Switzerland — [4]Universität München, Germany — [5]Paul Scherrer Institut, Switzerland — [6]University of Notre Dame, USA

The impact of electronic correlation effects in the CrMnFeCoNi high-entropy alloy on electronic transport, spectroscopies, and optics is investigated. Here we combine experiments like resonant photoemission, and optical response with DFT calculations utilizing the KKR Green's function method including many-body effects via dynamical mean-field theory (DMFT). Our results show excellent agreement between theory and experiment, particularly for onsite Coulomb interactions, mirroring those in the containing pure elements. Through linear response calculations, we demonstrate the influence of the imaginary part of the self-energy of localized d-band electrons. In terms of electrical resistivity, we confirm the known dominance of chemical and magnetic disorder at low temperatures, whereas at higher temperatures, the DMFT contribution becomes significant. In general, we find that many-body correlation effects especially affect states at higher binding energies, next to the strong band smearing due to chemical disorder, revealing the impact on different material properties and response.

O 42.4 Wed 11:15 HE 101

Mott Metal-Insulator Transition from Steady-State Density Functional Theory — •Stefan Kurth[1,2,3], David Jacob[1,2], and Gianluca Stefanucci[4,5] — [1]Univ. of the Basque Country UPV/EHU, San Sebastian, Spain — [2]IKERBASQUE, Basque Foundation for Science, Bilbao, Spain — [3]Donostia International Physics Center DIPC, San Sebastian, Spain — [4]Univ. of Rome Tor Vergata, Rome, Italy — [5]INFN, Section Rome Tor Vergata, Rome, Italy

We present a computationally efficient method to obtain the spectral function of bulk systems in the framework of steady-state density functional theory (i-DFT) using an idealized scanning tunneling microscope (STM) setup. We calculate the current through the STM tip and then extract the spectral function from the finite-bias differential conductance. The fictitious noninteracting system of i-DFT features an exchange-correlation (xc) contribution to the bias which guarantees the same current as in the true interacting system. Exact properties of the xc bias are established using Fermi-liquid theory and subsequently implemented to construct approximations for the Hubbard model. We show for two different lattice structures that the Mott metal-insulator transition is captured by i-DFT.

O 42.5 Wed 11:30 HE 101

Quasiparticle Self-Consistent *GW* Study of Simple Metals — •Christoph Friedrich, Stefan Blügel, and Dmitrii Nabok — Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany

The *GW* method is a standard method to calculate the electronic band structure from first principles. It has been applied to a large variety of semiconductors and insulators but less often to metallic systems, in particular, with respect to a self-consistent employment of the method. In this work, we take a look at all-electron quasiparticle self-consistent *GW* (QS*GW*) calculations for simple metals (alkali and alkaline earth metals) based on the full-potential linearized augmented-plane-wave approach and compare the results to single-shot (i.e., non-selfconsistent) G_0W_0 calculations, density-functional theory (DFT) calculations in the local-density approximation, and experimental measurements. We show that, while DFT overestimates the bandwidth of most of the materials, the *GW* quasiparticle renormalization corrects the bandwidths in the right direction, but a full self-consistent calculation is needed to consistently achieve good agreement with photoemission data. The results mainly confirm the common belief that simple metals can be regarded as nearly free electron gases with weak electronic correlation. The finding is particularly important in the light of a recent debate, in which this seemingly established view has been contested. D.N. and S.B. gratefully acknowledge financial support by the MaX Center of Excellence funded by the EU through the H2020-INFRAEDI-2018 (project: GA 824143).

O 42.6 Wed 11:45 HE 101

FAIR Spectroscopy Data in NOMAD: from Theory towards Experiments — •José M. Pizarro[1], Nathan Daelman[1], Joseph F. Rudzinski[1], Luca M. Ghiringhelli[2], and Silvana Botti[3] — [1]Institut für Physik und IRIS-Adlershof, Humboldt-Universität zu Berlin, Berlin — [2]Department of Materials Science and Engineering, Friedrich-Alexander-Universität, Erlangen-Nürnberg — [3]RC-FEMS and Faculty of Physics, Ruhr University Bochum, Bochum

The emergence of big data in science underscores the need for FAIR (Findable, Accessible, Interoperable, Reusable) [1] data management. NOMAD [nomad-lab.eu][2, 3] is an open-source data infrastructure that meets this demand in materials science, enabling cross-disciplinary data sharing and annotation for both computational and experimental users.

In this talk, I will present our recent work in extending NOMAD to support a range of many-body and excited state calculations, including *GW*, BSE, and DMFT, among others. I will demonstrate how NOMAD captures these workflows in an automated but flexible fashion, enabling findability and clear, visual overviews. Finally, I will present an outlook on NOMAD′s potential for large-scale interoperability and harmonization between computational and experimental data in the field of spectroscopy.

[1] Wilkinson, M. D. *et al.*, Sci. Data **3**, 160018 (2016).
[2] Scheffler, M. *et al.*, Nature **604**, 635-642 (2022).
[3] Scheidgen, M. *et al.*, JOSS **8**, 5388 (2023).

O 42.7 Wed 12:00 HE 101

Electronic structure of oxides and perovskites: a benchmark database — •Kshitij Sinha[1,2] and Claudia Draxl[2] — [1]Indian Institute of Science Education and Research, Tirupati, India — [2]Humboldt-Universität zu Berlin, Institut für Physik and IRIS Adlershof, 12489 Berlin, Germany

Both perovskites and oxide materials have gained significant attention in recent years due to their unique properties and, thus, a wide range of possible applications. In this work, we investigate from first principles metal oxide and halide perovskites, like $BaSnO_3$, $CsPbI_3$, etc., and large band-gap oxide materials, like Ga_2O_3, ZrO_2, etc. We focus on their electronic properties, computing their electronic band structure and band gap, the density of states, effective masses, and more by applying density-functional theory. We employ a range of density functionals, i.e. the local spin density approximation (LDA), the generalized gradient approximation (PBEsol), as well as the hybrid functional PBE0 and HSE06. All calculations are carried out with the full potential all-electron package **`exciting`** [1], that implements the linearized augmented planewave plus local orbital (LAPW+lo) basis to achieve highly precise results. **`exciting`** exhibits thus proven benchmark quality, reaching μHa precision [2]. Our calculations present benchmark calculations that can serve as valuable reference data for the community.

[1] A. Gulans *et al.* J. Phys.: Condens. Matter **26** (2014) 363202
[2] A. Gulans *et al.* Phys. Rev. B, **97**, 161105

O 42.8 Wed 12:15 HE 101

Beyond the GW approximation using effective interactions — •Abdallah El Sahili — École Polytechnique

Electron addition and removal spectra as well as the total ground state energy can be expressed in terms of the one-body Green*s function. Often, the Dyson equation with an approximate self-energy is solved to access this quantity. Today, the probably most widely used approximation is Hedin’s GW self-energy [1], which has become the state-of-the-art for the band structure calculations of solids. However, GW still suffers from the existence of a variety of different flavors that lead to a sometimes significant spread of results, it has problems to describe satellite structures in the spectral function, and it is limited to a range of materials where correlations are weak to moderate. Especially total energy calculations present a number of open questions. In the present work we concentrate on the screened interaction W. On one side, we discuss the different perturbation expansions for the self-energy that can be explored according to the choice of W. On the other side, we generalize the concept of the screened interaction to a generalized effective interaction, for which we derive in principle exact equations as well as promising approximations. We show that this allows one to go beyond the GW approximation in an efficient way, avoiding the computational complexity of full vertex corrections. Illustrations for the general findings are given using the half-filled symmetric Hubbard dimer.

[1] Lars Hedin, Phys. Rev. 139, A796 (1965)

O 43: Focus Session: Spins on Surfaces studied by Atomic Scale Spectroscopies IV

Time: Wednesday 10:30–13:00 Location: MA 004

O 43.1 Wed 10:30 MA 004

A scanning quantum sensor for atomic-scale electric and magnetic fields — •Taner Esat[1,2], Dmitriy Borodin[3,4], Jeongmin Oh[3,4], Andreas Heinrich[3,4], Stefan Tautz[1,2,5], Yujeong Bae[3,4], and Ruslan Temirov[1,2,6] — [1]Peter Grünberg Institute (PGI-3), Forschungszentrum Jülich; Jülich, Germany — [2]Jülich Aachen Research Alliance (JARA), Fundamentals of Future Information Technology; Jülich, Germany — [3]Center for Quantum Nanoscience (QNS), Institute for Basic Science (IBS); Seoul 03760, South Korea — [4]Department of Physics, Ewha Womans University; Seoul 03760, South Korea — [5]Experimentalphysik IV A, RWTH Aachen University; Aachen, Germany — [6]Faculty of Mathematics and Natural Sciences, Institute of Physics II, University of Cologne; Cologne, Germany

In this work, we report the fabrication of a quantum sensor delivering atomic resolution and single-spin sensitivity, assembled atom-by-atom at the apex of an ultra-high vacuum low temperature scanning tunnelling microscope (STM) tip. Our quantum sensor consists of a magnetic tip with an aromatic molecule attached by atomic-scale manipulation, forming a quantum mechanical two-level system in an external magnetic field. It is addressable by STM-based electron spin resonance and allows readout by tunneling magnetoresistance. We demonstrate the functionality of the quantum sensor by measuring the magnetic and electric dipole fields emanating from a single Fe atom and an Ag2 dimer with sub-microvolt energy and simultaneous atomic-scale spatial resolution.

O 43.2 Wed 10:45 MA 004

Controlling spin excitations of a single molecular magnet — •Maximilian Kögler, Nicolas Néel, and Jörg Kröger — Institut für Physik, TU Ilmenau, Ilmenau, D-98693, Germany

A single molecular magnet (SMM) with a robust spin S=1 was attached to the tip of a scanning tunneling microscope. The spin excitation spectrum was probed by inelastic electron tunneling spectroscopy as a function of the vertical distance to the metal surface, an adsorbed atom and an artificially crafted metal cluster. Above the clean metal surface the SMM spin excitation reduces its energy upon decreasing the distance. Possibly, the magnetic anisotropy energy is affected by relaxations of the junction geometry. In contrast, the excitation spectrum remains invariant atop the adsorbed atoms and vanishes abruptly upon collapsing the tunneling barrier. Funding by the Deutsche Forschungsgemeinschaft through KR 2912/21-1 is acknowledged.

O 43.3 Wed 11:00 MA 004

Spin engineering in artificial atom-molecule hybridsS — •Wantong Huang[1], Máté Stark[1], Paul Greule[1], Daria Sostina[2], Constantin Weeber[1], Jose Gálvez[3], Christoph Sürgers[1], Christoph Wolf[3], Wolfgang Wernsdorfer[1], and Philip Willke[1] — [1]Physikalisches Institut (PHI), Karlsruhe Institute of Technology, Karlsruhe, Germany — [2]Institute of Quantum Materials and Technologies (IQMT), Karlsruhe Institute of Technology, Karlsruhe, Germany — [3]Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul, Republic of Korea

Protecting individual qubits from decoherence and relaxation is one of the crucial challenges for quantum information processing. Both, individual atoms as well as magnetic molecules consisting of one central spin surrounded by ligands, are promising building blocks and offer a route for nanoscale spintronics and future quantum devices. Here, we explore the magnetic properties and spin dynamics of artificially built hybrids of atomic and molecular spins. The resulting magnetic system consisting of an FePc molecule coupled to an Fe atom, forms a mixed spin(1/2,1) Heisenberg antiferromagnet. We show that the strong exchange coupling leads to a ground-state doublet. Thus, this hybrid behaves effectively like a spin-1/2 system which can be driven into electron spin resonance by means of scanning tunneling microscopy. Strikingly, the relaxation time of the hybrid is significantly enhanced compared to pristine FePc reaching 1 μs. Moreover, we show by building dimers of hybrids that their spins can be coupled providing a path to realizing larger structures.

O 43.4 Wed 11:15 MA 004

Paramagnetic resonance of individual organic molecules driven by quantum spin torque — •Stepan Kovarik[1], Richard Schlitz[1,2], Aishwarya Vishwakarma[1], Dominic Ruckert[1], Pietro Gambardella[1], and Sebastian Stepanow[1] — [1]Department of Materials, ETH Zurich, Switzerland — [2]Department of Physics, University of Konstanz, Germany

The transfer of angular momentum between the current of spin-polarised electrons and spins of a magnetic material, the so-called spin torque, is used to control the magnetisation of modern magnetic memory devices [1]. Spin torque has also been used to excite magnetic resonance in ferromagnetic devices. [2]. Here, we use spin torque to drive the paramagnetic resonance of a single spin in a pentacene molecule adsorbed on ultrathin MgO with a time-dependent spin current injected from the tip of a scanning tunneling microscope. This novel approach to controlling single spins complements the well-established magnetic resonance driven by a time-dependent electromagnetic field. The spin-torque-driven resonance facilitates the observation of paramagnetic resonance even in regimes where the thermal polarisation of the measured spin is low. The observed signatures provide the first experimental insights into the spin-torque-driven resonance at the quantum level, previously studied only theoretically [3]. [1]D. C. Ralph, M. D. Stiles, JMMM. 320, 1190-1216 (2008). [2]T. Chen et al., Proc. IEEE. 104, 1919-1945 (2016). [3]A. M. Shakirov et al., PRB. 99, 054434 (2019).

O 43.5 Wed 11:30 MA 004

Magnetic resonance imaging of single organic radicals on a surface — •Christoph Wolf[1,2], Gregory Czap[3], Jose Reina-Gálvez[1,2], and Christopher P. Lutz[3] — [1]Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul, 03760 Republic of Korea — [2]Ewha Womans University, Seoul, 03760 Republic of Korea — [3]IBM Research Division, Almaden Research Center, San Jose, CA, 95120 USA

The combination of electron spin resonance and scanning tunneling microscopy resulted in a versatile surface probe with sub-nm spatial and μeV energy resolution. In this work, we will show that this technique can be applied to characterize singly occupied molecular orbitals of organic radicals. We show the general applicability of this technique by applying it across a series of small fused hydrocarbons adsorbed on thin layers of magnesium oxide used to decouple the spins from the underlying silver substrate. We utilize ab initio calculation and transport simulations using non-equilibriums Green*s functions to explain spatial and spectral variation of the magnetic resonance signal. Our work is the first demonstration of magnetic resonance imaging of a single molecular orbital with sub-molecular resolution of an organic radical of a surface.

O 43.6 Wed 11:45 MA 004

Magnetic resonance imaging of an electron spin residing in an extended molecular orbital — •Richard Schlitz[1,2], Stepan Kovarik[1], Aishwarya Vishwakarma[1], Dominic Ruckert[1], Pietro Gambardella[1], and Sebastian Stepanow[1] — [1]Department of Materials, ETH Zürich, 8093 Zürich, Switzerland — [2]Department of Physics, University of Konstanz, 78457 Konstanz, Germany

The combination of a scanning tunneling microscopy (STM) and electron paramagnetic resonance (EPR) allows addressing individual spins on surfaces with sub-μeV energy resolution and pm spatial resolution. Recent works demonstrated that EPR-STM can be used for magnetic resonance imaging (MRI), which reveals the interaction between the magnetic STM tip and the investigated spin species [1]. However, until now imaging was only performed for spins that were localized in an atomic orbital. In this talk, we will show the MRI of pentacene on two monolayers of MgO on Ag(001) by EPR-STM. Pentacene on MgO is singly charge with the electron residing in a delocalized molecular orbital. We discuss how the resonant slice is modified by the extended nature of the electron orbital and critically highlight differences to the atomic systems [2].

[1] Willke et al., Nature Physics 15, 1005-1010 (2019).

[2] Kovarik, Schlitz et al., submitted.

O 43.7 Wed 12:00 MA 004

Single-molecule electron-spin resonance by means of atomic force microscopy — Lisanne Sellies, •Sonja Bleher, and Jascha Repp — University of Regensburg, Regensburg, Germany

Recently, we combined electron spin resonance (ESR) and atomic force microscopy (AFM). To this end we drove electron spin transitions between the non-equilibrium triplet states of single molecules, which differ in their lifetimes. Driving these transitions changes the overall triplet lifetime [1], which can be read out with AFM using an electronic pump-probe scheme [2]. The resulting ESR-AFM spectra exhibit a sub-nanoelectronvolt spectral resolution and allow us to distinguish molecules that only differ in their isotopic configuration, as we demonstrated for pentacene [3].

This high spectral resolution allows us to detect minor influences of the local environment. For example, changing the voltage between the tip and the sample, we detected a considerable Stark shift. This Stark shift together with the cantilever oscillation likely contributes substantially to the small but finite linewidth. In this contribution, recent results obtained by ESR-AFM will be presented.

References:
[1] Köhler (1999). Physics Reports, 310, 261-339.
[2] Peng et al. (2021). Science, 373, 452-456.
[3] Sellies et al. (2022). arXiv, arXiv:2212.12244.

O 43.8 Wed 12:15 MA 004

Three-spin model for Dy atom adsorbed on graphene/Ir(111) — •Jindrich Kolorenc — Institute of Physics (FZU), Czech Academy of Sciences, Prague, Czech Republic

The inelastic electron tunneling spectroscopy (IETS) revealed that rare-earth atoms adsorbed on graphene/Ir(111) carry magnetic moments not only in their 4f shell but also in their valence shell(s) [1]. The measured IETS spectra can be accurately reproduced by assuming that there is one unpaired electron in the 6s orbital (or, more accurately, in some 6s–5d hybrid orbital) [1,2]. Recently, it was argued that to understand the spin dynamics in these adatoms, two separate spin-1/2 moments are needed to be present in the valence shells, probably one in the 6s orbital and one in the 5d orbitals [3]. Using theoretical modeling, we analyze what implications this additional spin has for the IETS spectra. We show that there should be one more step in the $\mathrm{d}I/\mathrm{d}V$ curve in addition to those reported in [1]. Combining first-principles and empirical parameters, we estimate the energy of this additional excitation.

[1] M. Pivetta *et al.*, Phys. Rev. X **10** (2020) 031054
[2] D. Kyvala, J. Kolorenc, `https://www.dpg-verhandlungen.de/year/2023/conference/skm/part/o/session/33/contribution/6`
[3] A. Curcella *et al.*, Phys. Rev. Lett. **130** (2023) 106702

O 43.9 Wed 12:30 MA 004

Electrically Driven Spin Resonance of 4*f* Electrons in a Single Atom on a Surface — •Stefano Reale[1,2,3], Jiyoon Hwang[1,4], Jeongmin Oh[1,4], Harald Brune[5], Andreas J. Heinrich[1,4], Fabio Donati[1,4], and Yujeong Bae[1,4] — [1]Center for Quantum Nanoscience (QNS), Institute for Basic Science (IBS), Seoul 03760, Republic of Korea — [2]Ewha Womans University, Seoul 03760, Republic of Korea — [3]Department of Energy, Politecnico di Milano, Milano 20133, Italy — [4]Department of Physics, Ewha Womans University, Seoul 03760, Republic of Korea — [5]Institute of Physics, Ecole Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland

Lanthanide atoms on surfaces are an exceptional platform for atomic-scale magnetic information storage. However, their potential as qubits is yet unexplored due to the limited number of experimental set-ups that can coherently drive the spins of single adatoms. Through x-ray magnetic circular dichroism and multiplet calculations, we identified erbium (Er) atoms on MgO(100)/Ag(100) as promising candidates for quantum coherent operations. This is due to their magnetic ground state and the inherent decoupling from the environment typical of the $4f$ shell. Here we employed scanning tunneling microscope to drive electron spin resonance (ESR) in a single Er atom. We exploited the well characterized titanium (Ti) atom to engineer a suitable Ti-Er dimer through atomic manipulation. This architecture allows us, through magnetic coupling, to access the spin state of Er, driving and detecting ESR transitions on its elusive $4f$ spins.

O 43.10 Wed 12:45 MA 004

A straight forward method to read the nuclear qudit of 4f-single molecular magnets: $^{163}\mathrm{DyPc}_2$ — •Hongyan Chen[1], Simon Gerber[1], Philip Schmid[1], Vera Schmeiser[1], Svetlana Klyatskaya[1], Eufemio Moreno-Pineda[2], Mario Ruben[1,3], and Wulf Wulfhekel[1] — [1]Karlsruhe Institute of Technology , Germany — [2]Universidad de Panamá — [3]Centre Européen de Sciences Quantiques, Strasbourg

The nuclear spin in single molecular magnets (SMMs) has been used for quantum information processing and has so far been detected by transitions of the magnetic moment during sweeping the magnetic field [1]. We here present a faster method to directly read the nuclear spin (I=5/2) of $^{163}\mathrm{DyPc}_2$ using spin-polarized scanning tunneling microscopy (Sp-STM) without the need for a magnetic field. For (I=0) DyPc_2 on Au(111), we recently demonstrated that the Dy^{3+} electron spin can be read out by Sp-STM but loses remanence due to the non-Kramers nature [2]. Adding the nuclear spin of I=5/2 restores Kramers protection and shifts the magnetization curve of Dy^{3+} by the hyperfine field. Measuring the spin polarization of the Kondo state at vanishing magnetic field thus directly reveals the quantum state of the nucleus. We observe T_1 times in excess of minutes at 35 mK. First results on nuclear magnetic resonance transitions between the nuclear states inducing by radio frequency voltages injected into the tunneling junction will be presented. [1] R. Vincent et al,. Nature 488, 357 (2012); S.Thiele et al,. Science 344, 1135 (2014). [2] T. Frauhammer, H. Chen et al,. Phys.Rev.Lett. 127, 123201 (2021).

O 44: 2D Materials III: Electronic Structure (joint session O/TT)

Time: Wednesday 10:30–13:00 Location: MA 005

O 44.1 Wed 10:30 MA 005

Semiclassical theory for plasmons in inhomogeneous two-dimensional systems — •Tjacco Koskamp, Koen Reijnders, and Mikhail Katsnelson — Radboud University, Nijmegen, The Netherlands

We construct a general theory for plasmons in inhomogeneous two-dimensional systems. Plasmons, quantized collective oscillations of conduction electrons in solids, can be used to manipulate and control light. This requires heterostructures of nanometer size, which are by definition spatially inhomogeneous, and difficult to describe analytically. Here, we present a novel semi-analytical method to describe plasmons in two-dimensional inhomogeneous media within the framework of the Random Phase Approximation (RPA). Our approach [1] is based on the semiclassical approximation, which is formally applicable when the length scale of the inhomogeneity is much larger than the plasmon wavelength. Within this framework, we obtain a classical Hamiltonian that describes the dynamics of quantum plasmons, given by the Lindhard function with spatially varying parameters. The classical trajectories generated by this Hamiltonian can be viewed as the analog of rays in geometrical optics. By subsequently adding the wave-like character of the plasmons to these classical trajectories, we can describe, for instance, plasmon scattering and plasmonic bound states. As an example, we compute the differential cross section for plasmon scattering by a radially symmetric impurity.

[1] T. M. Koskamp, M. I. Katsnelson, K. J. A. Reijnders, Phys. Rev. B 108, 085414 (2023)

O 44.2 Wed 10:45 MA 005

Imaging dielectric near-field modes of hexagonal boron nitride by photoemission electron microscopy — •Yaolong Li[1,2], Martin Aeschlimann[2], Quan Sun[1], Yunan Gao[1], Xiaoyong Hu[1], and Qihuang Gong[1] — [1]Department of Physics, Peking University, Beijing, China — [2]Department of Physics and Research Center OPTIMAS, University of Kaiserslautern-Landau, Germany

Low-loss dielectric modes are important features and functional bases of fundamental optical components in on-chip optical devices. However, dielectric near-field modes are challenging to reveal with high spatiotemporal resolution and fast direct imaging. Here, we present a method to address this issue by applying time-resolved photoemission electron microscopy to a low-dimensional wide-bandgap semiconductor, hexagonal boron nitride (hBN). Using a low-loss dielectric planar waveguide as the fundamental structure, static vector near-field vortices with different topological charges and the spatiotemporal evolution of waveguide modes are directly revealed. With the lowest order vortex structure, strong nanofocusing in real space is realized. Near-vertical photoemission in momentum space and narrow spread in energy space are simultaneously observed due to the atomically flat surface of hBN and the small photoemission horizon set by the limited photon energies. Our approach provides a strategy for the realization of flat photoemission emitters. Revealing low-loss dielectric near-field modes of hexagonal boron nitride by photoemission electron microscopy. Nat. Commun. 14, 4837 (2023).

O 44.3 Wed 11:00 MA 005

Noble metal dichalcogenides: Optoelectronic and non-linear response — George de Coster[1], Stefan Heiserer[1], Simon Schlosser[1], Zdenek Sofer[2], Tanja Stimpel-Lindner[1], Georg Duesberg[1], and •Paul Seifert[1] — [1]Institute of Physics, University of the Bundeswehr Munich, Faculty of Electrical Engineering and Information Technology, Werner-Heisenberg-Weg. 39, 85577 Neubiberg, Germany — [2]Department of Inorganic Chemistry, University of Chemistry and Technology Prague, Technická 5, 166 28 Prague 6, Czech Republic

Noble metal dichalcogenides belong to the material class of layered 2D materials. In particular, these materials transition from direct band gap semiconductor to semi-metal with increasing layer thickness and were shown to host type-II Dirac semi-metallic behavior, as well as topological surface states and superconductivity [1,2]. We analyze the low-frequency optoelectronic response of PdTe2 using THz time domain spectroscopy. Frequency resolved measurements reveal signatures of second order non-linear response whose symmetry constrains are consistent with the occurrence of anisotropy in its optoelectronic response to THz radiation [3]. The latter is verified in polarization resolved THz spectroscopy. The optical response at higher energies likewise exhibits this anisotropy. Our results elucidate the spectral optoelectronic response in PdTe2 at low energies and discuss its anisotropy in light of its underlying symmetry constraints. References: [1] W. Zheng et al., PRB 97, 235154 (2018) [2] O.J. Clark et al., PRL 120, 156401 (2018) [3] C. Guo et al. Sci. Adv. 6, 36 (2020)

O 44.4 Wed 11:15 MA 005

On the the nature of transient and metastable nonequilibrium phases in 1T-TaS2 — •Tanusree Saha[1,2], Arindam Pramanik[3], Barbara Ressel[1], Alessandra Ciavardini[1], Primož Rebernik Ribič[4], and Giovanni De Ninno[1,4] — [1]University of Nova Gorica, 5270 Ajdovščina, Slovenia — [2]Universität Duisburg-Essen, 47057 Duisburg, Germany — [3]Tata Institute of Fundamental Research, Mumbai 400005, India — [4]Elettra Sincrotrone Trieste, 34149 Trieste, Italy

Photoexcitation of materials with complex ground states can drive them into new out-of-equilibrium phases. In this talk, I will present the characteristics of these phases and the recovery dynamics in a complex system, the charge density wave (CDW)-Mott insulator 1T-TaS2, studied using time- and angle-resolved photoemission spectroscopy. We observe strong similarities between the band structures of the transient phase and the structurally undistorted equilibrium phase, with evidence for the coexistence of insulating and metallic phases. Following the transient phase, we find that the restorations of Mott and CDW orders begin around the same time, highlighting that the Mott transition is tied to the CDW distortion. During recovery, a metastable phase, driven by the CDW lattice order, emerges but only in the strong photoexcitation regime and is a commensurate CDW-Mott insulating phase but with a smaller CDW amplitude. Finally, I will briefly discuss our future work, where we aim to study how the nature of the metastable phase and electron correlations in photoexcited 1T-TaS2 can be tuned by an external parameter, e.g. pump fluence.

O 44.5 Wed 11:30 MA 005

Self-hybridized exciton-polaritons in thin films of Ruddlesden-Popper-Perovskites — •Maximilian Black[1], Parsa Darman[2], Sara Darbari[2], and Nahid Talebi[1] — [1]Institute for Experimental and Applied Physics, Kiel University, 24118 Kiel, Germany — [2]Faculty of Electrical and Computer Engineering, Tarbiat Modares University, 14115-111 Tehran, Iran

Lead halide perovskites have emerged as platforms for exciton-polaritonic studies at room temperature thanks to their excellent photoluminescence efficiency and great synthetic versatility. In this work we find proof of strong exciton-photon coupling in cavities formed by the layered crystals themselves, a phenomenon known as self-hybridization effect. We use multi-layers of high quality Ruddlesden-Popper perovskites in their 2D crystalline form, benefiting from their quantum-well excitonic resonances and the strong Fabry-Perot resonances resulting from the total-internal-reflection at their smooth surfaces. Optical spectroscopy reveals bending of the cavity modes typical for exciton-polariton formation, and photoluminescence spectroscopy shows thickness dependent splitting of the excitonic resonance. Additionally, local optical excitation of the flakes in photoluminescence measurements unveils long in-plane propagation of the excited modes. In previous works the influence of the incident angle is often overlooked, motivating this work to focus on tuning the in-plane momentum of the incident light to the polaritonic resonances. We therefore pave the way towards an effective way to study the rich physics of exciton-polaritons in Ruddlesden-Popper 2D perovskites.

O 44.6 Wed 11:45 MA 005

Inelastic tunneling into polaronic bound states in single-layer MoS$_2$ — Camiel van Efferen[1], Arne Schobert[2], Tfyeche Tounsi[1], Michael Winter[2], Mark Georger[1], Affan Safeer[1], Christian Krämer[1], Jeison Fischer[1], Jan Berges[3], Thomas Michely[1], •Roberto Mozara[2], Tim Wehling[2,4], and Wouter Jolie[1] — [1]II. Physikalisches Institut, U zu Köln — [2]I. Institut für Theoretische Physik, U Hamburg — [3]U Bremen Excellence Chair, Bremen Center for Computational Materials Science, and MAPEX Center for Materials and Processes — [4]The Hamburg Centre for Ultrafast Imaging

The presentation delves into the nuanced conductivity of two-dimensional MoS$_2$, a prominent transition metal dichalgogenide, by examining its response to doping-induced variations. In particular, we explore the intriguing phenomena of polarons, quasiparticles that emerge from the interplay of electrons with lattice vibrations. Employing advanced techniques such as scanning tunneling microscopy and spectroscopy, we unveil the manifestation of polaronic bound states in metallic 2D MoS$_2$, shedding light on their stability and formation dynamics. The investigation is enriched by density-functional theory calculations with a recently developed electron-lattice downfolding technique, emphasizing the role of renormalized M-phonons in shaping the electronic landscape of metallic MoS$_2$. This synthesis of experimental insights and theoretical perspectives offers a comprehensive understanding of the interplay between electrons and phonons in 2D MoS$_2$.

O 44.7 Wed 12:00 MA 005

Strain-dependent electromechanical and optoelectronic properties of free-standing PtSe2 films — •Natalie Galfe, Stefan Heiserer, Maximilian Wagner, Michael Loibl, Silke Boche, Simon Schlosser, Oliver Hartwig, Tanja Stimpel-Lindner, Cormac Ó Coileáin, Kangho Lee, George de Coster, Paul Seifert, and Georg S. Duesberg — University of the Bundeswehr Munich

We report on the piezoresistive and optoelectronic properties of free-standing noble metal dichalcogenide PtSe$_2$ films under controlled strain. Bridges of polycrystalline PtSe$_2$ films with different geometries were fabricated directly on target substrates. The pre-structered platinum channels were selenized through thermally assisted conversion and the resulting PtSe$_2$ electrically contacted and underetched. The controlled strain of the bridges was induced by application of back-gate voltages. This makes them an excellent platform to study the impact of strain on transport and optoelectronic properties.

Increasing tensile strain shows a decrease in the electrical resistance, which is attributable to an enhancement of the density of states at the Fermi level. Raman analysis of the channel under increasing static strain displays a blue-shift of the Raman modes, which can be attributed to a decrease in effective film thickness which is supported by finite element simulations of the polycrystalline films. By applying AC gate voltages, the geometry-dependent eigenfrequencies of the bridges can be determined proving their expected mechanical oscillations. The results lead to a deeper understanding of this novel material class and serve as a platform for further applications.

O 44.8 Wed 12:15 MA 005

Direct visualization of conduction band electrons in gated single layer TMDC via micro ARPES — •Chakradhar Sahoo[1], Yann in 't Veld[2], Alfred J. H. Jones[1], Zhihao Jiang[1], Paulina E. Majchrzak[1], Kimberly Hsieh[1], Kenji Watanabe[3], Takashi Taniguchi[4], Yong P. Chen[1], Jill A. Miwa[1], Malte Rösner[2], and Søren Ulstrup[1] — [1]Department of Physics and Astronomy, Interdisciplinary Nanoscience Center, Aarhus University, 8000 Aarhus C, Denmark — [2]Institute for Molecules and Materials, Radboud University, 6525 AJ Nijmegen, the Netherlands — [3]Research Center for Functional

Materials, National Institute for Materials Science, Tsukuba 305-0044, Japan — [4]International Center for Materials Nanoarchitectonics, National Institute for Materials Science, Tsukuba 305-0044, Japan

Electric field induced doping effects in the electronic structure of single-layer (SL) semiconductors is crucial for electronic and optoelectronic applications. However, direct visualization of doped electronic structure remains challenging for in situ gated devices. Here, we apply in operando micrometer scale angle-resolved photoemission spectroscopy at the ASTRID2 light source to characterize the electronic structure of a SL WS2 gated device. Using micromechanical cleaving and transfer methods, the SL WS2 is partially contacted to a graphene top electrode and placed on a boron nitrite dielectric on a graphite back-gate. We directly visualize distinct conduction band populations, band gap renormalization and charge transfer processes across the bare WS2 and graphene/WS2 interface. Our observations provide a better understanding of band renormalization and carrier doping in 2D devices.

O 44.9 Wed 12:30 MA 005

Deexcitation of highly charged ions at surfaces — •Anna Niggas, Matthias Werl, Daniel Thima, Filip Vukovic, Matthias Bernhart, Friedrich Aumayr, and Richard A. Wilhelm — TU Wien, Institute of Applied Physics, Vienna, Austria

Strong electronic excitations at the nanoscale can trigger nanopore formation on 2D materials and their heterostructures. One possible way to achieve these excitations are impacts of slow highly charged ions (HCIs), e.g., Xe^{40+} ions: Upon approaching the surface, resonant electron transfer leads to a population of high-n shells of the ion, with subsequent radiative and non-radiative decay, resulting in energy deposition in the very first surface layers of the material.

To unravel these deexcitation dynamics, we employ a complex coincidence spectrometer to detect correlated pairs of HCIs transmitted through 2D materials and electrons emitted from the material due to the ion impact. Filtering options for ion charge state, scattering angle, and energy loss, as well as electron number and energy, can be used for a detailed analysis of deexcitation channels. For instance, a charge-state-separated analysis of the HCI-induced electron yield from graphene shows that the number of emitted electrons increases continuously with the number of electrons captured by the projectile, reaching up to $100\,e^-$ per incident ion. Furthermore, we observe a sudden increase in the electron yield for ions filling up their valence shell.

In this contribution, we will discuss how these coincidence measurements help us understand the deexcitation of HCIs and how we can use our method to access material properties.

O 44.10 Wed 12:45 MA 005

Ultrafast Electron Diffuse Scattering as a Tool for Studying Phonon Transport: Phonon Hydrodynamics and Second Sound Oscillations — •Laurenz Kremeyer[1], Tristan Britt[1], Bradley Siwick[1,2], and Samuel Huberman[3] — [1]Department for Physics, McGill University, Montreal, Canada — [2]Department for Chemistry, McGill University, Montreal, Canada — [3]Department of Chemical Engineering, McGill University, Montreal, Canada

Hydrodynamic phonon transport phenomena, like second sound, have been observed in liquid Helium more than 50 years ago. More recently second sound has been observed in graphite at over 200 K using transient thermal grating techniques[1]. In this work we explore the signatures of second sound in ultrafast electron diffuse scattering (UEDS) patterns. We use density functional theory and solve the Boltzmann transport equation to determine time-resolved non-equilibrium phonon populations and subsequently calculate one-phonon structure factors and diffuse scattering patterns to simulate experimental data covering the regimes of ballistic, diffusive, and hydrodynamic phonon transport. For systems like graphite, UEDS is capable of extracting time-dependent phonon occupancies across the entire Brillouin zone [2] and ultimately lead to a more fundamental understanding of the hydrodynamic phonon transport regime.

[1] Ding et al. *Nat. Comm.* **13** 285 (2022)

[2] René de Cotret et al. *Phys. Rev. B.* **100** 214115 (2019)

O 45: Ultrafast Electron Dynamics at Surfaces and Interfaces IV

Time: Wednesday 10:30–12:45 Location: MA 041

O 45.1 Wed 10:30 MA 041

Ultrafast Charge Transfer in Graphene/Ag/SiC Studied Using Time- and Angle-Resolved Photoemission Spectroscopy — •Eduard Moos[1], Zhi-Yuan Deng[1], Hauke Beyer[1], Kai Rossnagel[1,2,3], and Michael Bauer[1,2] — [1]Institute of Experimental and Applied Physics, Kiel University, 24098 Kiel, Germany — [2]Kiel Nano, Surface and Interface Science KiNSIS, Kiel University, 24118 Kiel, Germany — [3]Deutsches ElektronenSynchrotron DESY, Ruprecht Haensel Lab, 22607 Hamburg, Germany

Monoatomic metals intercalated between graphene/SiC interfaces represent a new type of van der Waals heterostructures with extraordinary properties. For instance, silver exhibits a metal-semiconductor transition in the monolayer limit (MLAg) [1, 2]. The low binding energy of the valence band maximum of MLAg, the intrinsic band gap of a bilayer graphene (BLGr) overlayer, and a twisted orientation of the Brillouin zone of silver and graphene overlayer make this heterostructure an interesting model system for the exploration of ultrafast interlayer charge transfer. In this contribution, we use time- and angle-resolved photoemission spectroscopy (TRARPES) with a time resolution of 35 fs to study such types of processes in n-doped BLGr/MLAg/6H-SiC. Following the decay of the photoexcited carriers, we observe clear signatures of a net charge transfer between Ag and graphene on a 50 fs timescale. The detailed analysis of the data allows disentangling and identifying the relevant pathways.

[1] P. Rosenzweig, U. Starke, Phys. Rev. B **101**, 201407(R) (2020).

[2] W. Lee *et al.*, Nano Lett. **22**, 19, 7841-7847 (2022).

O 45.2 Wed 10:45 MA 041

Influence of twist angle on ultrafast charge transfer in WS2-graphene heterostructures — •Niklas Hofmann[1], Leonard Weigl[1], Johannes Gradl[1], Neeraj Mishra[2,3], Stiven Forti[2], Camilla Coletti[2,3], Amir Klein[4], Daniel Hernangomez-Peres[4], Sivan Refaely-Abramson[4], and Isabella Gierz[1] — [1]University of Regensburg, Germany — [2]Istituto Italiano di Tecnologia, Pisa, Italy — [3]Istituto Italiano di Tecnologia, Genova, Italy — [4]Weizmann Institute of Science, Rehovot, Israel

Ultrafast charge separation is crucial for efficiently converting sunlight into electrical energy. This phenomenon commonly occurs in different van der Waals heterostructures [1] where the transfer rates for electrons and holes are determined by the band alignment and interlayer hybridization. Both parameters are expected to be highly sensitive with respect to variations of the twist angle between the layers. This makes the twist angle the decisive tuning parameter for optimizing ultrafast charge transfer processes for various applications. Using time- and angle-resolved photoemission spectroscopy, we investigate the non-equilibrium carrier dynamics in WS2-graphene heterostructures with twist angles of 0° and 30°. We find that, for a twist angle of 0°, hole transfer from WS2 to graphene is significantly faster than electron transfer [2]. For a twist angle of 30°, however, we find no indication for ultrafast charge separation. We interpret our results with the help of ab-initio band structure calculations.

[1] C. Jin et al., Nature Nanotechnology 13, 994 (2018)

[2] S. Aeschlimann et al., Sci. Adv. 6, eaay0761 (2020)

O 45.3 Wed 11:00 MA 041

Probing electron-hole Coulomb correlations in the exciton landscape of a twisted semiconductor heterostructure — •Jan Philipp Bange[1], David Schmitt[1], Wiebke Bennecke[1], Giuseppe Meneghini[2], AbdulAziz AlMutairi[3], Daniel Steil[1], Sabine Steil[1], R. Thomas Weitz[1], G. S. Matthijs Jansen[1], Stephan Hofmann[3], Samuel Brem[2], Ermin Malic[2], Marcel Reutzel[1], and Stefan Mathias[1] — [1]Georg-August-Universität Göttingen, I. Physikalisches Institut, Germany — [2]Philipps-Universität Marburg, Germany — [3]University of Cambridge, U.K.

An exciton is a two-particle correlated state between an electron and a hole. In the type-II band aligned WSe_2/MoS_2 heterostructure, interlayer excitons can be formed after the resonant excitation of the WSe_2 intralayer excitons. In this process, the exciton's electron transfers across the interface while the hole remains rigid in the MoS_2 layer [1]. Moreover, the inverted process is possible: After exciting MoS_2

intralayer excitons, charge transfer of the exciton's hole across the interface leads to the formation of interlayer excitons. In this case, the exciton's electron remains rigid in WSe_2.

Here, we employ time-resolved momentum microscopy to study the ultrafast hole-transfer mechanism. Interestingly, we find a distinct photoemission feature that can only be described when considering the break-up of the Coulomb correlation between the exciton's electron and hole during the photoemission process [2].

[1] Schmitt, Bange et al., Nature 608, 499-503 (2022).

[2] Bange et al., arXiv:2303.17886 (2023).

O 45.4 Wed 11:15 MA 041

Effect of dynamical screening on the exciton-phono coupling in a layered semiconductor — •Selene Mor[1,2], Valentina Gosetti[1,2,3], Alejandro Molina-Sanchez[4], Davide Sangalli[5], Simona Achilli[5], and Stefania Pagliara[1,2] — [1]Università Cattolica del Sacro Cuore, Brescia, Italy — [2]I-Lamp Research Center, Brescia, Italy — [3]KU Leuven, Leuven, Belgium — [4]University of Valencia, Valencia, Spain — [5]University of Milan, Milan, Italy

The light-mediated interaction of fermionic and bosonic excitations governs the optoelectronic properties of condensed matter systems. In photoexcited semiconductors, the coupling of electron-hole pairs (excitons) to coherent optical phonons enables a modulation of the excitonic resonance on the picosecond timescale [1]. At the same time, due to the Coulombic nature of excitons, their dynamics are sensitive to transient changes in the screening by the photoexcited carriers. Interestingly, the effect of interplay between exciton-phonon coupling and dynamical screening is still elusive. By means of broadband transient reflectance spectroscopy, we set the spectral evidences of either the breaking or the activation of exciton-phonon coupling in a layered semiconductor, and we reveal that dynamical screening can favor the phonon-mediated modulation of the excitonic resonance. These findings move a step forward on the path towards the optical control of fundamental interactions on the ultrafast timescale. [1]Mor, S. et al., Phys. Rev. Research 3, 043175 (2021)

O 45.5 Wed 11:30 MA 041

Ultrafast nano-imaging of dark excitons — •David Schmitt[1], Jan Philipp Bange[1], Wiebke Bennecke[1], Giuseppe Meneghini[2], AbdulAziz AlMutairi[3], Marco Merboldt[1], Jonas Pöhls[1], Sabine Steil[1], Daniel Steil[1], R. Thomas Weitz[1], Stephan Hofmann[3], Samuel Brem[2], G. S. Matthijs Jansen[1], Ermin Malic[2], Stefan Mathias[1], and Marcel Reutzel[1] — [1]Georg-August-Universität Göttingen, I. Physikalisches Institut, Germany — [2]Fachbereich Physik, Philipps-Universität, Marburg, Germany — [3]Department of Engineering, University of Cambridge, Cambridge CB3 0FA, U.K.

The optical response of transition-metal dichalchodgenide (TMDs) heterostructures is determined by the formation of tightly-bound electron-hole pairs, or so-called excitons. For the application of TMDs in optoelectronic devices it is necessary to understand not only the temporal evolution of the formation, thermalization and relaxation of excitons within energy- and momentum-space [1], but also the impact of spatial inhomogeneities to the exciton landscape and ultrafast dynamics on the relevant nanometer to micrometer spatial scale. In this talk, we will demonstrate how our novel method of ultrafast dark-field momentum microscopy [2] can be used to track the ultrafast formation dynamics of excitons with unparalleled simultaneous spatio-temporal and spatio-spectral information.

[1] Schmitt *et al.*, Nature 608, 499-503 (2022)

[2] Schmitt *et al.*, arXiv:2305.18908 (2023)

O 45.6 Wed 11:45 MA 041

Influence of defects on ultrafast charge separation in van der Waals heterostructures — •Johannes Gradl[1], Niklas Hofmann[1], Leonard Weigl[1], Raül Perea-Causín[2], Ermin Malic[3], Daniel Hernangómez Pérez[4], Sivan Refaely-Abramson[4], and Isabella Gierz[1] — [1]University of Regensburg — [2]Chalmers University of Technology — [3]University of Marburg — [4]Weizmann Institute of Science

Ultrafast charge separation is crucial for efficiently converting sunlight into electrical energy. This phenomenon commonly occurs in different van der Waals heterostructures [1] where the transfer rates for electrons and holes are determined by the band alignment and interlayer hybridization. Recently, it has been realized that defects might enhance the lifetime of the charge separated state significantly [2,3]. A detailed microscopic understanding of defect-assisted charge transfer, however, is lacking. We deliberately generate sulfur vacancies in a prototypical WS2-graphene heterostructure and probe their influence on the charge transfer dynamics with time- and angle-resolved photoemission spectroscopy. We interpret our results based on existing theory [4,5].

[1] Nat. Nanotechnol. 13, 994 (2018)

[2] Sci. Adv. 7, eabd9061 (2021)

[3] Phys. Rev. Lett. 127, 276401 (2021)

[4] Phys. Rev. B 107, 075419 (2023)

[5] Nano Lett. 23, 5995 (2023)

O 45.7 Wed 12:00 MA 041

Ultrafast SHG imaging microscopy: imhomogeneities in space and time — •Marleen Axt[1], Markus B. Raschke[2], Gerson Mette[1], and Ulrich Höfer[1] — [1]Fachbereich Physik, Philipps-Universität Marburg, Germany — [2]Department of Physics and JILA, University of Colorado, Boulder, USA

Ultrafast processes in 2D materials such as exciton or charge-transfer dynamics exhibit a broad range of time scales. They have been found to vary from sample to sample, are substrate dependent, or influenced by edges, grain boundaries and defects. Distinguishing the influence of these extrinsic effects from the intrinsic carrier dynamics has been challenging. In particular for 2D heterostructures, the emergent quantum phenomena are controlled by interlayer coupling and crystallographic orientation that are particularly sensitive to structural heterogeneities. Here, we present multi-scale imaging in a combination of time-resolved second-harmonic generation microscopy with nano-optical imaging to probe WS_2/WSe_2 heterostructures that reveal a large influence of different inhomogeneities on carrier dynamics.

O 45.8 Wed 12:15 MA 041

Coherent Time- and Angle-Resolved Photoelectron Spectroscopy in a Low Energy Electron Microscope — •Alexander Neuhaus[1], Pascal Dreher[1], Florian Schütz[2], Helder Marchetto[2], Torsten Franz[2], Michael Horn-von Hoegen[1], and Frank J. Meyer zu Heringdorf[1,3] — [1]Faculty of Physics and Center for Nanointegration, Duisburg-Essen (CENIDE), University of Duisburg-Essen, 47048 Duisburg, Germany — [2]ELMITEC Elektronenmikroskopie GmbH, 38678 Clausthal-Zellerfeld, Germany — [3]Interdisciplinary Center for the Analytics on the Nanoscale (ICAN), 47057 Duisburg, Germany

In modern photoemission microscopes the electron optics allows different imaging modes such as real space imaging or momentum microscopy. Here, we describe how a spectroscopic and low energy electron microscope (SPE-LEEM) can be equipped with an additional slit at the entrance of the hemispherical analyzer to enable a ARPES like mode with micrometer spatial selectivity while maintaining all other imaging modes. We use a photogrammetric calibration to correct for image distortions caused by the magnetic projective system and to calibrate the momentum and energy axis. We demonstrate the capabilities of the new ARPES mode by analyzing the time-resolved nonlinear electron emission from a plasmonic nano-focus on a Au(111) platelet using a femtosecond laser.

O 45.9 Wed 12:30 MA 041

Modelling the temporal evolution of photoemission momentum maps of CuPc/Cu(001)-2O — •Alexa Adamkiewicz[1], Miriam Raths[2], Marcel Theilen[1], Lasse Münster[1], Monja Stettner[2], Sabine Wenzel[2], Mark Hutter[2], Sergey Soubatch[2], Christian Kumpf[2], Francois C. Bocquet[2], Robert Wallauer[1], F. Stefan Tautz[2], and Ulrich Höfer[1] — [1]Philipps-University Marburg, Germany — [2]Peter Grünberg Institute (PGI-3), Jülich Research Centre, Germany

Using time-resolved photoemission orbital tomography on a well-ordered monolayer of CuPc on Cu(001)-2O, we find a temporal evolution of the momentum pattern which gradually changes from a HOMO into a LUMO pattern [1]. This can be traced back to the presence of two excitation pathways with different dynamics: transient population of the CuPc LUMO via resonant HOMO→LUMO transition and coherent two-photon photoemission from the HOMO. For a theoretical description of the ultrafast electron dynamics, we numerically solve the optical Bloch equations for a three-level system, using a density matrix approach. This allows us to disentangle photoemission contributions from both excitation pathways. The model well reproduces the respective momentum distribution patterns as well as the measured shift in kinetic energy of photoelectrons for different detuning of the pump photon energy with respect to resonant excitation.

[1] A. Adamkiewicz *et al.*, J. Phys. Chem. C **127**, 20411 (2023).

O 46: Plasmonics and Nanooptics IV: Fabrication and Applications

Time: Wednesday 10:30–13:00 Location: MA 042

O 46.1 Wed 10:30 MA 042

Electron-beam-induced synthesis and characterization of random plasmonic gold nanoparticle assemblies — •Kristina Weinel[1,2], Johannes Schultz[2], Daniel Wolf[2], Leonardo Agudo Jácome[1], Axel Lubk[2,3], and Bernd Büchner[2,3] — [1]Federal institute of materials research and testing, Berlin, Germany — [2]Leibniz institute for solid state and materials research Dresden, Dresden, Germany — [3]Institute of solid state and materials physics, Technische Universitaet Dresden (TUD), Dresden, Germany

Several studies have been shown that the electron beam can be used to create nanomaterials from microparticle targets in situ in a transmission electron microscope (TEM). Here, we show how this method has to be modified in order to synthesize plasmonic gold nanoparticles (NPs) on insulating silicon oxide substrate by employing a scanning electron microscope with a comparatively low acceleration voltage of 30 kV.

The synthesized NPs exhibit a random distribution around the initial microparticle target: Their average size reduces from 150 nm to 3 nm with growing distance to the initial Au microparticle target. Similarly, their average distance increases. The synthesized NP assemblies therefore show distinctly different plasmonic behaviour with growing distance to the target, which allows to study consequences of random hybridization of surface plasmons in disordered system, such as Anderson localization. To reveal the surface plasmons and their localization behaviour we apply electron energy loss spectroscopy in the TEM.

O 46.2 Wed 10:45 MA 042

Controlling Nanoparticle Distance by On-surface DNA-origami Folding — •Zhe Liu[1], Zunhao Wang[2], Jannik Guckel[2], Daesung Park[2], Ziba Akbarian[1], Uta Schlickum[1], and Markus Etzkorn[1] — [1]Technische Universität Braunschweig, 38106 Braunschweig, Germany — [2]Physikalisch-Technische Bundesanstalt, 38116 Braunschweig, Germany

This study introduces a novel method that combines top-down lithography with bottom-up DNA origami techniques to achieve accurate manipulation of nano-objects on pre-patterned silicon surfaces. Using electron beam lithography, we determined specific sites where DNA origami-nanoparticle hybrid nanostructures can adsorb, enabling precise and controlled arrangement of adsorption patterns in several dimensions. The specific adsorption of the DNA origami leads to controllable deformations if the origami helices are defined by the shape of the pre-patterned adsorption site. This approach also works for origami functionalized with two gold nanoparticles (AuNPs). This unique behavior allows for tunable assembly of plasmonic dimer nanoarrays, showing the ability to manipulate the center-to-center distance of AuNPs dimers on the origami template. The efficiency and precision of this technique were confirmed with Raman spectroscopy of dye molecules coated on the AuNPs. The results show the great potential for creating nanoarrays with precise control over nanoscale dimensions and orientation. This opens up new possibilities for using these nanoarrays in fields such as biomedicine and nano-photonics.

O 46.3 Wed 11:00 MA 042

Comparison of plasmonic and dielectric phase-change materials by modifying magnetic infrared resonances — •Lukas Conrads, Andreas Hessler, Matthias Wuttig, and Thomas Taubner — I. Institute of Physics (IA), RWTH Aachen University

For miniaturized active nanophotonic components, resonance tuning of nanoantennas is a key ingredient. Phase-change materials (PCMs) have been established as prime candidates for non-volatile resonance tuning based on a change in refractive index [1]. Currently, a novel material class of switchable infrared plasmonic PCMs, like In_3SbTe_2 (IST), is emerging. Since IST can be locally optically switched between dielectric (amorphous phase) and metallic (crystalline phase) states in the whole infrared range, it becomes possible to directly change the geometry and size of nanoantennas to tune their infrared resonances [2]. Here, we demonstrate tuning magnetic dipole (MD) resonances of split-ring resonators (SRRs) by modifying the arm length of IST SRRs [3] and by locally addressing the antenna hotspots of aluminum SRRs covered by amorphous $Ge_3Sb_2Te_6$ [4]. Finally, we compare both PCMs by investigating the MD resonance tuning of slit antennas [5]. Our concepts are well-suited for rapid prototyping, speeding up workflows for engineering ultrathin, tunable, plasmonic devices for infrared nanophotonics, telecommunications or (bio)sensing.

[1] Wuttig et al., *Nat. Photon.* **11**, 465 (2017) [2] Heßler et al., *Nat. Commun.* **12**, 924 (2021) [3] Heßler, Conrads et al., *ACS Photon.* **9**, 5 (2022) [4] Conrads et al., *Adv. Opt. Mat.* **11**, 17 (2023) [5] Conrads et al., *ACS Nano* submitted

O 46.4 Wed 11:15 MA 042

Towards a Dynamic and Switchable All-Optical Image Processing Device — •Dominik Ludescher[1], Lukas Wesemann[2], Lincoln Clark[2], Mario Hentschel[1], Ann Roberts[2], and Harald Giessen[1] — [1]4th Physics Institute and Research Center SCoPE, University of Stuttgart, Germany — [2]School of Physics, the University of Melbourne, Melbourne, VIC 3010, Australia

The urge for all-optical image filtering without the need for postprocessing is increasing with the requirement for fast operation, reliability, and robustness. This study unveils a fascinating new approach towards switchable image filtering devices drawing inspiration from the concept of a Salisbury screen. This novel method paves the way to real-time dynamic image processing holding immense potential in various fields of applications such as microscopy, facial recognition, or biological imaging. The general concept is based on the possibility of altering the properties of a conducting polymer by driving its inherent electrochemical redox reaction. By simply changing the applied voltage, the material properties such as the refractive index can be adapted. This change in the refractive index can be directly utilized to adapt the functionality of devices such as an optical filter. Besides turning the operation of a system fully on and off, a gradual change of the refractive index can be used to continuously vary the performance of the image processing device. Combining this approach with the concept of Fourier filtering certain spatial frequencies known from static approaches such as the Salisbury screen generates the possibility of working our way towards devices with adaptive properties.

O 46.5 Wed 11:30 MA 042

Accelerating Plasmonic Hydrogen Sensors by Transformer-Based Deep Learning — •Viktor Martvall[1], Henrik Klein Moberg[1], Athanasios Theodoridis[1], David Tomeček[1], Pernilla Ekborg Tanner[1], Sara Nilsson[1], Giovanni Volpe[2], Paul Erhart[1], and Christoph Langhammer[1] — [1]Department of Physics, Chalmers University of Technology, Gothenburg, Sweden — [2]Department of Physics, University of Gothenburg, Gothenburg, Sweden

Fast and accurate H_2 sensors are needed for H_2 technologies to address safety concerns associated with the high flammability of H_2-air mixtures. Plasmonic optical hydrogen sensors, monitoring H_2 through changes in the localized surface plasmon resonance peak of metallic nanoparticles absorbing hydrogen, shows promise. In idealized H_2-vacuum conditions, they have met the US department of energy's target of a response time $< 1\,\mathrm{s}$ for concentrations $< 0.1\,\mathrm{vol.\%}$ H_2. However, further advances are required to meet this target in a realistic environment, where the presence of other molecules slows down the sensor response. Here, we accelerate sensor response by developing a deep learning (DL) model to predict the H_2 % from the time-dependent extinction spectrum. We apply the DL model to a $Pd_{70}Au_{30}$ alloy plasmonic sensor in Ar carrier gas. Compared to the conventional analysis, collapsing each spectrum to a single spectral descriptor related to the H_2 % via a power law, our model demonstrates up to a 40 times faster sensor response time during rapid H_2 pulses. Also, we illustrate that it can faster discern and quantify gradual changes in H_2 %.

O 46.6 Wed 11:45 MA 042

Mie voids for all-optical sizing and counting of microplastics — •Mario Hentschel[1], Julian Karst[1], Lukas Wesemann[2], and Harald Giessen[1] — [1]4th Physics Institute and Research Center

SCoPE, University of Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany — [2]3ARC Centre of Excellence for Transformative Meta-Optical Systems, School of Physics, The University of Melbourne, Victoria 3010, Australia

Manipulating light on the nanoscale has become a central challenge in metadevices, resonant surfaces, nanoscale optical sensors and many more, and it is largely based on resonant light confinement in dispersive and lossy metals and dielectrics. In contrast, Mie void resonances observed in air-filled cavities in dielectric host materials have been shown to resonantly confine light in air, thus circumventing this loss and dispersion. One of the key benefits afforded by the Mie void concept is the full access to the model volume inside the air-filled void. This allows for maximised interaction of the modal intensities with analytes and other systems of interest, consequently allowing to maximise sensitivities. We demonstrate this potential for nanophotonic sensing and show that Mie voids are ideal sensors for the detection and sizing of nano- and micro-sized particles. We utilize Mie voids of different size and depth for the characterization of ensembles of polystyrene beads as model system for micro- and nanoplastic. We demonstrate that our ansatz allows for the all-optical sizing and counting of micro- and nanoplastic samples, thus being of large environmental importance.

O 46.7 Wed 12:00 MA 042

High-refractive Index Nanodisk Arrays with Hyperuniform Disorder — •Davy Tesch, Koundinya Upadhyayula, Bodo Fuhrmann, Alexander Sprafke, and Ralf Wehrspohn — Martin Luther University Halle-Wittenberg, 06120 Halle, Germany

Light-scattering metasurfaces with tailored disorder, especially Hyperuniform disorder (HuD), have recently garnered interest within the photonics community. HuD promises various properties that, until now, have only been associated with either periodic or random structures. The combination of strong diffraction from periodic structures and the spectrally broadband response of disordered structures holds promise for light scattering.

In this work, we experimentally investigate HuD nanodisk arrays made of amorphous silicon (a-Si). The high refractive index of a-Si enables the excitation of pronounced Mie resonances in the nanodisks. However, the optical response of the nanodisk array is significantly impacted by the arrangement of the nanodisks, namely the structure factor. The interaction between the form factor and HuD structure factor is central to our investigations. We employ a scalable fabrication process to experimentally prepare HuD nanodisk arrays using a-Si optimized for low absorption as the nanodisk material. Our results indicate a substantial impact of HuD on the optical response of the system. Furthermore, we also examine more complex systems, such as HuD nanodisk arrays at a defined distance above metallic mirrors.

O 46.8 Wed 12:15 MA 042

A numeric analysis of the angular distribution of dielectric Mie void structures — •Benjamin Reichel[1], Michelle Pfahl[1], Serkan Arslan[1], Adrià Canós Valero[1], Mario Hentschel[2], Thomas Weiss[2], and Harald Giessen[1] — [1]4th Physics Institute and Research Center SCoPE, University of Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany — [2]Institute of Physics, University of Graz, and NAWI Graz, Universitätsplatz 5, Graz 8010, Austria

Controlling the behavior of light at the nanoscale is a significant challenge in various applications such as metadevices, diffraction gratings and resonant surfaces. The confinement of light is one of the typical interactions used in such devices. Recently, Mie voids have emerged as a promising platform for confining electromagnetic waves, possibly extending down to ultraviolet wavelengths in air. Therefore understanding and theoretical modeling the electromagnetic scattering behaviour of Mie voids are crucial for their effective use. In this study, we conduct a numerical study of the scattering behaviour for a periodic dielectric Mie void metasurface, alongside simulations for single Mie voids using widely used commercial software, COMSOL Multiphysics, and an in-house MATLAB code. These insights will be important in the development of resonant meta-structure designs. Finally leveraging the full resolution of electromagnetic fields within the void will open avenues for novel spectroscopy and active manipulation strategies, for example for coupling single quantum emitters via Mie voids to directed radiative modes.

O 46.9 Wed 12:30 MA 042

Dissipation-engineered plasmonic ratchet — •Anna Sidorenko[1], Jan Mathis Giesen[2], Sebastian Eggert[2], and Stefan Linden[1] — [1]Physikalisches Institut, Universität Bonn, Kreuzbergweg 24, 53115 Bonn, Germany — [2]Physics Department and Research Center OPTIMAS, University of Kaiserslautern-Landau, 67663 Kaiserslautern, Germany

A ratchet effect is an ability to convert periodic drive into directed motion without a bias force. The working principle of a ratchet relies on the breaking of space- and time-reversal symmetry that would otherwise not allow a directed current. Based on the quantum-optical analogy, we propose a new design of a plasmonic ratchet. Our implementation features a trimerized lattice where losses are periodically varied while hopping amplitudes and on-site potentials are kept constant. The plasmonic structures were fabricated by means of two-step electron beam lithography. In the produced arrays, the spatial evolution of surface plasmon polaritons (SPPs) was recorded by leakage radiation microscopy. We observe both in numerical calculations and in our measurements a directional transport of SPPs in such an array in a single preferred direction. Control of directionality purely by tailored dissipation distinguishes our system from a simple combination of directional couplers. We examine the effect of different dissipation strengths and durations on the efficiency of transport.

O 46.10 Wed 12:45 MA 042

Very thin plasmonic films for optical mirrors: influence of the dielectric function on the spectral properties — •Manuel Gonçalves — Ulm University - Inst. of Experimental Physics, Albert-Einstein-Allee 11, 89081 Ulm, Germany

Metallic thin films combined with dielectric layers allow a large variety of optical effects, as broadband high reflectivity, narrow spectral dips due to surface plasmon resonances and almost total absorption by surface nanostructuring.

In this contribution, it is shown that very thin films of few nanometers thickness can produce broadband resonances with high contrast in the vis-NIR reflectance spectrum, when interacting with an underlying high reflectance mirror. However, the design of such colored mirrors cannot employ the classical dielectric function of the bulk material. Several effective medium theory (EMT) models have been proposed for quasi-percolated thin films. A study of these models and their application in the design of the reflectance spectra is presented.

O 47: Metal Substrates I

Time: Wednesday 10:30–12:15 Location: MA 043

O 47.1 Wed 10:30 MA 043

Studying facet-selective adsorption of CO on Cu using the random phase approximation — •Seungchang Han and Stefan Ringe — Department of Chemistry, Korea University

Electrochemical CO2 reduction is a promising sustainable strategy for producing valuable chemicals and fuels. Cu is the only catalyst that produces significant amounts of higher reduced products like ethylene, ethanol, or methane. The product selectivity depends critically on the Cu active site environment, defined by the coverage and distribution of the central *CO intermediate. Unfortunately, one of the most successful approaches to studying adsorption events, the Perdew-Burke-Ernzerhof (PBE) functional based on generalized gradient approximation (GGA), has been shown to benefit from error compensation, which can affect their transferability to unknown systems. It also leads to a wrong prediction of adsorption trends across different surface facets and adsorption sites. Adding many-body corrections based on the random phase approximation (RPA) has been shown to critically improve the prediction of adsorption energies and long-range interactions. In this work, we first present a simple extrapolation scheme to overcome the difficulty of getting converged RPA-based adsorption energies. We then demonstrate the first trends of adsorption energies as well as the coverage dependence and compare them to conventional functionals.

O 47.2 Wed 10:45 MA 043

Electronic Structure Benchmark Calculations for CO2 Reduction Products on Cu(111)-Clusters from Projection-Based Embedding Theory — •Elena Kolodzeiski and Christopher Stein — TU Munich, TUM School of Natural Sciences, Department of Chemistry, Lichtenbergstr. 4, 85748 Garching, Germany

Copper surfaces are promising catalysts for the conversion of carbon dioxide. Despite the immense interest in such systems, current theoretical approaches have limitations either in numerical cost or accuracy. We propose the use of a cluster model described by the projection-based embedding theory (PBET) as a balanced compromise. This approach involves partitioning the system into an active system and an environmental, focusing the computational efforts on the active subsystem. The system partitioning is based on our recently developed ACE-of-SPADE algorithm, which has been proven to enable consistent active orbital space selection even for such challenging systems as transition metal clusters. Here, we present a benchmark study validating the accuracy of the cluster embedding approach for calculating the binding energy of a series of CO2 reduction products on Cu(111)-clusters. The cluster approach is first verified by comparison to PBE plane-wave calculations and then extensively validated regarding the impact of different cluster models. We showed that the proposed QM/QM embedding strategy considerably improves the accuracy compared to PBE plane-wave calculations. PBET, in combination with the ACE-of-SPADE algorithm, provides a highly accurate method for studying chemical reactions on cluster models of metal surfaces.

Topical Talk O 47.3 Wed 11:00 MA 043

Adsorbate motors for unidirectional translation and transport — •Grant J. Simpson[1], Mats Persson[2], and Leonhard Grill[1] — [1]University of Graz, Graz, Austria — [2]University of Liverpool, Liverpool, UK

Inspired by their biological counterparts, artificial molecular motors aim to convert external energy into unidirectional motion. However, despite having some form of inherent directionality imprinted into their chemical structure, synthetic motors often lose functionality when taken from their native aqueous phase and deposited onto a metallic surface. Here, we demonstrate a new concept in molecular motors which uses a small and simple chemical structure to achieve perfect unidirectionality when deposited onto a Cu(110) surface [1]. The motion is observed and quantified using scanning tunnelling microscopy and is shown to be triggered by an intramolecular proton transfer. We further demonstrate that this unidirectional motion can be harnessed to perform meaningful work by transporting small cargo molecules across the surface.

[1] G. J. Simpson, M. Persson, L. Grill, Nature, 621, 82-86 (2023)

O 47.4 Wed 11:30 MA 043

Functionalization of Cu and CuxO surfaces with heterocyclic molecules: a joint experimental and computational study — •Sergio Tosoni[1], Felix Landwehr[2], Mowpriya Das[3], Maximilian Koy[3], Jared P. Bruce[2], Ankita Das[3], Juan J. Navarro[2], Markus Heyde[2], Gianfranco Pacchioni[1], Frank Glorius[3], and Beatriz Roldan Cuenya[2] — [1]Dipartimento di Scienza dei Materiali, Università di Milano-Bicocca, Via Cozzi 55, 20125 Milano, Italy — [2]Fritz-Haber Institute of the Max Planck Society, Department of Interface Science, Faradayweg 4 6, 14195 Berlin, Germany — [3]Westfälische Wilhelms-Universität, Organisch-Chemisches Institut Corrensstraße 40, 48149 Münster, Germany)

N-Heterocyclic Carbenes (NHCs) and Olefins (NHOs) form strong chemical bonds to surfaces, which enables their use in surface functionalization for gas sensing, optoelectronics and (photo)catalysis. We deposit and characterize NHCs on clean and oxidized copper. State-of-the-art DFT calculations are used to interpret the results of STM and XPS measurements. We compare the adsorption and diffusion of NHCs and NHOs on Cu, also considering the important effect of the steric hindrance exerted by the side substituents. NHCs are bound stronger than NHOs on clean copper. Interestingly, bonds to oxygen are preferentially formed by NHCs on an oxidized Cu surface. The intriguing different behaviour shown by NHCs on Cu(111) and Cu(100) is analysed in details.

O 47.5 Wed 11:45 MA 043

Cluster formation and ordering at high CO coverages on Ru(0001): A combined DFT and STM study — •Hannah Illner[1], Sung Sakong[2], Axel Gross[2], and Joost Wintterlin[1] — [1]Ludwig-Maximilians-Universität München, Germany — [2]Universität Ulm, Germany

Dense coverages of adsorbates on metal surfaces provide insight into the relation between metal-adsorbate and adsorbate-adsorbate interactions. Depending on the variation of the adsorption energy with binding site, different types of structures can be formed in dense layers, namely moiré structures, structures formed by domain boundaries, and cluster structures. On Ru(0001), CO at 0.58 monolayers (ML) forms a closed ring cluster structure consisting of seven molecules in $(2\sqrt{3} \times 2\sqrt{3})$R 30° symmetry. Here we present a study in which CO at higher coverages than 0.58 ML is investigated by low-temperature STM and DFT simulations. We identify six compact cluster structures, which are separated by CO-free Ru rows and a triangular arrangement of CO molecules at the junctions. We discuss how the local configurations stabilize the cluster configuration. By analyzing the Fourier transform of the STM image we provide a new explanation of the LEED pattern of the CO adlayer, that previously has been interpreted as resulting from a moiré structure. The newly identified structures clarify the long-standing conflict between the assumed structure and the vibrational spectroscopy data.

O 47.6 Wed 12:00 MA 043

Single-molecule study of Heck cross-coupling with Pd nano-islands on Au(111) surface — •Donato Civita[1], Friedrich Esch[2], and Leonhard Grill[1] — [1]Department of Physical Chemistry, University of Graz, Austria — [2]Fakultät für Chemie, TU München, Germany

The interaction of organic molecules with metallic structures is particularly important in heterogeneous catalysis. It has been shown that model heterogeneous catalysts based on metal single crystals can be investigated by scanning tunnelling microscopy (STM) to determine the role of steps as active sites for dissociation or to study a thermally induced Heck cross-coupling [1]. Moreover, nanostructured catalysts are advantageous to obtain specific electronic properties, a large number of undercoordinated sites, and to reduce the amount of precious catalyst material. However, instability effects like leaching of catalyst atoms from the nanostructure or sintering of small particle into large ones requires a systematic investigation at the atomic scale.

Here, these questions are addressed by studying a Heck cross-coupling reaction catalysed by Palladium nano-islands on Au(111) surface by low temperature STM. A statistical analysis of the products and the Pd islands size, together with STM tip manipulation experiments of single intermediates, give insight into the activity and stability of this catalyst, and the dependence on its morphology.

[1] Shi, K.-J., et al., On-Surface Heck Reaction of Aryl Bromides with Alkene on Au(111) with Palladium as Catalyst. Organic Letters, 2017. 19(11): p. 2801.

O 48: Focus Session: Spin Phenomena in Chiral Molecular Systems II (joint session O/TT)

Time: Wednesday 10:30–12:30 Location: MA 141

Topical Talk O 48.1 Wed 10:30 MA 141

Chiral-induced Spin Selectivity in Hybrid Chiral Molecule/ Metal Systems — Ashish Moharana[1], Yael Kapon[2], Fabian Kammerbauer[1], David Anthofer[1], Shira Yochelis[2], Yossi Paltiel[2], and •Angela Wittmann[1] — [1]Johannes Gutenberg Universität Mainz, Germany — [2]Hebrew University Jerusalem, Israel

The chiral-induced spin selectivity (CISS) effect has recently gained significant attention in the field of spintronics. The remarkably high polarization efficiency of chiral molecules via the CISS effect paves the path toward novel, sustainable hybrid chiral molecule magnetic applications. While research has predominantly focused on transport properties so far, in our work, we explore spintronic phenomena at hybrid chiral molecule magnetic interfaces to elucidate the underlying mechanisms of the chiral-induced spin selectivity effect. For this, we investigate the interfacial spin-orbit coupling in chiral molecule/metal thin film heterostructures by probing the chirality and spin-dependent spin-to-charge conversion. Our findings validate the central role of spin angular momentum for the CISS effect, paving the path toward the functionalization of hybrid molecule-metal interfaces via chirality.

Topical Talk O 48.2 Wed 11:00 MA 141

Chirality-induced spin selectivity at the single-molecule scale — •Daniel Emil Bürgler[1], Mohammad Reza Safari[1], Frank Matthes[1], Nicolae Atodiresei[1], Claus Michael Schneider[1], and Karl-Heinz Ernst[2] — [1]Peter Grünberg Institut, Forschungszentrum Jülich, Germany — [2]Molecular Surface Science Group, Empa, Dübendorf, Switzerland

Chirality-induced spin selectivity (CISS) leads to spin-selective electron transport in chiral molecules and enantiospecific adsorption on magnetic surfaces. To advance the development of theoretical models, well-defined single-molecule experiments are needed. Here, we report CISS effects for single chiral heptahelicene molecules that are sublimed under ultra-high vacuum onto uncoated single-crystalline and perpendicularly magnetized Co nanoislands. We use spin-polarized scanning tunneling microscopy (SP-STM) to (i) determine the handedness of individual heptahelicenes and the magnetization direction of the underlying Co nanoisland and (ii) measure spin-polarized transport through single molecules. Analysis of more than 740 molecules provides unequivocal evidence for enantioselective adsorption and reveals that enantioselection must occur in a physisorbed transient precursor state. $I-V$ curves of two enantiomers under otherwise identical conditions show at 5 K magnetochiral conductance asymmetries of up to 50% when either the molecular handedness is exchanged or the magnetization of the STM tip or Co substrate is reversed. The results demonstrate that CISS is a single-molecule effect and rule out electron-phonon coupling and ensemble effects as its primary mechanisms.

O 48.3 Wed 11:30 MA 141

Spin polarization through a helical molecule- functinalized tip dependence on the tip-sample distance observed by ambient STM — •Thi Ngoc Ha Nguyen[1], Lech Tomasz Baczewski[2], Olav Hellwig[3], and Christoph Tegenkamp[1] — [1]Analysis of Solid Surfaces, Nanostructures and Quantum Materials, Chemnitz University of Technology, Germany — [2]Institute of Physics, Polish Academy of Sciences, Warszawa, Poland — [3]Functional Magnetic Materials, Chemnitz University of Technology, Chemnitz, Germany

Polyalanine (PA) with an alpha-helix conformation has gathered recently a lot of interest as the propagation of electrons along the helical backbone structure comes along with spin polarization of the transmitted electron. However, studies on a molecular scale are still rare, although this length scale provides direct insight into the role of molecular properties. We studied now in detail with a PA molecule-functionalized Au tip on magnetic Au/Co/Au/Pt/Al2O3 substrates and probed the transmission by local spectroscopy (STS). Because of the high spatial resolution, our setup allows to study this CISS effect on the nanoscale and probe the importance of cooperative effects. Using this functionalized tip, we found that the spin polarization (SP) significantly varies with tip-sample distance. Interestingly, the SP through the self-assembled film of PA on the same substrate at different non-functionalized Au tip-sample distance doesn't show significant change. Our observation provides that the overlaping tip and sample orbitals, the coupling as well as the electric field strength in the close proximity of tip - sample surface take the main roles on this SP variation.

O 48.4 Wed 11:45 MA 141

Chirality-induced spin selective quantum capacitance — •Theiler Pius Markus — ETH Zürich, Zürich, Switzerland

The absence of symmetries has a significant impact on physics, particularly in chiral molecules or crystals lacking mirror symmetry. When an electric charge interacts with such chiral materials, the spin of the charge aligns even at room temperature. This spin polarization is known as chirality-induced spin selectivity (CISS). Although the exact mechanism behind the effect remains unclear, it may have played a role in the origin of life and significantly impacted biological processes. The effect has potential applications in chemical catalysis, renewable energy, and quantum technologies. This work aims to elucidate the key mechanism behind CISS surface potential changes upon toggling the enantiomer or magnetic polarization of the substrate. For the first time, chiral α-helical polypeptide films are investigated with time-resolved Kelvin-probe atomic force microscopy to probe the dynamics of the surface potential and a CISS quantum capacitance. This discovery of the CISS quantum capacitance leads to the conclusion that CISS is a persistent effect and paths the way to a fundamental reinterpretation of the CISS effect.

O 48.5 Wed 12:00 MA 141

Magnetization generation in helical molecular junctions — •Richard Korytár[1], Jan van Ruitenbeek[2], and Ferdinand Evers[3] — [1]Univerzita Karlova, Prague, Czech Republic — [2]Leiden University — [3]University of Regensburg

Despite extensive experimental and theoretical literature on the spin-selective transport in helical molecules [1], a satisfactory theoretical explanation of the effect is lacking [2]. We present analytical calculations of charge and spin conductances in a minimal model of a helical molecule with spin orbit coupling attached to non-magnetic leads. The calculations extend previous studies, which focused on spin-polarization [5,3]. The band-structure of the model exhibits spin-momentum locked bands analogous the the edge modes of a quantum spin Hall system. The spin currents in the left and right lead carry opposite signs and consequently both leads pick up parallel magnetizations (in linear response). We discuss the feedback of resulting spin accumulations in the leads to the charge current [4].

[1] Ron Naaman, Yossi Paltiel & David H. Waldeck, Nature Reviews Chemistry, volume 3, pages 250*260 (2019)

[2] Evers et al., Adv. Mater. 2022, 34, 2106629

[3] J. M. van Ruitenbeek, R. Korytár, F. Evers, J. Chem. Phys. 159, 024710 (2023)

[4] R. Korytár, J. M. Ruitenbeek, F. Evers, in preparation

[5] K. Michaeli and R. Naaman, J. Phys. Chem. C 123, 17043 (2019)

O 48.6 Wed 12:15 MA 141

Study of magneto-optical properties of cobalt-layers by adsorption of α-helical polyalanine self-assembled monolayers — •Lokesh Rasabathina[1], Apoorva Sharma[1], Julia Krone[1], Annika Morgenstern[1], Thi Ngoc Ha Nguyen[1], Markus Gössler[2], Karin Leistner[2], Christoph Tegenkamp[1], Georgeta Salvan[1], and Olav Hellwig[1] — [1]Institute of Physics, Chemnitz University of Technology, 09126 Chemnitz, Germany — [2]Institute of Chemistry, Chemnitz University of Technology, Chemnitz, 09111, Germany

High spin polarization in helical polyalanine molecules enables selective electron transport with a defined spin direction, a phenomenon known as Chirality Induced Spin Selectivity (CISS). This discovery holds promising implications for organic spintronic devices. Furthermore, the adsorption of pure enantiomers of α-helical polyalanine on a gold-covered ferromagnetic thin film, termed Magnetism Induced by the Proximity of Adsorbed Chiral molecules (MIPAC), can influence the magnetization of the ferromagnetic thin film. In our ongoing research, we are delving into the magnetic properties of thin films and attempt to increase the size of atomically smooth terraces at the Au(111) surface. By varying parameters such as deposition pressure and annealing temperature, we aim to understand how alterations in the Au surface affect the arrangement of molecules and how the molecule adsorption at the Au surface depends on the magnetic properties and state of the underlying magnetic thin film.

O 49: Oxide and Insulator Interfaces I

Time: Wednesday 10:30–13:00 Location: MA 144

O 49.1 Wed 10:30 MA 144

Structure of the Al_2O_3(0001) surface — Jan Balajka[1], Andrea Conti[1], Johanna Hütner[1], Florian Mittendorfer[1], Georg Kresse[2], •Michael Schmid[1], and Ulrike Diebold[1] — [1]Institute of Applied Physics, TU Wien, Austria — [2]Computational Materials Physics, Universität Wien, Austria

Corundum (α-Al_2O_3) is the thermodynamically stable form of aluminum oxide and used in many fields of technology. Nevertheless, the structures of its surfaces have not been determined yet. Our high-quality ncAFM images of the $Al_2O_3(0001)$-$(\sqrt{31}\times\sqrt{31})R\pm9^\circ$ reconstruction, which forms after high-temperature annealing, rule out the aluminum termination proposed previously [1,2]. The surface is very similar to ultrathin alumina films, which have well-established structural models [3,4]. To determine the structure of the subsurface layers not directly imaged by ncAFM, we employed density functional theory calculations and machine-learned force fields to search the large configuration space, resulting in a lowest-energy structure that fits the experiment. DFT calculations also show that the standard model for the unreconstructed $Al_2O_3(0001)$-(1×1) surface cannot be thermodynamically stable at any conditions. We discuss reasons for the high stability of the reconstruction.

[1] Renaud et al. Phys. Rev. Lett. 73, 1825 (1994)
[2] Lauritsen et al. Phys. Rev. Lett. 103, 076103 (2009)
[3] Kresse et al. Science 308, 1440 (2005)
[4] Schmid et al. Phys. Rev. Lett. 99, 196104 (2007)

O 49.2 Wed 10:45 MA 144

Metal-oxide surfaces: Chemical selective nc-AFM imaging with O-terminated copper tips — •Philipp Wiesener[1], Johanna Hütner[2], Jan Balajka[2], Bertram Schulze Lammers[1], Harald Fuchs[1], Saeed Amirjalayer[1], and Harry Mönig[1] — [1]Physikalisches Institut, Universität Münster, Germany — [2]Institute of Applied Physics, TU Wien, Austria

Previously we investigated the performance of various atomically defined tip terminations on the Cu(110)O(2x1) surface with non-contact AFM [1]. A direct comparison of the imaging properties shows, that only for an oxygen terminated copper tip (CuOx-tip [2]) a chemical selective contrast between metal and oxygen atoms on the surface can be observed.

In this work we exhibit the generalized ability of CuOx-tips for an outstanding characterization of metal-oxide surfaces. By performing constant height measurements on a variety of copper-, iron-, titanium- and aluminum-oxide systems with increasing structural complexity the robustness of the chemical selectivity is demonstrated. For an additional contrast analysis we complement our site-selective microscopy with force spectroscopy measurements on the observerd metal and oxygen atoms and investigate predominant defects.

Supporting DFT calulations reveal that the chemical selectivity can be explained by purely electrostatic interactions, which are determined by the strongly electronegative tip termination.

[1] B. Schulze Lammers et. al., Nanoscale 13, 13617 (2021).
[2] H. Mönig et al., ACS Nano 10, 1201-1209 (2015).

O 49.3 Wed 11:00 MA 144

Observation of a dodecagonal europium oxide quasicrystal on Pd(111) — •Martin Haller, Sebastian Schenk, Stefan Förster, and Wolf Widdra — Martin-Luther-Universität Halle-Wittenberg, 06120 Halle, Germany

Oxide quasicrystals (OQCs) are 2D oxide films with dodecagonal symmetry grown on a hexagonal metal substrate. The dodecagonal symmetry results from a square-triangle-rhombus tiling of alkaline earth atoms, as Ba or Sr, in a Ti-O network [1]. Europium has a comparable ionic radius as Sr and the same 2+ oxidation state and is an interesting OQC substituent due to its half-filled 4f shell resulting in a strong magnetic moment. Here, we show that an OQC forms by Eu deposition onto a Ti-O network on Pd(111) and subsequent annealing. We analyze the Eu-Ti-O OQC and compare it with the prototypical Ba-Ti-O system based on LEED and STM data. From LEED, we deduce a reduction of the characteristic length scale and a close match with the theoretical diffraction pattern. In real space, the tiling frequencies are found to be 2.80:1:3.78 for squares:triangles:rhombi. Those are in good agreement to the ideal ratio of 2.73:1:0.37 [2]. Lastly, we analyze the datasets in hyperspace, where we observe the typical confinement of the projected points inside the dodecagonal acceptance domain.

[1] Schenk et al., Nature Communications, 13, 7542 (2022)
[2] Schenk et al., Acta Crystallographica A, 75, 307 (2019)

O 49.4 Wed 11:15 MA 144

Atomic layer deposition of cerium oxide monitored by operando ellipsometry and in-situ X-ray photoelectron spectroscopy — •Rudi Tschammer[1], Yuliia Kosto[1], Carlos Morales[1], Marcel Schmickler[2], Karsten Henkel[1], Anjana Devi[2], and Jan Ingo Flege[1] — [1]Applied Physics and Semiconductor Spectroscopy, BTU Cottbus-Senftenberg, Cottbus, Germany — [2]Inorganic Materials Chemistry, Ruhr University Bochum, Universitätsstraße 150, Bochum, Germany

Atomic layer deposition (ALD) has been used extensively to grow homogeneous films with excellent coverage and atomic-scale thickness control for a variety of applications. However, remaining challenges include the investigation of novel precursor-oxidant combinations for low-temperature deposition as well as unraveling the complex interplay between substrate and coating for ultrathin films. In this work, we present a detailed investigation of ultrathin cerium oxide films grown using the novel $Ce(dpdmg)_3$ precursor with H_2O and O_2. Following a surface science-based approach, we have combined operando spectroscopic ellipsometry and in-situ X-ray photoelectron spectroscopy to allow rapid process optimization and determination of the complex relation between oxide stoichiometry, film thickness and ALD growth parameters, revealing a distinct dependence of inital Ce^{3+} content on the film thickness and choice of oxidant. This offers the possibility of adjusting the oxide properties to application requirements e.g. in gas sensing by choosing a suitable precursor-oxidant combination.

O 49.5 Wed 11:30 MA 144

Bridging the Pressure and Materials Gap in Heterogeneous Catalysis: A Combined UHV, In Situ, and Operando Study Using Infrared Spectroscopy — •Lachlan Caulfield, Eric Sauter, and Christof Wöll — Karlsruher Institut für Technologie (KIT)/Campus Nord, Hermann-von-Helmholtz-Platz 1,Eggenstein-Leopoldshafen, 76433,Germany

In the last decades, ceria-based systems have seen an increase in popularity, due to its unique redox behavior and catalytic characteristics. Ceria powders are widely used in exhaust catalysts as three-way catalysts (TWC) well as many other catalytic cycles, due to the cheap and simple preparation methods. To gain a better understanding of the chemical and structural behavior of ceria nanoparticles and their interaction with gaseous molecules, an operando DRIFTs study was carried out in combination with in-situ FT-IR studies. By tuning the temperature of the sample under a constant flow of carbon monoxide the reactions on the surface could be monitored in real time, without changing the surface. Notably, increasing the temperature from liquid nitrogen temperature to room temperature and above, it is possible to observe very minor changes to the oxidation state of the ceria nanoparticles as the surface becomes increasingly reduced. Using operando DRIFTS in combination with carbon monoxide dually as a reducing agent and as a probe molecule, the chemical and structural changes of polycrystalline ceria and other metal oxides can easily be investigated.

O 49.6 Wed 11:45 MA 144

Atomic-scale Structure of K-Feldspar Microcline (001) — •Tobias Dickbreder[1], Franziska Sabath[1], Bernhard Reischl[2], Rasmus V. E. Nilsson[2], Adam S. Foster[3,4], Ralf Bechstein[1], and Angelika Kühnle[1] — [1]Physical Chemistry I, Bielefeld University, Germany — [2]Institute for Atmospheric and Earth System Research/Physics, Faculty of Science, University of Helsinki, Finland — [3]Department of Applied Physics, Aalto University, Finland — [4]Nano Life Science Institute (WPI-NanoLSI), Kanazawa University, Japan

Ice nucleation governs the aggregate state of water in clouds and, thus, understanding ice nucleation is essential for improving climate models. Most atmospheric ice nucleation is heterogeneous caused by ice nucleating particles (INP). Among the most important INP under mixed-phase cloud conditions are feldspar minerals. The mechanism by which feldspar minerals facilitate ice nucleation, however, remains elusive. A prerequisite for unravelling this mechanism is a solid understanding of the surface structure of feldspar minerals. In this regard, it is especially crucial that experimental studies on the atomic-scale structure of feldspar minerals are sparse. Here, we present atomic force microscopy images of K-feldspar microcline (001) taken in ultrahigh vacuum. Our high-resolution AFM data reveal features consistent with a hydroxyl-terminated surface as predicted by density functional theory. This finding suggests that water in the residual gas readily reacts with the surface. These insights into the surface structure will contribute to understanding the excellent ice nucleating ability of feldspar minerals.

O 49.7 Wed 12:00 MA 144

Atomic-scale imaging of K-feldspar surfaces and their interaction with water — •Luca Lezuo[1], Andrea Conti[1], Rainer Abart[2], Florian Mittendorfer[1], Michael Schmid[1], Ulrike Diebold[1], and Giada Franceschi[1] — [1]Institute of Applied Physics, TU Wien, Wiedner Hauptstrasse 8-10/E134, 1040 Wien, Austria — [2]Department of Lithospheric Research, Universität Wien, 1090 Wien, Austria

K-Feldspars ($KAlSi_3O_8$) are common minerals in the Earth's crust and play a crucial role as ice nucleators in atmospheric processes. In particular, the well-ordered low-temperature polymorph microcline is a very effective ice nucleator.[1] Understanding their interaction with water is essential for various scientific fields, including atmospheric chemistry and climate science.

This study presents experimental atomic-scale imaging of the interaction between K-feldspar and water.[2] We cleaved the mineral in ultra-high vacuum and analyzed the surface using non-contact atomic force microscopy (AFM) and X-ray photoelectron spectroscopy. The surfaces are prone to hydroxylation, exposing arrays of silanol (Si-OH) and aluminol (Al-OH) groups. We studied the onset of ice nucleation by introducing H_2O vapor at low temperatures on such hydroxylated surfaces. Ab-initio calculations in tandem with AFM simulations employing the Probe Particle Model [3] helped us interpret our results.

[1] A. Kumar, et al., Atmos. Chem. Phys. 18, 7057 (2018)
[2] G. Franceschi, et al., submitted (2023)
[3] P. Hapala, et al., Phys. Rev. B 90, 085421 (2014)

O 49.8 Wed 12:15 MA 144

Unraveling the effects of substrate interaction on the chemical properties of atomic layer deposited ultra-thin ceria layers — •Carlos Morales, Yuliia Kosto, Rudi Tschammer, Karsten Henkel, and Jan Ingo Flege — Applied Physics and Semiconductor Spectroscopy, Brandenburg University of Technology Cottbus-Senftenberg, Konrad-Zuse-Strasse 1, D-03046 Cottbus, Germany

Atomic layer deposition (ALD) is well known to lead to amorphous and defective, non-stoichiometric films, potentially resulting in modified material properties that can also be affected by film/substrate interaction in the case of ultra-thin growths. For example, the formation, diffusion, and recovery of oxygen vacancies can be favored in disordered, reducible metal oxides compared to more ordered deposits, whereas interdiffusion processes can critically affect the film/substrate interface region. These effects have extensively been studied for thin thermal-ALD ceria films (below 15 nm) by combining in-situ and ex-situ characterization techniques in our lab and at synchrotron radiation facilities. While using alumina or silica substrates modifies the initial growth rate, Ce^{3+}/Ce^{4+} ratio, and ceria morphology, the formation of different species at the interface affects its reactivity. Interestingly, the experiments have shown high reducibility of ALD-ceria ultrathin films on silica for very low hydrogen concentrations, even at room temperature, whereas for alumina substrates the formation of aluminates at the interface prevents further oxidation. Moreover, the comparison with more ordered films indicates a key role of the defective structure of ALD films in Ce^{3+}/Ce^{4+} conversion.

O 49.9 Wed 12:30 MA 144

Dynamics of an $Fe_3O_4(001)$ support for Pt_n cluster catalysts under the fast STM — Sebastian Kaiser[1], Johanna Reich[2], Ueli Heiz[1], Barbara A.J. Lechner[2], and •Friedrich Esch[1] — [1]Chair of Physical Chemistry, TUM School of Natural Sciences and Catalysis Research Center, Technische Universität München, Lichtenbergstr. 4, 85748 Garching — [2]Functional Nanomaterials Group, TUM School of Natural Sciences and Catalysis Research Center, Technische Universität München, Lichtenbergstr. 4, 85748 Garching

Reducible oxides are valuable supports for cluster catalysts in oxidation reactions due to their capability to contribute lattice oxygen. Here, we present a study on the surface dynamics of Fe_3O_4 in the absence and in the presence of supported size-selected Pt_5 clusters [1, 2]. With our dedicated FAST add-on module [3], we drive our variable temperature scanning probe microscope at choice in a fast-imaging mode at movie rates of several images per second and in a particle-tracking mode that allows to follow diffusion paths with a time resolution down to 10 ms. This paves the way to unravel in situ a variety of surface dynamics at the atomic scale that remain hidden to static imaging: Hydrogen diffusion, subsurface Fe interstitial transport, hole growth upon catalytic surface reduction, and, finally, cluster diffusion in the Smoluchowski ripening regime, at the verge of encapsulation.

[1] S. Kaiser et al., ACS Catalysis 11, 9519 (2021).
[2] S. Kaiser et al., ACS Catalysis 13, 6203 (2023).
[3] C. Dri et al., Ultramicroscopy, 205, 49 (2019).

O 49.10 Wed 12:45 MA 144

Imaging surface structure and premelting of hexagonal ice with atomic resolution — •Jiani Hong, Ye Tian, Tiancheng Liang, Xinmeng Liu, Limei Xu, Enge Wang, and Ying Jiang — School of Physics, Peking University, Beijing, P. R. China

The ice surfaces are closely relevant to many physical and chemical properties of ice, such as melting, freezing, friction, gas uptake, and atmospheric reaction. Despite massive experimental and theoretical investigations, the exact atomic structure of the ice interface still remains elusive due to the vulnerable surface hydrogen-bonding network and the complicated premelting process. Here, we realize the first atomic-resolution imaging of basal surface structure of hexagonal water ice by using qPlus-based cryogenic atomic force microscopy with a CO-functionalized tip. We find that the crystalline ice-Ih surface is composed of mixed Ih- and Ic-stacking nanodomains, forming periodic superstructures. DFT reveals that such a reconstructed ice surface is stabilized over the ideal ice surface mainly by minimizing the electrostatic repulsion between dangling OH bonds at the surface. Moreover, we find that the ice surface gradually becomes disordered with increasing temperature in experiment, revealing the onset of the premelting process. The surface premelting occurs from the defective boundaries between the Ih and Ic domains and can be promoted by the formation of a planar local structure. Those results put an end to the long-standing debate on the ice surface structure and shed new light on the molecular origin of ice premelting, which may lead to a paradigm shift in the understanding of ice physics and chemistry.

O 50: Supported Nanoclusters: Structure, Reaction, Catalysis

Time: Wednesday 10:30–12:30 Location: TC 006

O 50.1 Wed 10:30 TC 006

Interaction of Formate with Magnetite Nanoparticles Supported by Al_2O_3(0001) — •Mohammad Ebrahim Haji Naghi Tehrani[1,2], Daniel Silvan Dolling[1,2], Jan-Christian Schober[1,2], Esko Erick Beck[1,2], Mona Kohantorabi[1], Arno Jeromin[1], Thomas F. Keller[1], Vedran Vonk[1], Heshmat Noei[1,3], and Andreas Stierle[1,2] — [1]Deutsches Elektronen-Synchrotron (DESY), D-22603 Hamburg, Germany — [2]Fachbereich Physik Universität Hamburg, Jungiusstrasse 9, 20355 Hamburg, Germany — [3]Hamburg Centre for Ultrafast Imaging, Universität Hamburg, Hamburg, Germany

We studied the growth and adsorption properties of magnetite nanoparticles (NPs) grown on Al_2O_3(0001) support by means of physical vapor deposition. Grazing incidence X-ray diffraction (GIXRD) characterization indicated the formation of epitaxial (111)-oriented NPs. Scanning electron microscope (SEM) showed triangular-shaped NPs. Adsorption of formic acid as a probe molecule was studied on the magnetite NPs using Fourier transmission infrared reflection-absorption spectroscopy (FTIRRAS). According to the results, formate adsorbs in the forms of chelating and quasi-bidentate geometries on these NPs, similar to what we recently evidenced on a (111) magnetite single crystal surface. Moreover, X-ray photoelectron spectroscopy (XPS) proved a possible phase transition from magnetite to maghemite after air exposure. Our model system can be applied in order to design the next generation of hierarchical organic-linked magnetite.

O 50.2 Wed 10:45 TC 006

Elucidating the Role of the Degree of Reduction in the Alcohol Photooxidation over Titania(110) Photocatalysts — •Philip Petzoldt, Lucia Mengel, Anna Lemperle, Martin Tschurl, and Ueli Heiz — Chair of Physical Chemistry, School of Natural Sciences & Catalysis Research Center, Technische Universität München, Lichtenbergstr. 4, 85748 Garching, Germany

Titania is one of the most widely investigated metal-oxides in surface science and often considered the archetypical heterogeneous photocatalyst. Fundamental studies have deepened the understanding of its thermal and photochemical reactivity on the atomic scale as well as the interplay with nanoparticulate co-catalysts. However, a critical but commonly neglected parameter of titania is the degree of its reduction and consequential impacts on the (photo)reactivity.

In this contribution, we address this aspect by investigating the alcohol photooxidation reaction over Pt cluster-loaded Titania(110) single crystals, while systematically changing the degree of reduction of the photocatalyst. Our results provide new insights into different factors governing the photoreactivity and underline the importance of kinetics in photocatalysis.

O 50.3 Wed 11:00 TC 006

Influence of the Strong Metal-Support Interaction on the Photoactivity of Pt-loaded TiO_2(110) — •Lucia Mengel, Philip Petzoldt, Martin Tschurl, and Ueli Heiz — Chair of Physical Chemistry, School of Natural Sciences & Catalysis Research Center, Technische Universität München, Lichtenbergstr. 4, 85748 Garching, Germany

Pt-loaded TiO_2(110) is well-known for its capability of alcohol photoreforming. The hole-mediated photooxidation reaction yields hydrogen next to valuable organic compounds such as formaldehyde. The concept of encapsulation of clusters or nanoparticles by a metal oxide overlayer under reductive conditions is well established and has been extensively studied using a variety of techniques. Such encapsulations caused by the strong metal-support interaction (SMSI) are a promising tool in photo-/electrocatalysis to improve catalyst selectivity and high-temperature stability. However, the mechanistic influence of an SMSI overlayer on the photocatalytic hydrogen evolution reaction on Pt-loaded TiO_2(110) is still under investigation.

In our work, we focus on SMSI-induced effects on the hydrogen evolution reaction on Pt_{10}-loaded TiO_2(110) under ultra-high vacuum conditions. In particular, we investigate the SMSI state of the Pt_{10} clusters by thermal-programmed desorption of probe molecules. We employ methanol photoreforming as model reaction to study the evolution of hydrogen on the Pt_{10} clusters in the SMSI state.

O 50.4 Wed 11:15 TC 006

Cu Oxide Nanoparticles for Virus Inactivation — •Daniel Silvan Dolling[1,2], Miguel Blanco Garcia[1,2], Jan-Christian Schober[1,2], Mohammad Ebrahim Haji Naghi Tehrani[1,2], Ming Chao Kao[1,2], Andreas Stierle[1,2], and Heshmat Noei[2] — [1]Deutsches Elektronen Synchrotron (DESY) — [2]Universität Hamburg

Copper and its oxides are well known for their antiviral and antibacterial properties, more recently including the inactivation of SARS-CoV-2 [1, 2, 3]. The combination of Cu oxides with TiO2 has attracted interest due to the photocatalytic activity of the combined system. For the photocatalytic activity, the specific oxidation state of Cu is paramount, as the oxidation states offer different pathways for visible light activity. Up to now, most research regarding virus inactivation has focused on powder systems. Here, we investigate the effects of different Cu nanoparticle sizes and coverages on single crystalline TiO2(110) surface by X-ray photoelectron spectroscopy(XPS). Moreover, as the oxide state is playing a major role in the (photo-)activity, we investigate the in-situ oxidation of Cu nanoparticles via XRD, XPS and SEM. [1] M. Hosseini et al., Scientific Reports 12 (2021), 5919-5928. [2] A. Purniawan et al., Scientific Reports 12 (2022). [3] M. Liu et al., J. Mater. Chem. A 3 (2015), 17312-17319.

O 50.5 Wed 11:30 TC 006

Structure of Pd/CeO2/YSZ and Pd/Al2O3 model systems during methane oxidation light-off — •Jan Schober[1,2], Mona Kohantorabi[1], Birger Holtermann[3], Nadejda Firman[3], Thomas Keller[1], Vedran Vonk[1], Yolita Eggeler[3], and Andreas Stierle[1,2] — [1]DESY, Hamburg, Germany — [2]UHH, Hamburg, Germany — [3]KIT (LEM), Karlsruhe, Germany

High ecological impact of gasoline and diesel fuels causes a shift to natural gas and biofuel alternatives. Methane is one of the major components of these fuels and has a much higher greenhouse gas effect. To eliminate methane from exhaust gases, heterogeneous catalysts using noble metal NPs of the Pt group with oxides such as Al2O3 and CeO2 as support. [1,2]. To track the active sites and elucidate structure function relationships on an atomistic level we prepare fully oxidized and epitaxial CeO2 thin films for NP support, suitable for catalytic studies using grazing incidence X-ray diffraction. This allows us to investigate open questions about the interplay of the NPs with the support, role of the support in the reaction, sintering behavior and reaction mechanism in the low-temperature regime [1]. Using our operando catalysis environments, we investigated Pd/Al2O3 and Pd/CeO2/YSZ catalysts during methane oxidation light-off experiments at ID31, ESRF with HEGIXRD. With in-line mass spectrometry we were able to track activity while capturing detailed structural information on NPs and support by continuously tracking high-symmetry planes of reciprocal space. [1] DOI: 10.1021/acscatal.0c03338. [2] DOI: 10.1016/j.susc.2013.06.014.

O 50.6 Wed 11:45 TC 006

Bimetallic Pd-Rh core-shell nanoparticles supported on Co3O4(111): atomic ordering and stability — •Yaroslava Lykhach[1], Alexander Simanenko[1], Lukáš Fusek[1,2], Tomáš Skála[2], Nataliya Tsud[2], Sascha Mehl[3], Olaf Brummel[1], and Jörg Libuda[1] — [1]Friedrich-Alexander-Universität Erlangen-Nürnberg, Erlangen, Germany — [2]Charles University, Prague, Czech Republic — [3]Elettra-Sincrotrone Trieste SCpA, Basovizza-Trieste, Italy

The activity of bimetallic Pd-Rh catalysts can be efficiently tuned by geometric and electronic effects resulting from the atomic arrangement and bimetallic interactions between the Pd and Rh atoms. Additional functionalities arising from the electronic metal-support interaction (EMSI) can be introduced by the use of reducible oxide supports. We investigated the impact of the EMSI on the atomic ordering and stability of bimetallic Pd@Rh and Rh@Pd core@shell nanostructures supported on well-ordered Co3O4(111) films by means of synchrotron radiation photoelectron spectroscopy (SRPES). Depth profile analysis of the charge transfer and the oxidation states of Rh and Pd was performed using tunable synchrotron light during the preparation and annealing of the model systems in UHV. We found that the EMSI yields a fraction of Rh3+ species at the interface between metallic Rh nanoparticles and Co3O4(111). Annealing the model systems triggers atomic rearrangement in the core@shell nanostructures, leading to the formation of thermodynamically favorable Pd-rich shells and Rh-rich cores in both core@shell systems.

O 50.7 Wed 12:00 TC 006

Identifying Partial Reaction Steps in the Photocatalytic Conversion of Alcohols on Rutile Titania by Isotopic Labeling — •Martin Tschurl, Philip Petzoldt, and Ueli Heiz — Chair of Physical Chemistry, School of Natural Sciences & Catalysis Research Center, Technische Universität München, Lichtenbergstr. 4, 85748 Garching, Germany

The substitution of particular isotopes is a powerful method to uncover partial reactions in complex networks and to identify rate-determining steps by ascertaining kinetic isotope effects. Utilizing this methodology, we study the reaction of methanol on a rutile $TiO_2(110)$ single crystal decorated with Pt_{10} clusters under UV illumination in an ultra-high vacuum environment. By applying alcohols with different degrees of deuteration, we exemplarily reveal the reaction sequence in the photocatalysis of co-catalyst-loaded rutile titania for this class of molecules.

O 50.8 Wed 12:15 TC 006

Photocatalytic Reforming of Tertiary Alcohols at Ambient Conditions — •Paula Neumann, Clara Aletsee, Martin Tschurl, and Ueli Heiz — Chair of Physical Chemistry, School of Natural Sciences & Catalysis Research Center, Technische Universität München, Lichtenbergstr. 4, 85748 Garching, Germany

The selective oxidation of tertiary alcohols by heterogeneous photocatalysis is a mild alternative compared to harsh methods commonly employed in organic chemistry. What was first shown in a UHV study using a titania single crystal as a photocatalyst, we transferred to ambient conditions by means of our gas phase micro-reactor enabling sensitive and time-resolved product detection in a well-defined environment under the exclusion of oxygen. The results observed for a titania powder catalyst are in good agreement with UHV findings suggesting the absence of a significant pressure gap wich allows for a comprehensive interpretation of the surface photochemistry. Lastly, we discuss the impact of co-catalyst loading on the reaction and present the underlying mechanistic processes.

O 51: Focus Session: Molecular Nanostructures on Surfaces: On-Surface Synthesis and Single-Molecule Manipulation II

Time: Wednesday 15:00–17:45 Location: HE 101

Topical Talk O 51.1 Wed 15:00 HE 101

Exploring the Magnetic and Topological Properties in Carbon-based Nanomaterials — •Ping Yu — School of Physical Science and Technology, ShanghaiTech University, 201210 Shanghai, China

In low-dimensional carbon-based nanomaterials, quantum magnetism can be generated from the pi electrons, which have attracted lots of attention in the fields of on-surface synthesis and quantum spintronic materials. The talk will report on how to realize graphene-based quantum materials with desired magnetic or topological properties through on-surface synthesis. The electronic properties of designed graphene nanomaterials are comprehensively investigated by scanning probe microscope techniques and theoretical calculations. The results provide new strategies for designing and synthesizing nanographene materials with desired electronic functionalities.

O 51.2 Wed 15:30 HE 101

Polyradical State of an Extended Aza-Triangulene — •Francisco Romero-Lara[1], Alessio Vegliante[1], Manuel Vilas-Varela[2], Hector Briongos[1], Niklas Friedrich[1], Dongfei Wang[1], Ricardo Ortiz[3], Patrick Calupitan[3], Fabian Schulz[1], Thomas Frederiksen[3], Diego Peña[2], and Jose Ignacio Pascual[1] — [1]CIC nanoGUNE, Donostia, Spain — [2]CiQUS, Santiago de Compostela, Spain — [3]DIPC, Donostia, Spain

Triangulene nanographenes are the most paradigmatic case for the emergence of spin states due to the frustration of the bipartite lattice of graphene. Increasing their size implies an increase in sublattice imbalance, resulting in a larger spin state according to Lieb's theorem. Another way of controlling the spin state is by heteroatom substitution, e.g. N reduces the spin state by 1/2. In this work, we show that, by increasing the size of an aza-[3]-triangulene but maintaining the sublattice imbalance, the interplay between electron correlations and wavefunction hybridization is modified, resulting in a polyradical state. The successful synthesis of this extended triangulene on a Au(111) surface is confirmed by STM and nc-AFM with CO tips. We use STS to prove the magnetic character of the nanographene by measurements of the Kondo effect and spin excitations. Temperature and magnetic field dependence of the Kondo resonance indicates an $S=1/2$ ground state. Simultaneously, a spin excitation to a $S=3/2$ excited state was observed. These findings support the presence of a polyradical state in this nanographene conformed by three antiferromagnetically aligned radicals, in agreement with multireference CAS theoretical methods.

O 51.3 Wed 15:45 HE 101

Tuning the spin coupling in all-organic diradicals through mechanical manipulation — •Alessio Vegliante[1], Saleta Fernandez[2], Manuel Vilas-Varela[2], Ricardo Ortiz[3], Thomas Baum[4], Niklas Friedrich[1], Francisco R. Lara[1], Herre van der Zant[4], Thomas Frederiksen[3], Diego Peña[2], and Jose Ignacio Pascual[1] — [1]CIC NanoGUNE, Spain — [2]CiQUS-USC, Spain — [3]DIPC, Spain — [4]TU Delft, the Netherlands

Open-shell organic molecules have emerged as promising candidates for carbon-based spintronics. Organic diradicals, in particular, are interesting model systems for studying and manipulating intramolecular spin interactions at the atomic scale.

Here we report the tunable spin interactions of the molecular diradical 2OS on a Au(111) substrate using scanning tunneling microscopy and spectroscopy. 2OS is stable diradical, derivative of the Chichibabin*s hydrocarbon, characterized by a non-planar and flexible structure. With the support of theoretical calculations, we show that the spin interaction strongly depends on the structural conformation of the molecule: the adsorption on the Au surface induces a partial planarization that stabilizes a singlet ground state, while less planar conformations exhibit a significantly reduced spin coupling.

In this work, we demonstrate the possibility of tailoring the spin interaction through structural changes induced by approaching the STM tip or lifting the molecule from the substrate, thus confirming the influence of geometry on the molecule*s spin state.

O 51.4 Wed 16:00 HE 101

Charge states and electron correlation in graphene nanoribbons on MgO — •Leonard Edens[1], Amelia Domínguez Celorrio[2], Manuel Vilas-Varela[3], Sofia Sanz[4], Thomas Frederikssen[4], Diego Peña[3], Nacho Pascual[1], and David Serrate[2] — [1]CIC nanoGUNE, Spain — [2]CSIC-INMA and Universidad de Zaragoza, Spain — [3]CiQUS and Universidade de Santiago de Compostela, Spain — [4]DIPC, Spain

On-surface synthesis of graphene nanostructures provides tailor-made systems exhibiting quantum properties such as π magnetism. However, the underlying catalyst metal readily hybridizes with molecular orbitals and quenches spin. Here, we report the successful lateral STM manipulation of individual graphene nanoribbons onto epitaxial layers of MgO(001) after synthesis on Ag(001). We observe a rich electronic spectrum of large absolute gaps straddled by remarkably sharp resonances implying drastic lifetime enhancement. LDOS mapping allows exact determination of the large integer negative charge acquired due to the low insulator work function. We find that the integer charge state of the molecule depends on its length, leading to an opening and closing of the correlation gap with charge parity. Studying charge as a function of length shows that the electronic state alternates between open- and closed-shell as determined by the interplay between level evolution with length and electron correlation. We introduce a mean-field Hubbard model in the grand canonical ensemble that reproduces and predicts the charging behaviour and allows us to extract the energy of the graphene-metal interface dipole across the insulator.

O 51.5 Wed 16:15 HE 101

On-surface synthesis of non-planar carbon nanoribbons — Feifei Xiang[1], Sven Maisel[2], •Shreya Garg[1], Andreas Görling[2], and Sabine Maier[1] — [1]Department of Physics, Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany — [2]Department of Chemistry and Pharmacy, Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany

Graphene nanoribbons have great potential for applications in nanoelectronic devices such as transistors by combining the outstanding electronic properties of graphene with the introduction of an electronic band gap. In this study, we present a bottom-up synthesis approach for covalently linked non-planar carbon nanoribbons. We discuss the electronic properties of the non-planar carbon ribbons in relation to their adsorption geometry using low-temperature scanning tunneling microscopy and spectroscopy in combination with density functional theory. The synthesis involves bowl-shaped dibrominated indacenopicene precursors to create non-planar carbon ribbons through surface-assisted Ullmann-type dehalogenative coupling and cyclodehydrogenation on Au(111).

O 51.6 Wed 16:30 HE 101

Unipolar Resonant Hole Transport Through a Free-Standing Designer Graphene Nanoribbon — •Niklas Friedrich[1], Jingcheng Li[1], Iago Pozo[2], Diego Peña[2], and Nacho Pascual[1] — [1]CIC nanoGune, Spain — [2]CiQUS, Spain

Coherent electron transport through individual molecules is a vital technique to probe different quantum mechanical properties like energy level alignment, vibronic modes or spin states. Here, we investigate the electronic transport through 7-armchair graphene nanoribbons (GNRs) containing a single, substitutionally embedded 2B-dimer.

We find that the coherent electron transport through the 2B-GNR is unipolar with a singly occupied 2B-state (O2B) enabling resonant electron tunneling at both voltage polarities. The unipolar transport is favored by an exponential localization of the O2B resulting in a double tunneling barrier configuration. Further transport resonances reveal that resonant transport through the valence band exhibits a unipolar character, too. We find fingerprints of vibronic satellites and of band quantization in form of Fabry-Perot quantum well modes in our experiments.

The experiments were performed by lifting single 2B-GNRs using the tip of a low-temperature scanning tunneling microscope (STM) to create the molecular wires that bridges tip and substrate. Our results unravel the details of coherent resonant electron tunneling through molecular wires built from single 2B-doped GNRs, confirming their technological potential for single molecule electronics.

O 51.7 Wed 16:45 HE 101

On-Surface Synthesis of Edge-Extended Zigzag Graphene Nanoribbons — •Amogh Kinikar[1], Feifei Xiang[1], Xiushang Xu[2], Yanwei Gu[3], Akimitsu Narita[2], Klaus Müllen[2], Carlo Pignedoli[1], Oliver Gröning[1], Pascal Ruffieux[1], and Roman Fasel[1] — [1]Empa, Dübendorf 8600 Switzerland — [2]OIST Graduate University, Okinawa 904-0495, Japan — [3]MPI-P, 55128 Mainz, Germany

Graphene nanoribbons (GNRs) have gained significant attention in nanoelectronics due to their potential for precise tuning of electronic properties through variations in edge structure and ribbon width. However, the synthesis of GNRs with highly sought-after zigzag edges (ZGNRs) remains challenging. Here, we present a design motif for synthesizing novel edge-extended ZGNRs. This motif enables the controlled incorporation of edge extensions along the zigzag edges at regular intervals, opening up possibilities for synthesizing a diverse range of edge-extended ZGNRs. Examples of successfully synthesized structures are presented, characterized using Scanning Tunneling Microscopy, and complemented by Density Functional Theory calculations. These efforts elucidate the electronic and magnetic properties of these edge-extended ZGNRs. The diverse range of edge-extended ZGNRs now possible expands the structural landscape of GNRs and facilitates the exploration of their structure-dependent electronic properties.

[1] Kinikar, A. et al. "On-surface Synthesis of Edge-Extended Zigzag Graphene Nanoribbons." *Advanced Materials* (2023) **35**, 2306311.

O 51.8 Wed 17:00 HE 101

How conductive is a single polyene chain? — •Sifan You[1], Yixuan Gao[2], Shixuan Du[2], and Lifeng Chi[1] — [1]Soochow University, Suzhou, China. — [2]Institute of Physics, Beijing, China

Conjugated polymers are promising candidates for molecular wires in nanoelectronics, with flexibility in mechanics, stability in chemistry and variety in electrical conductivity. Polyene, as a segment of polyacetylene, is a typical conjugated polymer with straightforward structure and wide-range adjustable conductance. To obtain atomic scale understanding of charge transfer in polyene, we measured the conductance of a single polyene-based molecular chain via lifting it up with scanning tunneling microscopy tip. Different from semiconducting characters in pristine polyene (polyacetylene), high conductance and low decay constant were obtained, along with an electronic state around Fermi level and characteristic vibrational mode.

O 51.9 Wed 17:15 HE 101

A Challenge Human vs Machine: Building a Nanopattern of Unknown Adsorbates — •Bernhard Ramsauer[1], Grant Simpson[1], Johannes J. Cartus[1], Leonhard Grill[2], and Oliver T. Hofmann[2] — [1]Institute of Solid State Physics, NAWI Graz, Graz University of Technology, Graz, 8010, Austria — [2]Department of Physical Chemistry, Institute of Chemistry, NAWI Graz, University Graz, Graz, 8010, Austria

Scanning probe microscopes give us the possibility to precisely control the position and orientation of single molecules, thus unlocking the possibility to fabricate quantum-structures with novel properties. However, interaction processes at the nanoscale are stochastic, and the motion of molecules is often unintuitive and hard to predict even for human experts.

In this work we present an artificial intelligence challenging a human expert to build a pattern of unknown molecules as fast as possible. The challengers had one week each to learn the optimal manipulation parameters and to build the pre-defined nanopattern. In the building process every type of manipulation is allowed. Within this week one can attempt building the nanopattern as often as possible but the winner is determined by the shortest assembly time.

O 51.10 Wed 17:30 HE 101

Friction over single chemical bonds — Oliver Gretz[1], Lukas Hörmann[2,3], •Shinjae Nam[1], Oliver T. Hofmann[2], Franz J. Giessibl[1], and Alfred J. Weymouth[1] — [1]Universität Regensburg, Deutschland — [2]TU Graz, Österreich — [3]University of Warwick, UK

Friction is a phenomenon that acts on many different length scales. With a sharp nanometer-sized tip, lateral forces can be measured and phenomena like anisotropy or superlubricity can be observed [1, 2]. However, the *large* tip prevents measurements of single chemical bonds. But is this spatial resolution necessary to understand sliding friction? How much does the energy loss differ when sliding over different chemical bonds? We performed lateral force microscopy, in which the tip oscillates laterally above the surface, with small amplitudes and a CO-terminated tip to be directly sensitive to the dissipated energy above single chemical bonds [3]. By comparing the dissipation over different covalent bonds, we found that the local potential energy landscape plays an important role in sliding friction. We were also able to observe dissipation over single O**H bonds. Our findings show that a complete understanding of friction requires a description of the individual chemical bonds at the surface.

1.*Liley, M. et al., Science, 280(5361), 273-275, (1998). 2.*Dienwiebel, M. et al., Physical review letters, 92(12), 126101, (2004). 3.*Weymouth, A. J. et al., Physical Review Letters, 124(19), 196101, (2020).

O 52: Focus Session: Spins on Surfaces studied by Atomic Scale Spectroscopies V

Time: Wednesday 15:00–17:15 Location: MA 004

Topical Talk O 52.1 Wed 15:00 MA 004

Locally driven quantum phase transitions in a strongly correlated molecular monolayer — •Markus Ternes — Institute of Physics IIB, RWTH Aachen University, 52074 Aachen, Germany — 2 Peter-Grünberg-Institute (PGI 3), Research Center Jülich, 52425 Jülich, Germany — Jülich Aachen Research Alliance, 52425 Jülich, Germany

The Kondo effect, which appears as a strong zero-bias anomaly, is prototypical for strongly correlated states and therefore cannot be described by single-particle models. Its subtle interplay with magnetically ordered ground states in multi-site Kondo systems continues to attract interest. Here we report on a molecular system of NTCDA molecules on Ag(111) in which individual molecules do not show a Kondo effect even at temperatures of 1K because the π symmetry of their singly occupied orbital hybridizes only weakly with the electrons of the host Ag. In a perfectly ordered lattice, however, newly formed orbital superpositions dramatically increase the hybridization, so that the NTCDA molecules now form a Kondo lattice. Using the electric field exerted by the tip of an STM, we manipulate the orbital superpositions and drive the system through a cascade of quantum phase transitions in which the molecular building blocks change one by one from a Kondo screened to a new paramagnetic ground state, allowing us to reconstruct their complex interactions in detail.

O 52.2 Wed 15:30 MA 004

Evidence for spinarons in Co atoms on noble metal (111) surfaces — •Artem Odobesko, Felix Friedrich, and Matthias Bode — Julius-Maximilians-Universität Würzburg, Physikalisches Institut, Experimentelle Physik II, Am Hubland, 97074 Würzburg, Germany

The first Kondo effect's detection on individual Co atoms on Au(111) surface [1] was associated with a zero-bias anomaly (ZBA) in the differential conductance signal, explained by Fano resonance due to interfering tunneling paths into the Kondo state and atomic orbitals. Recent calculations [2] challenge this interpretation, suggesting that the ZBA is a mix of Co atom spin excitations and the spinaron, a magnetic polaron resulting from the interaction of spin excitations with conduction electrons. Our study used spin-polarised STS on Co atoms on Cu and Au (111) surfaces in magnetic fields up to 12 T. Our comprehensive study of the responses of the ZBA on Co atoms on Cu(111) to an external magnetic field, in conjunction with spin-polarized measurements, allowed us to discriminate between various theoretical models. We observe a field-induced energy shift and experimentally determine the spin character of the spectral features, exhibiting a behavior contrary to Kondo expectations but in line with the spinaron. Consequently, we invalidate the prevailing Kondo-based interpretation of the ZBA in favor of the spinaron and, for the first time experimentally, detect this novel many-body excitation [3].

[1] V. Madhavan *et al.*, Science 280, 567 (1998)
[2] J. Bouazizet *al.*, Nat. Comm. 11, 6112 (2020)
[3] F. Friedrich, *et al.*, Nat. Phys. (2023)

O 52.3 Wed 15:45 MA 004

Spin polarization of the Kondo system in mirror twin boundaries of MoS_2 — •Mahasweta Bagchi[1], Tfyeche Tounsi[1], Affan Safeer[1], Camiel van Efferen[1], Thomas Michely[1], Wouter Jolie[1], Theo A. Costi[2], and Jeison Fischer[1] — [1]II. Physikalisches Institut, Universität zu Köln, Zülpicher Str. 77, 50937 Cologne, Germany — [2]Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich, 52425 Jülich, Germany

We report spin-polarized scanning tunneling microscopy measurements of the Kondo effect observed in mirror twin boundaries of MoS_2 on graphene, which are consistent with numerical renormalization group calculations. A Kondo resonance appears because the magnetic moment of a singly occupied quantum confined state of the mirror twin boundary is screened by the conduction electrons from the substrate [1]. Using a spin-polarized tip, we measure the Kondo resonance and the singly and doubly occupied confined levels simultaneously, characterizing the full Anderson system. Clear changes in the peak heights of the confined states as well as the magnetic field-split Kondo state provide evidence of their full spin polarization. The magnetization of the confined level as a function of magnetic field and temperature can be described as single quantum spin. This, along with the absence of any higher spin excitations, establishes the mirror twin boundary of MoS_2 as an ideal spin 1/2 system.

[1] van Efferen, et al., Modulated Kondo screening along magnetic mirror twin boundaries in monolayer MoS_2. Nat. Phys. (2023).

O 52.4 Wed 16:00 MA 004

Temperature evolution of the Kondo peak beyond Fermi liquid theory — •David Jacob — University of the Basque Country UPV/EHU, San Sebastian, Spain — IKERBASQUE, Basque Foundation for Science, Bilbao, Spain

The limitation of Fermi liquid theory to very low energies and temperatures poses a fundamental problem for describing the temperature evolution of the Kondo peak. Here Fermi liquid theory for the single impurity Anderson model is extended beyond the low-energy and low-temperature regime by means of an Ansatz for the impurity self-energy based on the accurate description of the Kondo peak by the Frota function and by exploiting Fermi liquid conditions. Analytic expressions for the temperature dependence of the Kondo peak height and width derived from this Ansatz are in excellent agreement with numerical renormalization group data for temperatures up to and beyond the Kondo temperature. The derived expression thus allows to unambiguously determine the intrinsic Kondo peak width and Kondo temperature from finite temperature measurements of the Kondo resonance, as measured by scanning tunneling spectroscopy of magnetic adatoms and molecules on conducting surfaces.

References: D. Jacob, Phys. Rev. B **108**, L161109 (2023); E. Turco *et al.*, arXiv:2310.09326 (2023)

O 52.5 Wed 16:15 MA 004

Accurate Kondo temperature determination of spin-1/2 magnetic impurities — Elia Turco[1], Markus Aapro[2], Somesh C. Ganguli[2], •Nils Krane[1], Robert Drost[2], Nahual Sobrino[3], Annika Bernhardt[4], Michal Juríček[4], Roman Fasel[1,5], Pascal Ruffieux[1], Peter Liljeroth[2], and David Jacob[3,6] — [1]Empa, Dübendorf, Switzerland — [2]Aalto University, Aalto, Finland — [3]Universidad del País Vasco UPV/EHU, San Sebastián, Spain — [4]University of Zurich, Zurich, Switzerland — [5]University of Bern, Bern, Switzerland — [6]IKERBASQUE, Bilbao, Spain

A localized spin interacting with the electron bath of a metallic surface might give rise to the Kondo effect. The energy scale of this interaction is related to the Kondo temperature T_K and an important quantity when it comes to application of nanoscale magnets. In scanning tunneling spectroscopy (STS) the Kondo effect is observed as a resonance at zero bias. The characteristic temperature evolution of this resonance can be used to prove the Kondo nature of the zero-bias resonance, but only when all other significant broadening methods have been taken properly into account. Using Phenalenyl on Au(111) as an ideal spin-1/2 Kondo system, we demonstrate that the lineshape of a Kondo resonance, measured by STS at finite temperatures, can be described very well by a Hurwitz ζ-function. The extracted intrinsic Kondo linewidth fit very well with a recently derived expression for the temperature evolution of the Kondo resonance. Utilizing the new methodology, we are able to extract the Kondo temperature T_K of a system reliably by a single spectra taken at finite temperature.

O 52.6 Wed 16:30 MA 004

Stable π radical BDPA on Cu(100): adsorption and Kondo signature — •Jacob Teeter[1], Daniel Miller[2], and Stefan Müllegger[1] — [1]Solid State Physics Department, Johannes Kepler University Linz, 4040 Linz, Austria — [2]Hofstra University, New York, USA

Introduction

Stable organic radicals can serve as model systems for investigating metal-free magnetic phenomena. We have investigated one such species, α, γ-bisdiphenylene-β-phenylallyl (BDPA), at the single-molecule level on Cu(100).

Methods

Ultra-high vacuum (UHV) scanning tunneling microscopy (STM) and spectroscopy (STS) were performed using a commercial Omicron Polar STM at 6 K. Spectroscopic measurements were acquired with a W tip using an external lock-in amplifier and a typical modulation voltage of 2.5 mV. The substrate employed was a Cu(100) crystal obtained from Surface Preparation Laboratory. Density functional theory computations which account for London dispersion forces were performed to elucidate upon the adsorption structure and orientation of BDPA on the Cu(100) surface.

Results and Discussion

Our investigations revealed structural and electronic features of BDPA on Cu(100), including a preferential adsorption orientation and the presence of a Kondo-like feature in differential conductance measurements that suggests survival of the unpaired electron spin.

O 52.7 Wed 16:45 MA 004

Spin excitations and correlations in nanographene-based multi-spin platforms — •Elia Turco[1], Fupeng Wu[2], Nils Krane[1], Ji Ma[2], Roman Fasel[1], Xinliang Feng[2], and Pascal Ruffieux[1] — [1]EMPA, Duebendorf, Switzerland — [2]Faculty of Chemistry, Technical University of Dresden, Dresden, Germany

Chemical design offers the unique opportunity to realize robust molecular spin qubits with tailored magnetic properties, strong qubit interactions, and with practical bottom-up scalability. In this context, much attention has recently been given to open-shell nanographenes (NGs), whose spin interactions can be engineered with atomic precision by on-surface synthesis[1]. In particular, zigzag-edged triangular NGs (triangulenes) are regarded as prototypical magnetic building blocks, with a total spin S scaling with molecular size.

In this contribution, I will discuss multi-spin platforms fabricated via on-surface synthesis from the two smallest $S = 1/2$ and $S = 1$ triangulenes[2]. Scanning probe microscopy & spectroscopy of antiferromagnetically coupled hetero-dimers and trimers allow a thorough characterization of these coupled quantum spin systems, with their multiple inelastic spin excitations unambiguously reflecting the underlying spin Hamiltonians. The degenerate spin ground state in asymmetrically coupled spin systems, where each spin unit has a different Kondo exchange with the substrate, also allows to gain novel insights into Kondo correlations. [1] De Oteyza D.G. et al., J. Phys.: Condens. Matter 34 (2022); [2] Turco E. et al., JACS Au, 3, (2023)

O 52.8 Wed 17:00 MA 004

Strong exchange interactions between open-shell nanographenes and a rare earth-gold surface alloy — •Nicolo' Bassi[1], Feifei Xiang[1], Nils Krane[1], Carlo Pignedoli[1], Jan Jan Wilhelm[2], Michal Juríček[3], Roman Fasel[1], and Pascal Ruffieux[1] — [1]Empa, Duebendorf, Switzerland — [2]Institute of Theoretical Physics, Regensburg, Germany — [3]University of Zurich, Zurich, Switzerland

Rare-earth-based intermetallic compounds belong to a family of novel substrates, which is becoming a promising platform to control properties of nanomaterials via specific surface-adsorbates interactions. Different combinations, including GdAu2(1) and TbAu2(2), have been so far studied. They are all characterized by a ordered hexagonal superstructure with similar lattice constants. Here, we investigate various open-shell nanographenes on TbAu2 alloy by means of scanning tunneling techniques. For on-surface synthesized 7 armchair graphene nanoribbons (7-AGNRs), we find that the predicted spin properties of the end states are indeed conserved on TbAu2. Thanks to its lower workfunction, the 7-AGNRs are uncharged and the occupied and unoccupied end states have a spin polarization-induced energy splitting of 1.4eV. In addition, we investigated phenalenyl, the smallest open-shell molecule with spin $S = 1/2(4)$. Low bias spectroscopy reveals a splitting of more than 20mV, which we assign to the exchange interaction between the molecular spin and Tb atoms of the surface layer. These results shows a new substrate for studying different open-shell C structures. 1 Corso, M. et al. ACS Nano 4,(2010). 2 Que, Y. et al. J. Phys. Chem. Lett. 11,(2020). 3 Turco, E. et al. jJACS Au (2023)

O 53: 2D Materials IV: Graphene (joint session O/TT)

Time: Wednesday 15:00–18:00 Location: MA 005

O 53.1 Wed 15:00 MA 005

Ultra-large polymer-free suspended graphene films — •Lukas Kalkhoff, Sebastian Matschy, Ann-Sophie Meyer, Leon Lasnig, Nele Junker, Lars Breuer, and Marika Schleberger — Universität Duisburg-Essen, Fakultät für Physik, Germany

The quest to preserve graphene's unique characteristics has intensified the demand for advanced preparation methods. The production of large area graphene films has been constrained by the necessity of a polymer film, like PMMA, during the transfer process, leading to unavoidable contaminations. In this talk, we show our approach to fabricate triple-layer graphene films, without the use of polymers, freely suspended across circular holes with diameters of 100-300 micrometers. This technique achieves a high yield and also ensures remarkable uniformity of the electronic properties of graphene, both across and within individual holes [1].

Using confocal Raman and THz spectroscopy we can confirm that the structural and electronic properties of these triple-layer films are akin to monolayer graphene. This discovery has significant implications, particularly in their application as ion-electron converters in time-of-flight mass spectrometry and similar fields. Remarkably, these graphene films are two orders of magnitude thinner than conventional carbon foils used in such experiments, yet they maintain robustness and a high electron yield, as proven under picosecond ion bombardment. This marks a critical step toward replacing existing carbon films or polymer-based graphene with cleaner, more precisely defined graphene films. [1] L. Kalkhoff *et al.*, 2023, arXiv:2311.08137

O 53.2 Wed 15:15 MA 005

Design of electronic structure and transport in S-doped few-layer graphene — •Armin Sahinovic[1], Paolo Fortugno[2], Nicholas Wilson[3], Hartmut Wiggers[2], and Rossitza Pentcheva[1] — [1]Department of Physics, Universität Duisburg-Essen — [2]Institute for Energy and Materials Processes - Reactive Fluids, Universität Duisburg-Essen — [3]Department of Chemical Engineering, University of Waterloo

While the properties of N- and S-doped graphene have been addressed previously [1], the interplay between defects, doping and the layered structure of FLG are largely unknown. Based on density functional theory calculations we explore the formation energies and electronic structure of different defect types as a function of concentration and distribution. We find a layer dependence of the doping allowing for a design of the electronic structure. The surface layers are favored for doping, giving rise to buckling and a modified band structure such as a band gap opening not found for doping the center layers. Experiments on the synthesis of S-doped FLG in a microwave plasma reactor show S incorporation and a conductivity increase of up to 50%. This is consistent with the transport properties, obtained via BoltZtrap2 [2], revealing that S doping enhances the conductivity in FLG. We hereby show that tailoring the S incorporation into FLG enables one to design the electronic structure in view of energy conversion and storage applications.

[1] J.H Lee et al., Nanomaterials, 9, 268 (2019)

[2] G.K.H. Madsen et al., Comput. Phys. Commun., 231, 140 (2018)

Topical Talk O 53.3 Wed 15:30 MA 005

Tuning quantum electronic transport in nanoporous graphene — •Aran Garcia-Lekue — Donostia International Physics Center (DIPC), San Sebastian, Spain — Ikerbasque, Basque Foundation for Science, Bilbao, Spain

Recent experimental advances have demonstrated that graphene nanoribbons (GNRs) can be laterally coupled with atomic precision to obtain a nanoporous graphene (NPG) structure with highly anisotropic electronic properties.[1] Moreover, simulations have shown that the lateral coupling of GNRs leads to sizable interribbon transmission giving rise to intriguing interference patterns,[2] and that such interribbon transmission can be switched on/off by the chemical modification of the coupling bridges.[3]

Using density functional theory (DFT) and a multiscale transport method based on DFT combined with nonequilibrium Green's functions (NEGF), we have carried out electronic structure and current simulations for different NPG systems. First, in collaboration with our experimental colleagues, we have explored a new NPG that, provided by specifically designed coupling bridges, shows additional degrees of freedom to control the in-plane current.[4] Besides, we have investigated a bilayer composed of NPG and graphene, concluding that the interlayer current can be tuned by changing their relative twist angle.

[1] Moreno et al., Science 360, 199 (2018) [2] Calogero et al., ; Nano Lett. 19, 576 (2019) [3] Calogero et al., JACS 141, 13081 (2019) [4] Moreno et al., JACS 145, 8988 (2023)

O 53.4 Wed 16:00 MA 005

Visualizing band hybridization and moiré effects in gate-tunable twisted graphene layers using nanoARPES — •Zhihao Jiang[1], Kimberly Hsieh[1], Paulina Majchrzak[1], Alfred Jones[1], Chakradhar Sahoo[1], Youngju Park[2], Dongkyu Lee[2], Kenji Watanabe[3], Takashi Taniguchi[3], Jill Miwa[1], Jeil Jung[2], Yong P. Chen[1], and Søren Ulstrup[1] — [1]Aarhus University, Denmark — [2]University of Seoul, Korea — [3]National Institute for Materials Science, Japan

Twisted graphene layers have emerged as an intriguing class of quantum materials that display surprising correlation effects, including superconductivity, Mott insulators as well as strange metal phases. The possibility to tune these states using twist angle and electrostatic doping provides a promising route to interrogate the underpinning interactions between the electronic states. Here, we use angle-resolved photoemission spectroscopy with spatial resolution at the new micro- and nanoARPES branch at the ASTRID2 synchrotron light source at Aarhus University in Denmark to visualize the electronic states of twisted graphene layers integrated in device architectures. Specifically, we reveal the flat bands in twisted bilayer and double-bilayer graphene around the magic angles and systematically track the evolution of hybridization effects and moiré bands with small twist angles up to 6°. The interactions in the systems are further tuned by in situ electrostatic doping using a back-gate electrode. Our study paves the way for directly engineering band structure and correlation effects in twisted two-dimensional materials.

O 53.5 Wed 16:15 MA 005

Direct Mn implantation into graphene on Cu(111) substrate: understanding defect production in 2D materials from first-principles calculations — •Silvan Kretschmer[1], Renan Villarreal[2], Lino M. C. Pereira[2], and Arkady V. Krasheninnikov[1] — [1]Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf, Dresden 01328, Germany — [2]Quantum Solid State Physics, KU Leuven, 3001 Leuven, Belgium

Defects influence the properties of 2D materials tremendously, as they essentially consist of surface-only. Contrary to what their name implies, defects not only have detrimental effects, but also are introduced to tune the magnetic, electronic and optical response of 2D materials. The defect induced changes in the material and the concentration of defects produced, e.g by ion irradiation can be rationalized using first-principles calculations [1,2]. Here, we report on our recent simulation results obtained in collaboration with an experimental group on the probability to dope graphene with Mn atoms. Specifically, using ab-initio molecular dynamics we calculated the formation probability of Mn substitutional impurities in graphene on Cu(111) substrate under low-energy ion irradiation.

[1] S. Kretschmer, et.al, ACS Appl. Mater. Interfaces 10 (36), 30827–30836 (2018)

[2] S. Kretschmer, et. al, J. Phys. Chem. Lett. 13, 514–519 (2022)

O 53.6 Wed 16:30 MA 005

Investigating swift heavy ion induced defects in graphene and MoS$_2$ on SiO$_2$/Si via Raman spectroscopy — •Kevin Vomschee[1], Yossarian Liebsch[1], Leon Lasnig[1], Osamah Kharsah[1], Lars Breuer[1], Henning Lebius[2], Abdenacer Benyagoub[2], Clara Grygiel[2], and Marika Schleberger[1] — [1]Faculty of Physics and CENIDE, University of Duisburg-Essen, Duisburg, Germany — [2]CIMAP/GANIL, Caen, France

In the past years, ions have been used as a tool for the so-called defect engineering of 2D materials such as graphene and single-layer molybdenum disulfide (MoS$_2$). The potential impact of swift heavy ion irradiations on strain or doping in 2D materials and, in the case of graphene, also on defect formation can be effectively characterized using Raman spectroscopy. The irradiation of graphene on SiO$_2$/Si substrates with 91 MeV ^{129}Xe^{23+} and 967 MeV ^{129}Xe^{43+} at GANIL creates a so called D-peak indicating the presence of defects. The area ratio of the D-peak to the G-peak, another Raman peak that is not defect dependent, is recorded for accumulated ion fluences of up to $2.5\cdot10^{13}$cm^{-2}. Our Raman spectroscopy measurements show that swift heavy ion-induced defects in graphene are smaller in size than those investigated in earlier experiments by highly charged, slow Xe ions and keV Ar$^+$ irradiation. We also observed that the 91 MeV ^{129}Xe^{23+} ion beam does not significantly change strain or doping of CVD-grown MoS$_2$ monolayers on a similar substrate for fluences of up to $4\cdot10^{12}$cm^{-2}, but sputters away the monolayer at a fluence of $2.5\cdot10^{13}$cm^{-2}. The fluence dependent sputtering can be observed in a decaying intensity of the spectra.

O 53.7 Wed 16:45 MA 005

Hyperlens enabled defect imaging in hBN-covered few-layer graphene — •Lina Jäckering, Konstantin G. Wirth, and Thomas Taubner — I. Institute of Physics (IA), RWTH Aachen University

Most of the unique phenomena of few-layer graphene (FLG) can only be observed when the FLG flake is encapsulated in hexagonal Boron Nitride (hBN) [1]. The fabrication process of encapsulated graphene devices can alter the stacking order and induce defects within the FLG flake [2]. The present stacking order and possible defects significantly influence the graphene sample's electronic properties. Therefore, the visualization of stacking domains and defects in graphene flakes before, during, and after the fabrication of a transport device is of great interest. Here, we show that scanning near-field optical microscopy (s-SNOM) can visualize submicrometer-sized defects in FLG below 33 nm hBN. We attribute the achieved super-resolution imaging of buried defects to the hyberbolic nature of hBN [3]. hBN hosts hyperbolic phonon polaritons that enable super-resolution focusing through a cover layer of hBN, the so called hyperlensing effect [3]. Here, we present the first practical application of the hyperlensing effect in a hBN-FLG heterostructure. Our work paves the way for characterization of FLG devices during fabrication. [1] Dean et al. Nat. Nanotechnol. **5**, 722 (2010). [2] Geisenhof et al. ACS Appl. Nano Mater. **2**, 6067 (2019). [3] Li et al. Nat. Commun. **6**, 7507 (2015).

O 53.8 Wed 17:00 MA 005

Alkali metal (AM) adsorption on pristine and defective graphite surfaces — •Jafar Azizi, Holger Euchner, and Axel Gross — Institute of Theoretical Chemistry, Ulm University, 89069 Ulm, Germany

While carbon derivatives are still the anodes of choice for Li- and post-Li ion batteries, the quest for improving their properties is ongoing. In particular, when considering soft and hard carbon materials the impact of heteroatoms on Alkali metal adsorption and storage is hardly investigated on the atomistic scale. To gain more insight into the underlying mechanisms, a density functional theory-based study of Li, Na, and K adsorption on pristine and defective graphite surfaces, considering different heteroatom impurities (such as N, S, Si, and O), has been performed. Our results show that on the surface of pure graphite,the adsorption of Li and K ions is energetically more favorable as compared to Na ions. However, in the presence of defects and impurities, Na adsorption also becomes feasible. In general, AM adsorption in the vicinity of defects and impurities is largely favored, putting constrain on the number of AM atoms that participate in charge/discharge processes.

O 53.9 Wed 17:15 MA 005

Enhancement of graphene phonon excitation by a chemically engineered molecular resonance — Xiaocui Wu[1], •Nicolas Néel[1], Mads Brandbyge[2], and Jörg Kröger[1] — [1]Institut für Physik, Technische Universität Ilmenau, D-98693 Ilmenau, Germany — [2]Center of Nanostructured Graphene, Department of Physics, Technical University of Denmark, DK-2800 Kongens Lyngby, Denmark

The abstraction of pyrrolic hydrogen from a single phthalocyanine on graphene turns the molecule into a sensitive probe for graphene phonons. The inelastic electron transport measured with a scanning tunneling microscope across the molecular adsorbate and graphene becomes strongly enhanced for a graphene out-of-plane acoustic phonon mode. Supporting density functional and transport calculations elucidate the underlying physical mechanism. A molecular orbital resonance close to the Fermi energy controls the inelastic current while specific phonon modes of graphene are magnified due to their coupling to symmetry-equivalent vibrational quanta of the molecule. Funding by the Deutsche Forschungsgemeinschaft through Grant No. KR 2912/17-1 is acknowledged.

O 53.10 Wed 17:30 MA 005

Accelerated First-Principles Exploration of Structure and reactivity in Graphene Oxide — •Zakariya El-Machachi, Damyan Frantzov, Nijamudheen Abdulrahiman, and Volker L.

Deringer — Department of Chemistry, University of Oxford, Oxford OX1 3QR, UK

Graphene oxide (GO) materials are widely studied, and yet their atomic-scale structures remain to be fully understood. Here we show that the chemical and configurational space of GO can be rapidly explored by advanced machine-learning methods, combining on-the-fly acceleration for first-principles molecular dynamics with message-passing neural-network potentials. The first step allows for the rapid sampling of chemical structures with very little prior knowledge required; the second step affords state-of-the-art accuracy and predictive power. We showcase the performance of the method by applying it to a model of a partially disordered GO nanoflake and its thermal evolution. Our work provides a platform for routine, quantum-mechanically accurate, and realistic-scale simulations of diverse carbonaceous materials.

O 53.11 Wed 17:45 MA 005

Let's Go on Graphs: X-ray Absorption Spectroscopy of Graphene Oxide using Graph Neural Networks — •Samuel J. Hall[1], Kanishka Singh[1,2], Qinyuan Zhou[1,2], and Annika Bande[1,3] — [1]Helmholtz-Zentrum Berlin, Germany — [2]Institute of Chemistry and Biochemistry, Freie Universität Berlin, Germany — [3]Leibniz Universität Hannover, Germany

Graphene oxide (GO) materials, while promising for various applications, can be difficult to fully understand and predict its properties due to the highly irregular molecular structure arising from several oxygen functionalizations across the surface. X-ray absorption spectroscopy (XAS) experiments and simulations can help provide valuable insight by characterizing the electronic structure of materials. However, there are problems with complex spectra being hard to interpret and the prohibitive computational simulation cost for large extended systems. We have developed a machine learning model utilizing graph neural networks (GNN) based on a database of 319 GO-derivative molecules, consisting of 7984 individual atomic XAS spectra calculated with time-dependent density functional theory (TDDFT), that can accurately simulate XAS spectra at a significant lower cost. We show how the model can learn through either the combined spectra of the GO-derivative molecules or the individual atomic spectra to make predictions based on either the larger global environment or the local atomic environment and can further be applied to larger extended systems.

O 54: Scanning Probe Microscopy: Light Matter Interaction at Atomic Scales I

Time: Wednesday 15:00–17:45 Location: MA 041

Topical Talk O 54.1 Wed 15:00 MA 041

Ultrafast scanning tunneling microscopy as a local probe of femtosecond electron and coherent phonon dynamics — •Melanie Müller — Fritz Haber Institute, Berlin, Germany

Ultrafast scanning tunneling microscopy (USTM) has emerged as a powerful tool for imaging electronic and structural excitations at surfaces with angstrom spatial and femtosecond temporal resolution. THz-lightwave-driven STM (THz-STM) and optical photon-assisted STM (ph-STM) offer two approaches [1] to probe the dynamics of charge, orbital and lattice degrees of freedom at the atomic scale. While THz-STM uses single-cycle THz pulses to drive tunneling via classical rectification, ph-STM is based on photon absorption-induced tunneling. Both allow to probe coherent phonons (CPs) and ultrafast changes in the occupation and local density of electronic states (LDOS). CPs modulate the electronic structure of solids, and thus the electronic states involved in tunneling. Here we use ph-STM to probe CPs in ultrathin ZnO/Ag(111) via their coupling to an optical resonance that enhances photon-assisted tunneling [2]. On the other hand, THz-STM allows more direct access to the LDOS by quasi-static tunneling. We study the commensurate charge density wave (CDW) phase in 1T-TaS$_2$, and probe the photoinduced Mott collapse and coherent CDW amplitude motion with THz-STM. The ability to probe both ultrafast local atomic and electronic structure makes USTM unique for imaging spatially inhomogeneous nonequilibrium dynamics in strongly correlated materials at the atomic scale. [1] M. Müller, Prog. Surf. Sci. 100727 (2023) [2] S. Liu et al., Sci. Adv. abq5682 (2022)

O 54.2 Wed 15:30 MA 041

THz waveform optimization for ultrafast time-domain spectroscopy in the scanning tunneling microscope — Shaoxiang Sheng[1,2], Kurt Lichtenberg[1], Felix Huber[1], Johannes Schust[1], Li Chen[3], Susanne Baumann[1], and •Sebastian Loth[1] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]Max Planck Institute for Solid State Research, Stuttgart, Germany — [3]Program in Materials Science and Engineering, University of California San Diego, La Jolla, CA, USA

Exciting the tunnel junction of a scanning tunneling microscope (STM) with sub-cycle THz pulses enables ultrafast spectroscopy of electron, phonon and molecular motion dynamics on surfaces with atomic spatial and sub-picosecond temporal resolution. THz pulses coupled to the STM tip induce short bursts of electron tunneling that are used to sample the ultrafast dynamics. Hence, the sensitivity and time resolution achievable with this technique critically depends on the quality of the THz pulse's electric-field waveform. We use electro-optic sampling of tip-scattered THz light (s-EOS) from the STM tip and pulse-pulse correlation measurements of the THz-induced tunnel current to characterize the shape of the THz electric field in the tunnel junction. Comparison with finite-element modeling of the THz near field identifies reflections and spectral distortions that originate from THz surface plasmon propagation along the STM tip. We find an optimized tip geometry that flattens the electric field waveform in the tunnel junction and enables accurate time-domain spectroscopy.

O 54.3 Wed 15:45 MA 041

Time-domain Detection of Ultrafast Voltage Transients in an STM Tunnel Junction — Vedran Jelic[1], Stefanie Adams[1], Mohamed Hassan[1], Kaedon Cleland-Host[1], •Spencer E. Ammerman[2], and Tyler L. Cocker[1] — [1]Department of Physics and Astronomy, Michigan State University, East Lansing, MI 48824, USA — [2]EMPA, Swiss Federal Laboratories for Materials Science and Technology, 8600 Dübendorf, Switzerland

Lightwave-driven terahertz scanning tunneling microscopy (THz-STM) has emerged as a powerful tool for investigating ultrafast dynamics at atomic length scales. Illuminating the tunnel junction of an STM with a phase-stable single-cycle THz pulse has been shown to induce sub-picosecond tunneling currents coherently driven by the electric field of the THz transient, providing access to the local density of electronic states. A critical factor for interpretation of THz-STM measurements is knowledge of the ultrashort voltage transient. However, the THz transient is subject to spectral modification by variation in the nanoscale dielectric function, as shown by scanning near-field optical microscopy experiments. In order to distinguish between local samples properties and effects arising from terahertz-pulse coupling, a method for extracting the near-field transient is needed. Here, we demonstrate a THz-STM cross-correlation experiment for local time-domain sampling of the THz voltage transient. Combining self-consistent modeling and waveform shaping we introduce a generally applicable scheme for atomic-scale terahertz time-domain spectroscopy, spatially confined to an STM tunnel junction.

O 54.4 Wed 16:00 MA 041

Efficient and continuous THz carrier-envelope phase control for ultrafast lightwave-driven STM — •Jonas Allerbeck[1], Joel Kuttruff[2], Laric Bobzien[1], Lysander Huberich[1], Maxim Tsarev[2], and Bruno Schuler[1] — [1]nanotech@surfaces, Empa, Swiss Federal Laboratories for Material Science and Technology, Überlandstrasse 129, 8600 Dübendorf Switzerland — [2]University of Konstanz, Universitätsstrasse 10, 78464 Konstanz, Germany

THz lightwave-driven scanning tunneling microscopy (THz-STM) facilitates the study of atomically precise structures with sub-cycle picosecond temporal resolution, leveraging the fundamental understanding of quantum systems and carrier dynamics at the nanoscale. Here, I present a versatile THz-STM toolbox, operating at up to 41 MHz repetition rate and using an efficient method to continuously control the carrier-envelope phase of single-cycle THz pulses, established by frustrated internal reflection in a right-angle polymer prism. The setup achieves peak THz voltages at the tunneling junction ranging from 1 V at 41 MHz to few-10 V at 1 MHz, where multi-MHz repetition rates enable state-selective tunneling to localized orbital states of atomic quantum defects below 0.01 electrons per THz pulse. Photoemission sampling and lightwave-driven THz cross correlation benchmark the near-field THz amplitude. We study atomic point defects in few-monolayer transition metal dichalcogenide epitaxial crystals, featuring few-ps charge state lifetime. The precise control of the transient THz waveform at the tunneling junction paves the way towards exploring local carrier dynamics with atomic resolution.

O 54.5 Wed 16:15 MA 041

Towards STM-based atomic-scale scanning near-field optical microscopy — •Fabian Schulz[1], Jun Nishida[2], Adnan Hammud[3], Shuyi Liu[4], Takashi Kumagai[2], Martin Wolf[3], Akitoshi Shiotari[3], and Melanie Müller[3] — [1]CIC nanoGUNE, San Sebastian, Spain — [2]Institute for Molecular Science, Okazaki, Japan — [3]Fritz Haber Institute of the Max Planck Society, Berlin, Germany — [4]Huazhong University of Science and Technology, Wuhan, China

Scattering-type scanning near-field optical microscopy (sSNOM) enables measuring the optical properties of surfaces with a lateral reslolution beyond the diffraction limit. Conventional sSNOM is based on cantilever atomic force microscopy with the resolution limited to typically 10 - 20 nm. Here, using plasmonic nanocavities in a low-temperature scanning tunneling microscope (LT-STM), we demonstrate the potential of STM-based sSNOM in the visible regime to reach a resolution of at least ~1 nm. As a prerequisite for future plasmonic sSNOM using LT-STM, we investigate the interplay between the measured near-field signal and the gap plasmon formed inside the STM junction.

Topical Talk O 54.6 Wed 16:30 MA 041

Ultrafast scanning tunnelling spectroscopy of a phonon-driven atomic vacancy in a monolayer crystal — Carmen Roelcke, Lukas Kastner, Maximilian Graml, Andreas Biereder, Jan Wilhelm, Jascha Repp, Rupert Huber, and •Yaroslav Gerasimenko — Department of Physics and Regensburg Center for Ultrafast Nanoscopy (RUN), University of Regensburg, 93040 Regensburg, Germany

Directly observing in actual microscopic videography how the motion of a specific atom affects the electronic structure and the functionality of solids has been a long-held dream of modern science. Here, we break this ground with tomographic lightwave-driven scanning tunnelling spectroscopy by taking snapshots of the electronic spectrum of a phonon-driven atomic defect faster than the vibration period. We directly resolve in space, time, and energy how atomic motion transiently modulates the bound state of a selenium vacancy, the archetypal single-photon emitter, in a WSe_2 monolayer. By combining atomically-confined excitation of a drum-like phonon with ultrafast tunnelling spectroscopy reaching atomic spatial and 300 fs temporal resolution, we reveal transient energy shifts of the lowest bound defect state by up to 40 meV, depending on the amplitude and phase of the phonon. This combination marks a disruptive development towards understanding and control of quantum matter by accessing key mechanisms at their intrinsic length, time and energy scales.

O 54.7 Wed 17:00 MA 041

Observing correlated electron dynamics in the local density of states of 1T-TaS_2 by THz-STM — •Luis Parra López[1], Alkisti Vaitsi[1], Vivien Sleziona[1], Fabian Schulz[2], Martin Wolf[1], and Melanie Müller[1] — [1]Fritz Haber Institute, Berlin, Germany — [2]CIC nanoGUNE, San Sebastian, Spain

THz scanning tunneling microscopy (THz-STM) has emerged as a promising technique for imaging surfaces with femtosecond temporal and angstrom spatial resolution. The prospect of imaging both local charge and lattice dynamics is of particular interest for the study of strongly correlated materials whose physical properties are governed by strong electron-electron and electron-phonon interactions. 1T-TaS_2 is a fascinating example, where the formation of a commensurate charge density wave (C-CDW) phase is accompanied by the opening of a Mott gap. Femtosecond optical excitation leads to ultrafast collapse of the Mott gap and launches coherent amplitude motion of the CDW. We discuss to what extent both can be probed by THz-STM and decipher the mechanisms by which transient electronic temperatures, ultrafast Mott collapse and coherent AM oscillations modulate the tunneling current. We present a model to extract ultrafast changes in the local density of states in the C-CDW phase of 1T-TaS_2. We reproduce the transient change of the rectified current measured in THz-STM after global ultrafast photoexcitation of 1T-TaS_2. Our results provide the basis to explore ultrafast insulator-to-metal transitions and coherent CDW oscillations with the angstrom spatial resolution provided by THz-STM.

O 54.8 Wed 17:15 MA 041

Combining THz Pump-Probe spectroscopy with Scanning Tunneling Luminescence — •Kurt Lichtenberg, Johannes Schust, Felix Huber, Susanne Baumann, and Sebastian Loth — University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany

Inelastic electron tunnelling in scanning tunnelling microscopes (STM) can trigger atomically-localized emission of light from metallic tunnel junctions, molecules or semiconducting surfaces. This enables studies of light-matter interaction at sub-molecular length scales. Fast gating of the tunnelling current by pulsed voltage sources gives access to the dynamics of the underlying excitation and light emission processes. Electronic pulse generators provide nanosecond time resolution [1], but much faster voltage transients are possible by exciting the STM's tunnel junction with single-cycle THz pulses, which were shown to generate electroluminescence [2]. We combine THz excitation [3] with single-photon detection in a variable-temperature STM and investigate the possibilities for ultrafast and atomically resolved measurements of electroluminescence using THz pump-probe spectroscopy.

[1] C. Grosse, et al. Appl. Phys. Lett. 103 183108 (2013)
[2] K. Kimura, et al. ACS Photonics 8 982 (2021)
[3] M. Abdo, et al. ACS Photonics 8 702 (2021)

O 54.9 Wed 17:30 MA 041

Amplitude Calibration for THz-STM of Atomic Defects in 2D Semiconductors — •Laric Bobzien, Jonas Allerbeck, Spencer Eve Ammermann, and Bruno Schuler — nanotech@surfaces, Empa -Swiss Federal Laboratories for Material Science and Technology, Überlandstrasse 129, 8600 Dübendorf Switzerland

The fundamental understanding of quantum dynamics in low-dimensional materials requires simultaneous ultrafast temporal and atomic spatial resolution, achievable with ultrafast STM. Here, single-cycle THz-pulse-driven STM (THz-STM) combines the atomic spatial resolution with picosecond temporal resolution of the THz field. Point defects in 2D semiconductors, such as vacancies or impurities, are a perfect model systems to investigate strongly correlated dynamics in low dimensions and offer a rich playground for understanding dynamics in the atomic regime. We study point defects in a prototypical family of 2D semiconductors, transition metal dichalcogenides (TMDs), with our multi-MHz repetition rates and efficient THz-STM.

I will show recent measurements of the THz rectified charge as a function of bias and THz field amplitude on 2D TMDs. This allows to map the complex energy landscape of localized states with a resolution of down to 0.01 electrons per pulse enabling precise near-field amplitude calibration. Preliminary THz-STM and THz-STS measurements of a sulfur vacancy in monolayer MoS2 demonstrate state-selective tunneling into distinct electronic orbitals of the quantum defect. These findings pave the way for exploring atomic systems at their native length and time scale.

O 55: Topology and Symmetry protected Materials (joint session O/TT)

Time: Wednesday 15:00–17:45 Location: HL 001

O 55.1 Wed 15:00 HL 001

Interaction Effects in a 1D Flat Band at a Topological Crystalline Step Edge — •Souvik Das[1], Glenn Wagner[2], Johannes Jung[3], Artem Odobesko[3], Felix Kuester[1], Florian Keller[3], Jedrzej Korczak[4], Andrzej Szczerbakow[5], Tomasz Story[4], Ronny Thomale[6], Titus Neupert[2], Matthias Bode[3], Paolo Sessi[1], and Stuart S. P. Parkin[1] — [1]Max Planck Institute of Microstructure Physics, Halle 06120,Germany — [2]Department of Physics, University of Zürich, 8057 Zürich, Switzerland — [3]Physikalisches Institut, Experimentelle Physik II, Universität Würzburg, 97074 Würzburg, Germany — [4]Institute of Physics and International Research Centre MagTop, Institute of Physics, Polish Academy of Sciences, 02-668 Warsaw, Poland — [5]Institute of Physics, Polish Academy of Sciences, 02-668 Warsaw, Poland — [6]Institut für Theoretische Physik und Astrophysik Universität Würzburg, 97074 Würzburg, Germany

We utilize scanning tunneling microscopy and spectroscopy to explore the behavior of the 1D step-edge channels in PbSnSe, a topological crystalline insulator, under the influence of doping. By doping distinct 3d adatoms in PbSnSe we observed that once the energy position of the step edge is brought close to the Fermi level, a new correlation gap starts to open. Our experimental findings are rationalized in terms of enhanced interaction effects since the electron density of states condenses into a 1D channel. This enables us to realize a unique system to study how topology and many-body electronic effects intertwine.

O 55.2 Wed 15:15 HL 001

Quantifying the Electronic Structure of at the Boundary of Topological Insulators by Means of Combined STM and AFM — •Christoph S. Setescak[1], Adrian Weindl[1], Andrea Donarini[2], and Franz J. Giessibl[1] — [1]Institute of Experimental and Applied Physics, University of Regensburg, Universitätstraße 31, 93080 Regensburg, Germany — [2]Institute of Theoretical Physics, University of Regensburg, Universitätstraße 31, 93080 Regensburg, Germany

We show that combined STM and AFM can characterize the surface electronic structure of topological insulators in the Bi2Se3-family. The electronic structure is described by an effective tight-binding Hamiltonian, which is derived from a GW-DFT calculation. A circumspect application of Bardeens model of tunneling current enables us to rigorously interpret tunneling spectroscopy experiments and thus identify characteristic points in the halfspace bandstructure. Furthermore, our experiments demonstrate, that spatially resolved maps of the local density of states can resolve individual electronic degrees of freedom energetically as well as spatially. We observed quasiparticle interference in the topological boundary mode of Bi2Te3 at one-dimensional defects, which enables us to quantify subtle hexagonal warping terms in the dispersion relation of the boundary mode. Our recent measurements, in combination with the ability to structurally characterize defects and the crystal structure at the picometer scale, underscore the potential of combined AFM and STM in exploring the evolution of topological phases amidst varying degrees of disorder.

O 55.3 Wed 15:30 HL 001

Discovery of Segmented Fermi Surface — •Hao Zheng — Shanghai Jiao Tong University, Shanghai, China

Since the early days of Bardeen-Cooper-Schrieffer theory, it has been predicted that a sufficiently large supercurrent can close the energy gap in a superconductor and create gapless Bogoliubov quasiparticles through the Doppler shift of quasiparticle energy due to the Cooper pair momentum[1]. In this gapless superconducting state, zero-energy quasiparticles reside on a segment of the normal state Fermi surface, while its remaining part is still gapped. However, the segmented Fermi surface of a finite-momentum state carrying a supercurrent has never been detected directly. We use quasiparticle interference (QPI) technique to image field-controlled Fermi surface of Bi2Te3 thin films proximitized by the superconductor NbSe2. By applying a small in-plane magnetic field, a screening supercurrent is induced which leads to finite-momentum pairing on topological surface states of Bi2Te3[2]. Our measurements and analysis reveal the strong impact of finite Cooper pair momentum on the quasiparticle spectrum, and thus pave the way for STM study of pair density wave and FFLO states in unconventional superconductors. [1] Phys. Rev. 137, A783-A787 (1965) [2] Science 374, 1381-1385(2021)

O 55.4 Wed 15:45 HL 001

Carrier Injection Observed by Interface-Enhanced Raman Scattering from Topological Insulators on Gold Substrates. — Sarah Scheitz, Tomke Eva Glier, Christian Nweze, •Malte Felix van Heek, Isa Moch, Robert Zierold, Robert Blick, Nils Huse, and Michael Rübhausen — Institute of Nanostructure and Solid-State Physics, University of Hamburg, Hamburg, 22761, Germany

The electron-phonon interaction at the interface between topological insulator (TI) of bismuth/tellurium and gold substrate as a function of TI nanoflakes thickness is studied with a sub-micron Raman spectroscopy. We show the presence of interface-enhanced Raman Scattering and strong phonon renormalization induced by carriers injected from the gold substrate into the topological surface in contact with the gold substrate. The associated electron-phonon coupling shows an approximate linear behavior as function of nanoflake thickness. The strongly nonlinear change of the Raman scattering cross-section as a function of flake thickness can be associated with band bending effects at the metal-TI interface. This provides spectroscopic evidence for strongly modified band structure in the first few quintuple layers of bismuth selenide and tellurium selenide TI.

O 55.5 Wed 16:00 HL 001

Plasmonic Hot carrier Injection from Single Gold Nanoparticles into Topological Insulator (Bi$_2$Se$_3$) Nanoribbons — Christian Nweze[1], Tomke Eva Glier[1], •Mika Rerrer[1], Sarah Scheitz[1], Yalan Huang[2], Robert Zierold[2], Robert Blick[2], Wolfgang Parak[2], Nils Huse[2], and Michael Rübhausen[1] — [1]Institut für Nanostruktur- und Festkörperphysik, Centre for Free Electron Laser Science (CFEL), Universität Hamburg, Luruper Chaussee 149, 22761, Hamburg, Germany — [2]Institut für Nanostruktur- und Festkörperphysik, Centre for Hybrid Nanostructures (CHyN), Universität Hamburg, Luruper Chaussee 149, 22761, Hamburg, Germany

Plasmonic gold nanoparticles (AuNP) injecting hot carriers (HC) into the topological insulator (TI) are studied with sub-micron spatially resolved Raman spectroscopy. We study the impact of single AuNP with sizes between 40 nm–140 nm on the topological surface states of single nanoribbons. In resonance at 633 nm excitation wavelength, we find phonon renormalization in the E_g^2- and A_{1g}^2- modes that we attribute to plasmonic hot carrier injection filling the Dirac cone of the Bi$_2$Se$_3$ TI. The phonon modes are enhanced by a factor of 350 when tuning the excitation wavelengths into interband transition of the Bi$_2$Se$_3$ TI and in resonance with the localized surface plasmon of AuNP. AuNP size-dependent Raman studies show strongest HC injection for particles with a size of 108 nm in agreement with the resonance energy of the localized surface plasmons in AuNP. HC injection opens the opportunity to locally manipulate the electronic properties of the TI.

O 55.6 Wed 16:15 HL 001

Phase-resolved near-field mapping of tight-binding lattices — •Hans-Joachim Schill, Anna Sidorenko, and Stefan Linden — Physikalisches Institut, Universität Bonn, D-53115-Bonn, Germany

The propagation of light in coupled arrays of dielectric loaded surface plasmon polariton waveguides (DLSPPWs) closely resembles the dynamics of a lattice Schrödinger equation, making it an excellent platform for simulating condensed matter lattice systems. The real and momentum space intensity distributions can be simply recorded by leakage radiation microscopy. In order to also capture the local near-field amplitude and phase with sub-wavelength resolution, we employ scattering-type scanning near-field optical microscopy in transmission mode. As an example, to discuss the capabilities of this dual-technique approach, we investigate the topological edge state in a plasmonic variant of the Su-Schrieffer-Heeger model. The topological nature of this state has been shown previously by spectral imaging of the edge mode residing in the band gap [1], but the characteristic electric field oscillation remained elusive, until now.

[1] F. Bleckmann, Z. Cherpakova (Fedorova), S. Linden and A. Alberti, "Spectral imaging of topological edge states in plasmonic waveg-

uide arrays", Phys. Rev. B 96, 045417 (2017)

O 55.7 Wed 16:30 HL 001

The Trimer Chain: Robust Chiral Edge Mode in Artificial Electronic Lattices — •Rian Ligthart[1], Amber Visser[1], Dario Bercioux[2], and Ingmar Swart[1] — [1]Debye Institute for Nanomaterials Science, Utrecht University, the Netherlands — [2]Donostia International Physics Center, 20018 Donostia-San Sebastian, Spain

Artificial electronic lattices are a promising tool to study topology on an atomic scale. The Scanning Tunnelling Microscope (STM) allows to build the topological lattices by manipulating single atoms with nanoscale precision and to probe their electronic properties. Here, the semiconductor InAs(111)A surface with adsorbed Cs atoms is studied. Vertical and lateral manipulation allow to place multiple Cs-adatoms in vicinity of each other. Lines of positively charged Cs atoms create a potential well that confines the surface state electrons of InAs due to local band bending. The confined state acts as an artificial atom and can be used to construct a variety of structures. [1]

Artificial atoms can therefore be used as a platform to study the topological properties of model systems such as the SSH [2] and trimer chain. The trimer chain is similar to the SSH chain but has an extra atom in its unit cell. The extra hopping term introduces an extra degree of freedom in the system allowing to separate the the edge modes on the left and right side of the chain in energy. The trimer chain can therefore host a robust chiral edge mode on one side of the chain.

[1] E. Sierda, et al. Science 380, 1048-1052(2023)

[2] Van Dong Pham, et al. Phys. Rev. B, 105, 125418

O 55.8 Wed 16:45 HL 001

Manipulating Sunken Adatoms on Topological Insulator Bi_2Se_3 — Emma Grasser, •Adrian Weindl, Alfred J. Weymouth, and Franz J. Giessibl — Faculty of Physics, University of Regensburg, D-93053 Regensburg, Germany

Topological insulators (TIs) are a class of materials whose bulk is insulating, while their surface houses gapless topological boundary modes. These surface states possess a linear, spin-polarized dispersion relation and are thus protected from back-scattering by time-reversal symmetry. An open question remains whether surface doping of magnetic elements can suppress this topological protection. Nanostructures made from magnetic adatoms are proposed to enhance the magnetic scattering and may enable the observation of backscattering and the emergence of exotic quantum well states on the surfaces of topological insulators.

To this end, we study the manipulation characteristics of Fe adatoms on the surface of topological insulator Bi_2Se_3 with atomic force microscopy (AFM). The Fe adatoms adsorb in a sunken position in both hollow sites, rendering manipulation difficult. Nonetheless, we find a manipulation mechanism where the Fe atom is pulled from its sunken adsorption site towards the tip. We propose that the Fe atom develops a chemical bond to the probe tip while still bound to the surface as well. This state is stable for tip-sample distances of up to several hundreds of pm and can be used for controlled lateral manipulation. We demonstrate the controllability of manipulation by the construction of a small nanostructure.

O 55.9 Wed 17:00 HL 001

Giant tunable out-of-plane spin polarization in topological antimonene — •Polina Sheverdyaeva[1], Conor Hogan[2,3], Gustav Bihlmayer[4], Jun Fujii[5], Ivana Vobornik[5], Matteo Jugovac[1,6], Asish K. Kundu[1,7], Sandra Gardonio[8], Zipporah Rini Benher[8], Giovanni Di Santo[9], Sara Gonzalez[9], Luca Petaccia[9], Carlo Carbone[1], and Paolo Moras[1] — [1]ISM-CNR, Trieste, Italy — [2]ISM-CNR, Roma, Italy — [3]Dipartimento di Fisica, Università di Roma "Tor Vergata", Roma, Italy — [4]PGI and Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, Jülich, Germany — [5]IOM-CNR, Trieste, Italy — [6]PGI, Forschungszentrum Jülich, Jülich, Germany — [7]ICTP, Trieste, Italy — [8]University of Nova Gorica, Ajdovščina, Slovenia — [9]Elettra - Sincrotrone Trieste S.C.p.A.,Trieste, Italy

We show first-principles calculations and angle- and spin- resolved photoemission studies of a heterointerface built of a single bilayer of β-antimonene and a bulk bismuth selenide. The trivial insulator β-antimonene inherits the topological surface state of the substrate as a result of a topological proximity effect. The new topological state exhibits an unusually high, almost complete out-of-plane spin polarization within the substrate gap, that we explain by a symmetry-protected band crossing of spin-polarized surface states. The spin polarization can be finely and reversibly tuned from nearly full out-of-plane to nearly full in-plane by electron doping. Our findings pave the way towards advanced spintronics applications exploiting the giant out-of-plane spin polarization of topological surface states.

O 55.10 Wed 17:15 HL 001

Orbital-momentum locking in chiral topological semimetal CoSi — •Stefanie Suzanne Brinkman[1], Xin Liang Tan[1,2], Øyvind Finnseth[1], Anders Christian Mathisen[1], Ying-Jiun Chen[2], Christian Tusche[2], and Hendrik Bentmann[1] — [1]Center for Quantum Spintronics, Department of Physics, NTNU, Norway — [2]Forschungszentrum Jülich, Peter Grünberg Institut, Jülich, Germany

The chiral topological semimetal CoSi has a rich electronic structure in which unconventional chiral fermions and long surface Fermi arcs are observed near the Fermi level [1]. We studied the bulk electronic structure in CoSi using soft X-ray angle-resolved photoemission (ARPES) experiments via the ASPHERE endstation at PETRA III (DESY, Hamburg). We investigated the orbital character of the electronic states using circular dichroism in ARPES. Our results reveal the effect of the chiral crystal symmetry on the orbital structure in momentum space, and provide evidence of orbital-momentum locking as recently theoretically predicted [2]. This chirality-driven orbital texture with an accompanying large orbital Hall effect [2] paves the way towards applications of topological semimetals in spin- and orbitronics.

[1] Rao, Z. et al. Observation of unconventional chiral fermions with long Fermi arcs in CoSi. Nature 567, 496-499 (2019).

[2] Yang, Qun et al. Monopole-like orbital-momentum locking and the induced orbital transport in topological chiral semimetals. PNAS 120, 48 (2023).

O 55.11 Wed 17:30 HL 001

Boundary-induced delocalization of vortices in the topological superconductor — •Qiao-Yan Yu — Shanghai Jiao Tong University, Shanghai, China

Planar defects are predicted to induce a delocalization of Abrikosov vortices in superconductors. These delocalized vortices are perturbated by the supercurrent and thus might carry a fraction of a flux quantum. In this work, we constructed a topological superconductor heterostructure by growing Bi2Te3 thin films with molecular beam epitaxy on the superconductor NbSe2. We reveal that the domain boundary induced partial spatial distribution of vortex and screened in-gap bound states within the vortex which crossing the boundary. Our results show the signature of impact of domain boundary on the Majorana zero mode.

O 56: Metal Substrates II

Time: Wednesday 15:00–16:00 Location: MA 043

O 56.1 Wed 15:00 MA 043

In situ observation of the on-surface thermal dehydrogenation of n-octane on Pt(111) — Daniel Arribas[1], Víctor Villalobos-Vilda[1], Ezequiel Tosi[1], Paolo Lacovig[2], Alessandro Baraldi[2], Luca Bignardi[2], Silvano Lizzit[2], José Ignacio Martínez[1], Pedro de Andres[1], Alejandro Gutiérrez[3], José Ángel Martín-Gago[1], and •Pablo Merino[1] — [1]Instituto de Ciencia de Materiales de Madrid — [2]Elettra-Sincrotrone Trieste S.C.p.A. — [3]Universidad Autónoma de Madrid

The catalytic dehydrogenation of alkanes constitutes a key step for the industrial conversion of these inert sp3-bonded carbon chains into other valuable unsaturated chemicals. To this end, platinum-based materials are among the most widely used catalysts. In this work, we characterize the thermal dehydrogenation of n-octane (n-C8H18) on Pt(111) under ultra-high vacuum using synchrotron-radiation X-ray photoelectron spectroscopy, temperature-programmed desorption and scanning tunneling microscopy, combined with ab initio calculations. At low activation temperatures, two different dehydrogenation stages are observed. At 330 K, n-C8H18 effectively undergoes a 100% regioselective single C-H bond cleavage at one methyl end. At 600 K, the chemisorbed molecules undergo a double dehydrogenation, yielding double bonds in their carbon skeletons. Diffusion of the dehydrogenated species leads to the formation of carbon molecular clusters, which represents the first step towards poisoning of the catalyst. Our results disclose the chemical mechanisms behind the first stages of alkane dehydrogenation on a Pt surface at the atomic scale.

O 56.2 Wed 15:15 MA 043

How to measure cones of reaction for single-molecule collisions — •Matthew James Timm[1], Stefan Hecht[2], and Leonhard Grill[1] — [1]Institute of Chemistry, University of Graz, Heinrichstraße 28/IV, 8010 Graz, Austria — [2]Department of Chemistry & IRIS Adlershof, Humboldt-Universität zu Berlin, Brook-Taylor-Str. 2, 12489 Berlin, Germany

Collisions between molecules are essential for chemistry, as they are required to form bonds. Their outcome depends on the collision energy, the relative alignment of reagents and on the impact parameter, which is the miss-distance between the centers of mass of the colliding species. Selection of impact parameter has been demonstrated for on-surface reactions by a "surface-molecular-beam" of CF2 projectiles, formed by dissociating CF3 molecules on a Cu(110) surface with a scanning tunneling microscope tip. These projectiles are directed along the underlying rows of Cu atoms toward a chemisorbed molecular target [1-2]. Control over the relative alignment of the target, however, has remained elusive so far. Here, a singly-debrominated molecular target has been chosen as it can adopt multiple possible adsorption alignments relative to the incoming CF2 projectile. This allows collision at a selected distance from, and alignment with, the center of mass of the target. Accordingly, it paves the way towards an unprecedented ability to map how collision geometry contributes to collision outcome. [1] Anggara, K.; Leung, L.; Timm, M. J.; Hu, Z.; Polanyi, J. C.; Sci Adv., 2018, 4, eaau2821. [2] Anggara, K.; Leung, L.; Timm, M. J.; Hu, Z.; Polanyi, J. C.; Faraday Discuss., 2019, 214, 89-103.

O 56.3 Wed 15:30 MA 043

Crystal growth of alkali and alkali earth metal on the transition metal surfaces — •Yuanyuan Zhou[1] and Chunye Zhu[2] — [1]Technical University of Denmark, Copenhagen, Denmark — [2]Guangdong University of Technology, Jieyang, China

Electrochemical nitrogen reduction is an attractive alternative to the Haber-Bosch process for making ammonia. The promising electrochemical system that produces large amounts of ammonia is the Li- and Ca-mediated process, which has achieved nearly 100% selectivity [Li *et al.* Joule 6, 1-19 (2022), Fu *et al.* Nat. Mater. (2023)]. However, the formation of the Li/Ca dendrites reduce the selectivity and deteriorate the stability of the Li- and Ca-mediated process. Therefore, a prerequisite for avoiding the dendrite formation is an in-depth understanding towards the growth mechanisms in an atomistic level.

We coupled our developed replica-exchange grand-canonical (REGC) [Zhou *et al.* Phys. Rev. B. 100, 174106(2019), Zhou *et al.* Phys. Rev. Lett. 128, 246101(2022)] method with machine-learned interatomic potentials (MLIPs) [Schütt *et al.* PMLR 139, (2021)] to simulate larger length scale and time scale. The MLIPs is constructed in an autonomous active learning during the REGC simulations. We investigated the Li/Ca growth on the transition metal surface (Cu and Fe) using REGC MLIPs-accelerated molecular dynamics. This framework vividly showcased growth process of Li/Ca on Cu/Fe, pinpointing the origin of different growth behavior between Li and Ca.

O 56.4 Wed 15:45 MA 043

Epitaxial CoCrFeNi films for surface investigations — Holger Schwarz[1], Jonathan Apell[1,2], Robert Wonneberger[1], Andreas Undisz[1], Julian Ledieu[3], Vincent Fournée[3], Peter Richter[1], and •Thomas Seyller[1] — [1]Chemnitz University of Technology, Chemnitz, Germany — [2]Friedrich Schiller University Jena, Jena, Germany — [3]Université de Lorraine, Nancy, France

High-entropy alloys (HEAs) are discussed for applications in the fields of corrosion, wear protection and electrocatalysis. Although the surface properties play a central role in these applications, they are still largely unexplored, which is caused by the unavailability of single-crystalline samples. In this presentation, recent progress is reported on the growth and subsequent characterization of epitaxial CoCrFeNi films [1], which were deposited by DC magnetron sputtering from spark-plasma sintered targets [2] on single-crystalline oxide substrates. A characterization of structural, chemical and electronic properties of the films was performed by different techniques including X-ray diffraction, scanning electron and transmission electron microscopy, energy-dispersive X-ray spectroscopy, photoelectron spectroscopy, low-energy electron diffraction and, more recently, by scanning tunneling microscopy. It is demonstrated that epitaxially grown HEA films have the potential to fill the sample gap, allowing for fundamental studies of properties of and processes on well-defined HEA surfaces over the full compositional space. [1] H. Schwarz, et al., Advanced Materials 35 (2023) 2301526. [2] H. Schwarz, et al., Coatings 11 (2021) 468.

O 57: New Methods: Theory

Time: Wednesday 16:15–18:00 Location: MA 043

O 57.1 Wed 16:15 MA 043

Periodic coupled-cluster theory for the ground and excited states with atom-centered basis functions — •Evgeny Moerman[1], Felix Hummel[2], Andreas Irmler[2], Alejandro Gallo[2], Andreas Grüneis[2], and Matthias Scheffler[1] — [1]The NOMAD Laboratory at the FHI-MPG, Berlin — [2]Inst. for Th. Phys., TU Wien, Vienna

Periodic coupled-cluster (CC) theory promises to be a reliable, highly accurate electronic structure method in materials science [1]. The all-electron code FHI-aims [2], which employs numeric atom-centered orbitals (NAOs), has been interfaced to the Cc4s code [3,4], making CC theory for both the ground and excited states accessible to FHI-aims. For molecules, the equation-of-motion-CC (EOM-CC) method predicts quasi-particle energies more accurately than the GW approximation[5], and we expect that this will also hold for bandstructures of solids. Like most correlated wave function methods, CC methods exhibit excessively slow convergence with the size of the super cell. So far, no efficient approach to address this problem exists for NAOs. We present the current state of the CC theory framework available in FHI-aims and possible avenues to address the finite-size error.
[1] G. Booth *et al.*, *Nature*, **493**, 365-370 (2013)
[2] The FHI-aims web page, https://fhi-aims.org
[3] F. Hummel *et al.*, *JCP*, **146**, 124105 (2017)
[4] E. Moerman *et al.*, *JOSS*, **7**, 4 (2022)
[5] M. Lange *et al.*,*JCTC*, **14**, 4224-4236 (2018)

O 57.2 Wed 16:30 MA 043

Efficient and Accurate Calculation of Tunneling Conductance in Molecular Junctions Using Density Functional Theory — •Enrique Montes and Héctor Vázquez — Institute of Physics, Czech Academy of Sciences, Cukrovarnická 10, Prague 162 00, Czech Republic

We present a novel method for the calculation using density functional theory (DFT) of tunneling conductance in molecular junctions for thousands of junction structures. Conductance is estimated within a single scaling parameter by examining clusters where a molecule is bonded to one Au atom at each end and employing unconstrained ab initio molecular dynamics simulations [1]. This method yields accurate DFT-based conductance values across a wide range of geometries.

We investigate two junctions involving conjugated molecules. By examining scenarios where the central benzene ring can freely rotate or is constrained by chemical design [2], we uncover trends in transport properties and establish a clear mapping between molecular structure and junction conductance. Additionally, we evaluate the robustness of our methodology under varied conditions by employing different chemical linker groups (coupling regime) and metallic electrodes [3].

This method overcomes computational limitations, providing reliable conductance evaluation while reducing computational costs by a factor ∼400× as compared to DFT-NEGF transport calculations.
[1] H. Vázquez, J. Phys. Chem. Lett., 13, 9326 (2022).
[2] Y. S. Park, et al. J. Am. Chem. Soc., 131, 10820 (2009).
[3] T. A. Su, et al. Nat. Rev. Mater., 1, 16002 (2016).

O 57.3 Wed 16:45 MA 043

STM simulations with asymmetric/tilted tips: the case of Cu and CO on Cu(111) — Ivan Abilio[1,2] and •Krisztián Palotás[1,2] — [1]Wigner Research Center for Physics, Budapest, Hungary — [2]Institute of Physics, Budapest University of Technology and Economics, Budapest, Hungary

The correct interpretation of experimental scanning tunneling microscopy (STM) images is very important. The presence of asymmetric/tilted tips complicates this procedure and results in reduced symmetry of the STM images. There are only a few theoretical methods, which are able to treat arbitrary tilting of the tip in STM simulations [1,2,3,4]. In the present work, we employ the revised Chen's derivative rules [4] to study the effect of tilted tips on the STM imaging of a Cu adatom and a CO molecule on a Cu(111) surface. Apart from obtaining good agreement with published experimental images [5], our method enables an analysis of the tunneling current in terms of tip orbital contributions, and thus provides a deeper theoretical understanding of the STM contrast formation. We point out important differences between pure and CO-functionalized metal (symmetric and tilted) STM tips. [1] G. Mándi et al., J. Phys. Condens. Matter 25, 445009 (2013). [2] G. Mándi et al., J. Phys. Condens. Matter 26, 485007 (2014). [3] G. Mándi et al., Prog. Surf. Sci. 90, 223 (2015). [4] G. Mándi and K. Palotás, Phys. Rev. B 91, 165406 (2015). [5] A. Gustafsson et al., Phys. Rev. B 96, 085415 (2017).

O 57.4 Wed 17:00 MA 043

Sub-nanometer depth profiling of native metal oxide layers within single fixed-angle X-ray photoelectron spectra — •Martin Wortmann[1], Klaus Viertel[2], Michael Westphal[1], Dominik Graulich[1], Yang Yang[1], Maik Gärner[1], Jan Schmalhorst[1], Natalie Frese[3], and Timo Kuschel[1] — [1]Bielefeld University, Bielefeld, Germany — [2]Bielefeld University of Applied Sciences, Bielefeld, Germany — [3]University of Hawaii, Honolulu, USA

Many metals form nanometer-thin self-passivating native oxide layers upon exposure to the atmosphere, which affect their interfacial properties and corrosion behavior. Such oxide layers are commonly analyzed by X-ray photoelectron spectroscopy (XPS). Here, we propose a simple and accessible approach for the depth profiling of ultrathin oxide layers within single fixed-angle XPS spectra. Instead of using only one peak in the spectrum, as is usually the case, we utilize all peaks within the energy range of a standard lab device, thus resembling energy-resolved XPS without the need for a synchrotron. We derived and tested new models that allow the calculation of depth-resolved concentration profiles at the oxide-metal interface, which are also valid for angular- and energy-resolved XPS. The proposed method not only improves the accuracy of earlier approaches but also paves the way for a more holistic understanding of the XPS spectrum.

O 57.5 Wed 17:15 MA 043

Adapting Explainable Machine Learning to Study Mechanical Properties of 2D Hybrid Perovskites — •Yuxuan Yao[1,2], Dan Han[3,4], Hubert Ebert[4], Aron Walsh[5], David O. Scanlon[3], and Harald Oberhofer[2] — [1]Chair for Theoretical Chemistry, Technical University of Munich — [2]Chair for Theoretical Physics VII, University of Bayreuth — [3]School of Chemistry, University of Birmingham — [4]Department of Chemistry and Center for NanoScience, University of Munich — [5]Department of Materials, Imperial College London

The fast increase of power conversion efficiency (PCE) and low-cost preparation of lead-based halide perovskite photovoltaics are of great interest for use in optoelectronic devices. 2D hybrid organic and inorganic perovskites (HOIPs) have been used as capping layers on top of 3D perovskites to increase the stability and PCE. On the other hand, the soft and stable HOIPs are attractive in sustaining flexible electronic devices. In our work, we utilize explainable machine learning (ML) techniques to accelerate the in silico prediction of elasticities of 2D perovskites, as indicated by their Young's moduli. Our ML models allow us to distinguish between stiff and nonstiff HOIPs and to extract the materials' features most strongly influencing the Young's modulus. The Pb-halogen-Pb bond angle emerges as the dominant physical feature with an inverse correlation to the structural non-stiffness. Furthermore, the cations' steric effect index (STEI) was found to yield rough estimates of non-stiffness. Finally, the deformation of the octahedra strongly affects the mechanical properties, allowing us to perform transfer learning from single layered to multi-layered 2D perovskites.

O 57.6 Wed 17:30 MA 043

Enhanced Sampling of Rare Events Using Boltzmann Generators — •David Greten[1], Karsten Reuter[1], and Johannes T. Margraf[1,2] — [1]Fritz-Haber-Institut der MPG, Berlin — [2]University of Bayreuth

The kinetics of catalytic processes in surface chemistry are determined by rare events such as bond breaking or bond forming reactions. These events are highly challenging to describe in atomistic simulations because brute-force molecular dynamics (MD) simulations would require an unfeasible number of time steps to observe them. Biased MD methods like Metadynamics or Umbrella Sampling represent the most rigorous approach to overcome this limitation. Such enhanced sampling methods are thus the gold standard for the computation of free energy barriers. Nonetheless, they inherit some of the drawbacks of conventional MD, namely long equilibration periods and correlation between samples along the trajectory. In this context, generative machine learning models (such as Boltzmann Generators, BGs) may offer significant advantages, as they can directly generate statistically independent samples from the Boltzmann distribution. In this contribution, we explore how BGs can be used in enhanced sampling simulations of rare events.

O 57.7 Wed 17:45 MA 043

Surface segregation in high-entropy alloys from alchemical machine learning — •Arslan Mazitov and Michele Ceriotti — Laboratory of Computational Science and Modeling, Institute of Materials, École Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland

High-entropy alloys (HEAs), containing several metallic elements in near-equimolar proportions, have long been of interest for their unique bulk properties. More recently, they have emerged as a promising platform for the development of novel heterogeneous catalysts, because of the large design space, and the synergistic effects between their components. In this work we use a machine-learning potential that can model simultaneously up to 25 transition metals to study the tendency of different elements to segregate at the surface of a HEA. We show that, thanks to the physically-inspired functional form of the model, a small amount of data is sufficient to extend a potential that was previously developed using exclusively crystalline bulk phases, so that it can also accurately model defective configurations and surfaces. We then present several computational studies of surface segregation, including both a simulation of a 25-element alloy, that provides a rough estimate of the relative surface propensity of the various elements, and targeted studies of CoCrFeMnNi and IrFeCoNiCu, which provide further validation of the model, and insights to guide the modeling and design of alloys for heterogeneous catalysis.

O 58: Focus Session: Novel Approaches to Surface Plasmon Generated Charges and Heat for Photocatalysis I (joint session O/CPP)

Plasmonic nanostructures present efficent optical antennas to generate energetic charges and/or localized heat for the use in photocatalysis. The time and length scales of typical plasmon-induced processes however pose a formidable challenge, which today still hinders the large-scale implementation of plasmon photocatalysis. The aim of this focus session is to discuss the physical mechanism of plasmon-induced charge-transfer and heat generation processes and exchange ideas on novel materials and devices utilizing these processes.

Organizers: Marc Herzog, Wouter Koopman (U Potsdam)

Time: Wednesday 15:00–17:30 Location: MA 141

Topical Talk O 58.1 Wed 15:00 MA 141

Plasmonic photothermal chemistry on single nanoparticles — •Andrea Baldi — Vrije Universiteit Amsterdam, The Netherlands

Metal nanoparticles strongly absorb light thanks to light-driven oscillations of their free electrons called plasmon resonances. The decay of these resonances can be used to drive chemical reactions via heating of the surrounding environment. In this talk, I will show how we quantify photothermal heating on single nanoparticles using a novel Raman-based thermometry [1] and how we use localized temperature gradients to drive the synthesis of hierarchical core-shell nanostructures [2].

[1] P. Li, S. H. C. Askes, E. del Pino Rosendo, F. Ariese, C. Ramanan, E. von Hauff, and A. Baldi, Nanoscale Thermometry of Plasmonic Structures via Raman Shifts in Copper Phthalocyanine, J. Phys. Chem. C 2023, 127, 9690-9698

[2] R. Kamarudheen, G. Kumari, and A. Baldi, Plasmon-driven synthesis of individual metal@semiconductor core@shell nanoparticles, Nature Communications 2020, 11:3957

O 58.2 Wed 15:30 MA 141

Single molecule layer temperature control via electrically connected SERS gratings — •Thorsten Feichtner[1], Pooja Sutheeshnan[2], Paul Mörk[1], Enno Schatz[3], and Bert Hecht[1] — [1]Nano- Optics & Biophotonics Group, Department of Experimental Physics 5, Röntgen Research Center for Complex Material Research (RCCM), Physics Institute, University of Würzburg, Am Hubland, D-97074 Würzburg, Germany — [2]School of Physics, IISER Thiruvananthapuram, India — [3]NanoStruct GmbH, Friedrich-Bergius-Ring 15, 97076, Würzburg

It would be beneficial for photoelectrochemical experiments in microfluidic environments to be able to locally heat and directly measure the realized temperature. Surface enhanced Raman scattering can be used to locally measure the temperature of molecules, as the scattering peak heights depend on the probability density of vibrational states.

Here we provide evidence that our recently developed two wire grating SERS substrates [1] can be heated very locally using its Ohmic resistance. We measure its resulting temperature using the Raman peaks of a self-assembled monolayer of 4-MBA molecules in dependency of the heating current and fitting a probability density function approach. We back up the experiment with optical and heat simulations. This platform is ready to be implemented in microfluidic systems to measure SERS at the very same position where heat is applied.

[1] Sweedan, Amro, et al., arXiv preprint arXiv:2308.01395 (2023).

O 58.3 Wed 15:45 MA 141

Superlattice excitation driven by electron-phonon interaction measured with UXRD — •Jan-Etienne Pudell[1], Marc Herzog[2], Max Mattern[2], Alexander von Reppert[2], Daniel Schick[3], Ulrike Boesenberg[1], Angel Rodriguez-Fernandez[1], Wonhyuk Jo[1], Roman Shayduk[1], Wei Lu[1], Gregory Malinowski[5], Michel Hehn[5], Matias Bargheer[2,4], and Anders Madsen[1] — [1]European XFEL, Germany — [2]Institut für Physik, Uni Potsdam, Germany — [3]MBI, Berlin — [4]HZB Berlin, Germany — [5]IJL, Université Lorraine, France

The transport of heat (or energy in general) in nanoscopic heterostructures is of great fundamental as well as technological interest, e.g. in the context of thermal management in devices, heat-assisted magnetic recording or plasmonic catalysis. In metal heterostructures, heat conduction is usually dominated by electrons which can be at strong non-equilibrium with lattice vibrations on ultrashort time scales. Utilizing ultrafast x-ray diffraction (UXRD) at the MID end-station at European XFEL, we investigate the ultrafast structural response of a laser-excited metal-metal superlattice (SL) consisting of few-nm thick metal layers with low (Cu) and high (Pt) electron-phonon coupling on top of a 100 nm Cu and 20 nm Ni layer. Using the transient shift and intensity modulations of the respective Bragg peaks, we can quantify the ultrafast heat flow in the SL by comparison to a time-dependent diffusive two-temperature transport model. We identify the excitation of the SL phonon mode as well as an efficient heat transport from the laser excited SL to the Ni-layer via hot electrons.

O 58.4 Wed 16:00 MA 141

How to evaluate temperature at single-molecule, single-particle and ensemble levels — •Simone Ezendam, Julian Gargiulo, and Emiliano Cortés — Physics department, University of Munich, Germany

Understanding temperature dynamics across various scales (single-molecule, single-particle, and ensemble) is pivotal in harnessing plasmonic catalysis for enhanced chemical reactivity. We present three studies investigating this intricate interplay. First, employing super-resolution fluorescence microscopy, we probe single-particle plasmonic catalysis, unveiling distinct pathways and spatial reactivity [1]. Second, through single-particle thermometry, we explore morphology-driven light-to-heat conversion in Au/Pd nanoparticles, unraveling intriguing photothermal responses [2]. Finally, we introduce Anti Stokes thermometry to measure temperature in plasmonic ensembles, validating its efficacy in understanding light-induced heating of nanoparticle arrays [3]. These findings deepen our understanding of plasmonic heating, guiding the design of future nanocatalysts.

[1] 10.1021/acsnano.3c07833, [2] 10.1038/s41467-023-38982-9 [3] 10.1002/adom.202301496

O 58.5 Wed 16:15 MA 141

Measurement of temperature gradients at plasmonic metal surface using thermosensitive microgels — •Yulia Gordievskaya[1], Nino Lomadze[1], Se-Hyeong Jung[2], Andrij Pich[2,3], and Svetlana Santer[1] — [1]University of Potsdam, Potsdam, Germany — [2]RWTH Aachen University, Aachen, Germany — [3]Aachen Maastricht Institute for Biobased Materials (AMIBM), Geleen, The Netherlands

Microgels are recognized for their remarkable responsiveness to changes in environmental conditions[1]. Here, we presented a novel method for local temperature determination in aqueous solutions by observing the inhomogeneous collapse of microgels positioned at plasmonic metal surface. The surface is a variable-thickness gold layer(5-30nm) deposited on glass coated with titanium oxide(<1nm). The localized laser irradiation of the substrate results in the absorption of light by the gold layer, converting it into heat [2]. We studied how the laser wavelength and power levels, as well as distance from the irradiation spot influence the size and shape of the thermosensitive PNIPAM microgel. The obtained dependences allowed us to construct a comprehensive temperature distribution profile relative to the irradiation point.

We thank SFB 1636: Elementary processes of light driven reactions at the nanoscale. [1] J. Jelken, S.-H. Jung, N. Lomadze, Yu.D. Gordievskaya, E.Yu. Kramarenko, A. Pich, S. Santer. Advanced Functional Materials 32.2 (2022), p. 2107946. [2] V. Muraveva, M. Bekir, N. Lomadze, R. Großmann, C. Beta, S.Santer. Applied Physics Letters 120.23 (2022), p. 231905.

Topical Talk O 58.6 Wed 16:30 MA 141

Disentangling plasmonic catalysis contributions by time-resolved spectroscopy — •Holger Lange — The Hamburg Centre for Ultrafast Imaging — Institute of Physics and Astronomy, University of Potsdam

According to current understanding, the nanoparticle plasmon decay is a series of sequential scattering events, separated by their timescales of vastly increasing duration. At first, Landau damping polarizes the electron density. Electron-electron scattering equilibrates the excess energy and subsequent electron-phonon coupling leads to rapid electron cooling and a simultaneous increase in lattice temperature. Several of these processes might contribute to the reaction rate enhancement on plasmonic nanoparticles. I will discuss a range of experiments to specifically address the individual steps of the plasmon decay. The temperature of the thermalized hot electron gas can be observed as a contrast in optical pump-probe experiments, the excited lattice is accessible by time-resolved x-ray scattering. I will present the first reconstruction of the decaying gold nanoparticle plasmon field in the time domain by the detection of transient currents. This data combined with reaction dynamics studies under monochromatic excitation reveals both, non-equilibrium carriers and field-induced excitation as most relevant contributions for two different reactions, highlighting the selectivity of the processes.

O 58.7 Wed 17:00 MA 141

In situ Observation of Nanoparticle Photocharging: Gold Nanorods as Photochemical Capacitors — •Felix Stete[1], Wouter Koopman[1], and Matias Bargheer[1,2] — [1]Institut für Physik & Astronomie, Universität Potsdam, Karl-Liebknecht-Str. 24-25, 14476 Potsdam, Germany — [2]Helmholtz Zentrum Berlin, Albert-Einstein-Str. 15, 12489 Berlin, Germany

Light can induce charge on plasmonic nanoparticles. This idea was used in the last years to explain experimental results on chemical reactions on gold and silver nanoparticles. Here, we give spectroscopic evidence for this photocharging effect and present a method to examine the charge transfer from a hole scavenger to gold nanorods in situ. In contrast to the plasmon resonance in gold nanospheres, the longitudinal resonance of gold nanorods is very sensitive to the free electron density in the metal. We show here how the illumination with light blue-shifts the resonance visualizing the transfer of electrons from ethanol in the solution to the nanoparticles and model the intensity-dependence in a capacitor model. The peak can also be shifted back again after illumination when the particles are left in dark or exposed to molecules with a low reduction potential. We discuss how particle size, oxygen in the solution or the absence of ethanol influence the particle charging.

O 58.8 Wed 17:15 MA 141

Catalytic Electron Transfer Inhibited by Coulomb Blockade in Au Nanoparticles — •Monalisa Garai and Thomas Klar — Institute of Applied Physics, Johannes Kepler University Linz, Altenberger Straße 69, 4040 Linz

Colloidal noble metal nanoparticles (NPs) are excellent catalysts for various chemical redox reactions. They offer alternative reaction pathways and thereby significantly reduce the activation energy for chemical transformations. There are several strategies to reduce the activation energy for kinetically sluggish catalytic reactions, e.g. by changing the size, shape and composition of the NPs and also by changing the specific reaction conditions. To investigate the reaction mechanism of Au NPs of different sizes below 10 nm, we used the 1-electron transfer from ferricyanide (Fe3+) to ferrocyanide (Fe2+) as a model reaction system. We compared the reaction rate of the conversion from Fe3+ to Fe2+ in the presence and absence of the reducing agent triethanolamine (TEOA). It has been shown that the reaction pathway is significantly altered with and without TEOA. TEOA generally transfers electrons to Au NP and causes a negative charging on the Au NP surface. However, if the size of the NPs is very small, especially below 5 nm, the negative charging of the Au NPs is inhibited by the Coulomb blockade effect. As a result, the subsequent catalytic electron transfer from Au NP to Fe3+ is also inhibited, which decreases the overall Fe3+ reduction rate. These experimental results and theoretical calculations will help to develop novel metal NPs for various catalytic processes for environmental remediation.

O 59: Oxide and Insulator Interfaces II

Time: Wednesday 15:00–17:30 Location: MA 144

O 59.1 Wed 15:00 MA 144

Multitechnique characterization of rhodium single atoms on rutile TiO_2(110) — •Moritz Eder, David Rath, Jiri Pavelec, and Gareth S. Parkinson — Institute of Applied Physics, TU Wien, Vienna, Austria

Single-atom catalysis (SAC) has emerged as a prominent research topic in the last decade. The goal is to minimize the noble metal content on metal oxide surfaces by utilizing isolated atoms as active sites. A persistent challenge in the field is the characterization of these single atoms on the surface and showing that they are the dominant sites. Generally, there are few suitable analytical techniques for this on powder catalysts.

We will show a comprehensive surface science approach to the problem. Using isolated rhodium carbonyls on titania as an example, we characterize these single sites using x-ray photoelectron spectroscopy (XPS), temperature-programmed desorption (TPD), and a newly designed infrared reflection absorption spectroscopy (IRAS) apparatus to get the bigger picture. We compare the results to studies on powder catalysts in the literature and show that multitechnique approaches are imperative for characterizing single-atom catalysts.

O 59.2 Wed 15:15 MA 144

Crystal-facet-dependent surface transformation dictates the oxygen evolution reaction activity in lanthanum nickelate — •Achim Füngerlings[1], Marcus Wohlgemuth[2], Denis Antipin[3], Emma van der Minne[4], Ellen Marijn Kiens[4], Javier Villalobos[3], Marcel Risch[3], Felix Gunkel[2], Christoph Bäumer[4], and Rossitza Pentcheva[1] — [1]Department of Physics and Center for Nanointegration Duisburg-Essen (CENIDE), University of Duisburg-Essen, Duisburg, Germany — [2]Peter Gruenberg Institute and JARA-FIT, Forschungszentrum JuelichGmbH, Juelich, Germany — [3]Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Berlin, Germany — [4]MESA+ Institute for Nanotechnology, Faculty of Science and Technology,University of Twente, Enschede, Netherlands

We present a combined experimental and theoretical study of the (001), (110) and (111) facets of $LaNiO_{3-\delta}$ electrocatalyst for water oxidation using electrochemical measurements, X-ray spectroscopy, and density functional theory calculations with a Hubbard U-term. We reveal that the (111) overpotential is $\approx 30-60$ mV lower than for the other facets. While a surface transformation into oxyhydroxide-like NiOO(H) may occur for all three orientations, it is more pronounced for (111). A structural mismatch of the transformed layer with the underlying perovskite for (001) and (110) influences the ratio of Ni^{2+} and Ni^{3+} to Ni^{4+} sites during the reaction and thereby the binding energy of reaction intermediates, resulting in the distinct catalytic activities of the transformed facets.

O 59.3 Wed 15:30 MA 144

How single water molecules adsorb on the calcite(104)-(2x1) surface — Jonas Heggemann[1], Yashasvi Ranawat[2], Jie Huang[2], Adam S. Foster[2], and •Philipp Rahe[1] — [1]Universität Osnabrück, Germany — [2]Aalto University, Finland

Calcite, the most stable polymorph of calcium carbonate and one of the most abundant minerals in the earth's crust, is critically involved in fluid-based dissolution-precipitation processes [1]. The hydrated (104) surface has intensively been studied [2], still, it was only very recently established that the pristine calcite(104) surface expresses a (2x1) reconstruction [3] that is lifted by water adsorption [4].

Here, we investigate the adsorption of single water molecules on calcite(104)-(2x1) by a combination of high-resolution atomic force microscopy (AFM) at 5K using CO-functionalised tips, density functional theory (DFT), and AFM image calculations. From DFT, we find two structurally similar but energetically different adsorption geometries. The excellent agreement between experimental and simulated AFM images confirms the presence of these two structures und unravels microscopic details of the water/calcite system. Our study clarifies the interaction of single water molecules with this most important mineral.

[1] A. Putnis, Rev. Min. & Geochem. 70, 87 (2009)

[2] P. Fenter et al., J. Phys. Chem. C 117, 5028 (2013)

[3] J. Heggemann et al., J. Phys. Chem. Lett. 14, 1983 (2023)

[4] J. Heggemann et al., Phys. Chem. Chem. Phys., Adv. Art. (DOI: 10.1039/d3cp01408h), (2023)

O 59.4 Wed 15:45 MA 144

Physisorption of a small aromatic hydrocarbon: Benzene on In_2O_3(111) — Andreas Ziegler[1], Matthias Preidl[2], Michael Schmid[2], Ulrike Diebold[2], Bernd Meyer[1], and •Margareta Wagner[2] — [1]FAU-Erlangen-Nürnberg, Germany — [2]TU Wien, Austria

In_2O_3 is a complex oxide material with a large unit cell and its (111) surface features different combinations of bulk-like and undercoordinated In and O atoms with 3-fold symmetry. Thus, small molecules can choose their adsorption site(s) from a variety of electronically and chemically different configurations.

In this work we study benzene, the smallest organic aromatic hydrocarbon, on bare and hydroxylated In_2O_3(111). We find that benzene adsorbs in the vicinity of a high-symmetry site of the surface lattice, with three molecules per unit cell in symmetry-equivalent sites. Hydroxylating the In_2O_3(111) surface blocks the preferred adsorption site of the benzene molecules. Still, one molecule can be accommodated in the less-favorable high-symmetry spot of the surface lattice, creating an array of well-spaced single benzene molecules. Both systems are studied with temperature programmed desorption (TPD) and noncontact atomic force microscopy (ncAFM); density functional theory (DFT) calculations and molecular dynamics (MD) simulations provide further insights into the adsorption and mobility of the molecules.

O 59.5 Wed 16:00 MA 144

Bare and Pd-doped ceria thin films prepared by ALD and EBE for hydrogen detection — •Yuliia Kosto[1], Paulina Kapuscik[2], Rudi Tschammer[1], Dominic Guttmann[1], Ewa Mankowska[2], Peter Matvija[3], Carlos Morales[1], Michal Mazur[2], Karsten Henkel[1], Iva Matolinova[3], Jaroslaw Domaradzki[2], and Jan Ingo Flege[1] — [1]BTU Cottbus-Senftenberg, Appl Phys & Semicond Spect, Cottbus, Germany — [2]Wroclaw Univ Sci & Technol, Fac Elect Photon & Microsyst, Wroclaw, Poland — [3]Charles Univ, Dept Surface & Plasma Sci, Prague , Czech

The need to store and use hydrogen safely as part of green economy based on renewable energy evokes a necessity to reliably detect it at ambient conditions. The majority of currently used sensors are working at elevated temperatures (200-500 °C). In this work, we demonstrate that ceria films deposited on a commercial electrode by atomic layer deposition (ALD) and electron beam evaporation (EBE) electrically respond to hydrogen (from 20 to 500 ppm) at much lower temperatures (50-200 °C). The results reveal that <1.5 nm thin Pd adlayer increases the electrical response by several orders of magnitude for both ceria films. The NAP-XPS study under changing oxidative/reductive atmospheres sheds light on the mechanism of Pd-CeOx thermal activation and the role of the deposition technique in the reactivity of the oxide.

O 59.6 Wed 16:15 MA 144

Infrared spectroscopic investigations of the interaction between formic acid and ceria surfaces — •Zairan Yu, Shuang Chen, Alexei Nefedov, Christof Wöll, and Yuemin Wang — Institute of Functional Interfaces(IFG),Karlsruhe Institute of Technology(KIT), Eggenstein-Leopoldshafen, 76344, Germany

Formic acid (HCOOH) as a simplest carboxylic acid with carboxylate functional group is involved in a wide variety of industrial processes. Ceria shows excellent catalytic activity in numerous reactions due to the unique redox properties and high oxygen storage capacity. Given the great complexity of nanostructured ceria catalysts, a surface science study based on well-controlled model systems is required to provide reliable and comprehensive reference data. In this work, we present a systematic IR study of the interaction between HCOOH and CeO2 surfaces of both single crystals and nanoparticles using a sophisticated UHV apparatus dedicated to IRRAS experiments at grazing incidence on monocrystals and FTIR transmission measurements for powders. The polarization-resolved IRRAS revealed that the dissociative adsorption of HCOOH occurs on both oxidized and reduced CeO2(111) surfaces, giving rise to formate species in different adsorption configurations. The reliable reference IRRAS data acquired for ceria single-crystal surfaces allowed us to unambiguously assign the IR bands observed on ceria powder samples. This work was funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation)-Project-ID 426888090-SFB 1441.

O 59.7 Wed 16:30 MA 144

Selective reduction of α-Fe_2O_3 (0001) by hydrogen adsorption and incorporation — •Muhammad Munawar and Rossitza Pentcheva — Department of Physics, University of Duisburg Essen, Germany.

The reduction of iron oxides using hydrogen as a reducing agent may be an alternative route towards the production of green steel. In this context, we perform DFT+U calculations to explore the initial adsorption and incorporation of hydrogen in the near surface region of hematite (0001). Starting with the Fe-O_3-Fe termination, which is stable in a wide range of oxygen chemical potentials, we vary the H concentration and distribution in the surface layers. The adsorption energy per H indicates that incorporation of H on the surface or in the near-surface region is favored, in contrast to incorporation in bulk hematite. H binds to an oxygen ion, while simultaneously an Fe^{3+} in the vicinity turns into Fe^{2+}, leading to a pronounced distortion of the lattice. The extent of localization of the donated electron depends on the layer where H is incorporated, suggesting a varying mobility of excess electrons. We find that the bandgap decreases with increasing hydrogen concentration at the surface, whereas a semiconductor-to-half metal transition is observed when H is incorporated in deeper layers.

Funding within IMPRS SUSMET and computational time at Leibniz Rechenzentrum are acknowledged

O 59.8 Wed 16:45 MA 144

Extended support structure dictates the reactivity of model single-atom catalysts for dissociative oxygen adsorption — •Faith J. Lewis, Ali Rafsanjani-Abbasi, Matthias Meier, Michael Schmid, Ulrike Diebold, and Gareth S. Parkinson — Institute of Applied Physics, TU Wien, Vienna, Austria

A goal of single-atom catalysis (SAC) is to find a support that stabilizes single metal adatoms in geometries that make them catalytically active.[1] For this to be possible, the adatoms must be able to change their coordination state by forming and breaking bonds. Iron oxides are popular supports used in SAC because of their low cost, chemical stability, and non-toxicity.[2] Given its ubiquity in catalysis, platinum is an attractive metal to be used in SAC.

I will discuss the similarities and differences between Pt adatoms on hematite, α-Fe_2O_3(012)-(1x1), and magnetite, Fe_3O_4(001). Scanning tunneling microscopy (STM), x-ray photoelectron spectroscopy (XPS), and density functional theory (DFT) were used to characterize these surfaces and how Pt atoms bind to them. In both cases, Pt is 2-fold coordinated to lattice oxygen atoms, but the reactivity differs. Interestingly, we find that the second coordination sphere plays an important role defining the reactivity to molecular oxygen.

1. Kraushofer, F., Single Atom Catalysis: Insights from Model Systems, Chemical Reviews, 2022. 122, 18, 14911-14939.

2. Parkinson, G.S., Iron oxide surfaces. Surface Science Reports, 2016. 71(1): p. 272-365.

O 59.9 Wed 17:00 MA 144

Calculating core-level binding energies of small molecules on Fe_3O_4(111) — Pauline Schütt[1,2,3], Heshmat Noei[2], Wernfried Mayr-Schmölzer[1], Andreas Stierle[2,3], and •Gregor Vonbun-Feldbauer[1] — [1]Institute of Advanced Ceramics, TU Hamburg, Germany — [2]DESY NanoLab, Deutsches Elektronen-Synchrotron, Hamburg, Germany — [3]Fachbereich Physik, Universität Hamburg, Germany

The identification of species adsorbed on surfaces, especially under reactive conditions, is crucial for a better understanding and subsequent optimization of processes in various applications ranging from heterogeneous catalysis to hybrid nanocomposites. Here, various small organic and inorganic carbon compounds adsorbed on a magnetite (111) surface are studied computationally using Density Functional Theory (DFT). These molecules on magnetite surfaces are highly relevant, for example, in Fischer-Tropsch synthesis. After geometry optimization and a stability analysis for selected molecules, the core-level binding energies of the oxygen and carbon atoms involved are calculated using different approximations within DFT. The different approaches are compared. The results are rationalized and connected with experimental X-ray photoemission spectra to support the identification of species.

O 59.10 Wed 17:15 MA 144

Surface Structure of Vicinal Magnetite and the Adsorption of Formic Acid — •Marcus Creutzburg[1], Heshmat Noei[1], Vedran Vonk[1], Wernfried Mayr-Schmölzer[2], Gregor Vonbun-Feldbauer[2], and Andreas Stierle[1,3] — [1]Centre for X-ray and Nano Science (CXNS), Deutsches Elektronen-Synchrotron DESY, Hamburg — [2]Institut für Keramische Hochleistungswerstoffe, Technische Universität Hamburg — [3]Fachbereich Physik, Universität Hamburg

The study of vicinal magnetite (Fe_3O_4) surfaces, with regularly ordered terraces and steps, and their interaction with carboxylic acids is one approach towards an atomic understanding of the oxide/organic interface in complex nanomaterials and active facets during catalytic reactions. In this contribution a (112)-oriented magnetite single crystal is studied using various experimental techniques complemented by DFT calculations. We can show using LEED and STM that this surface is stable after UHV preparation and presents (001)- and (111)-oriented nanosized terraces, confirmed by SXRD. Formic acid (HCOOH) is typically used as a probe molecule and as a representative for larger carboxylic acids. FT-IRRAS reveals that three different formate species are present on the surface after adsorption of formic acid: bidentate bridging on the (001) facets and chelating and quasi-bidentate on the (111) facets, similar to the respective planar surfaces [1,2]. Polarization-dependent FT-IRRAS reveals that the majority of formate is adsorbed across the step edges. [1] Arndt et al., Comm. Chem. 2(92) (2019) [2] Creutzburg et al., J. Phys. Chem. Lett. 12(15) (2021)

O 60: Solid-Liquid Interfaces III: Reactions and Electrochemistry

Time: Wednesday 15:00–17:45 Location: MA 042

O 60.1 Wed 15:00 MA 042

An from insightful ab-initio view molecular on free dynamics energies at metal-water interfaces — •Fabiola Dominguez Flores, Sung Sakong, and Gross Axel — Institute of Theoretical Chemistry, Ulm University

Solid-liquid interfaces are a key element of all technological processes and devices in interfacial electrochemistry, where atomistic modeling encounter major challenges in measuring accurately thermodynamic properties comparable to experiments. Ab initio molecular dynamics (AIMD) simulations have been used to sample the statistical properties of the electrochemical interfaces. However, there is no feasible method to treat the entropy, only the structural properties of the simulations can be safely analyzed. Using the two-phase thermodynamic (2PT) model, which estimates the entropy from the vibrational density of states of liquids, we obtained the free energy of the electrochemical interfaces of ion-free water on several metal surfaces. Based on the grand canonical thermodynamic scheme, we will discuss the free energy of solvation on metal surfaces and their respective energetics at the interfaces. The information is essential to understand the energetics at the electrochemical interfaces as well as for further development of effective solvent modeling and parametrization of solvent models.

O 60.2 Wed 15:15 MA 042

Oxidation of functionalized monolayers on gold in classical molecular dynamics (MD) — •Ludwig Ahrens-Iwers, Gregor Vonbun-Feldbauer, and Robert Meissner — Hamburg University of Technology, Hamburg, Germany

Monolayers of thiols can self-assemble on gold surfaces. If they are functionalized with ferrocene, this functional group can oxidize and transfer an electron to the gold surface in the process. In order to allow an electron transfer in classical MD, partial charges of gold atoms are modeled using the constant potential method (CPM) which distributes charges on metal electrodes. The charge transfer from the ferrocene to the gold surface is modeled using free energy perturbation methods giving access to the reduction potential.

As an implementation of the CPM, the ELECTRODE package for the MD code LAMMPS is presented[1]. This package features a particle-mesh solver to greatly reduce computation times of the long-range Coulomb interactions[2].

[1] L. Ahrens-Iwers et al., J. Chem. Phys. 157, 084801 (2022)

[2] L. Ahrens-Iwers, R. Meißner, J. Chem. Phys. 155, 104104 (2021)

O 60.3 Wed 15:30 MA 042

SiO2-water interface with long-range Charge Equilibration (Qeq)

— •Kamila Savvidi[1], Ludwig Ahrens-Iwers[1], and Robert Meissner[1,2] — [1]Hamburg University of Technology, Hamburg, Germany — [2]Helmholtz-Zentrum Hereon, Geesthacht, Germany

Charge equilibration (Qeq) methods enables the accurate calculation of partial charges in molecular dynamics. The charges are predicted based on the minimization of the electrostatic potential energy, which is in principle calculated up to a cut-off radius. Long-range contributions improve the accuracy of the Coulomb interactions in a system with periodic images, that can advance the prediction of the atomic charges. To this end, we implemented a long-range Qeq scheme in a similar way to the original Qeq method [1] by adding terms with the electronegativity and atomic hardness to the electrostatic energy. The long-range contributions to the Coulomb energy are calculated using Ewald summation and gaussian charges. This method is part of the ELECTRODE package in LAMMPS [2] and shows agreement with the constant potential method when an electric potential is applied. SiO2 and Si electrodes with aqueous electrolytes are studied using the method and compared to results with other Qeq models.

[1] A. Rappe, W. Goddard, Phys. Chem., 95, 3358-3363 (1991) [2] L. Ahrens-Iwers, M. Janssen, S. Tee, R. Meißner, J. Chem. Phys. 157, 084801 (2022)

O 60.4 Wed 15:45 MA 042

Understanding Competitive Adsorption at Metal-Water Interfaces via Cavity Formation

— •Thorben Eggert[1,2], Nicolas G. Hörmann[1,2], and Karsten Reuter[1] — [1]Fritz-Haber-Institut der MPG, Berlin — [2]Technische Universität München

Adsorption energies from the gas phase differ significantly from their respective values in aqueous environments, e.g. the reduced adsorption energy of phenol on Pt(111) in water [1]. While computationally costly, explicit solvation models can partially capture this reduction, adsorption energies of phenol are not changed in implicit solvation models [2]. The latter deficiency arises from the common approximation to base the cavity formation cost, i.e. the exclusion of solvent by the adsorbate, solely on bulk liquid energetics.

To measure the missing energetic contribution from the competitive binding of adsorbate *and* solvent, we study the free energy of cavity formation with classical molecular dynamics simulations and free energy perturbation via the Multistate Bennett Acceptance Ratio. We demonstrate that cavity formation at interfaces depends on the interaction strength between the substrate and the solvent, which can be rationalized by the respective adsorption energy [3]. These results allow for a quantification of the competitive nature of adsorption processes at solid-liquid interfaces, which is a prerequisite for a parametrized substrate-specificity in improved solvation models.

[1] N. Singh and C.T. Campbell, ACS Catal. **9**, 8116 (2019).

[2] P. Clabaut *et al.*, J. Chem. Theory Comput. **16**, 6539 (2020).

[3] T. Eggert *et al.*, J. Chem. Phys. **159**, 194702 (2023).

Topical Talk O 60.5 Wed 16:00 MA 042

Ab initio insights into atomistic processes at electrified solid/liquid interfaces

— •Mira Todorova[1], Jing Yang[1], Sudarsan Surendralal[1], Florian Deissenbeck[1], Stefan Wippermann[1,2], and Jörg Neugebauer[1] — [1]Max-Planck-Insitut für Eisenforschung GmbH, Düsseldorf, Germany — [2]Phillips-Universität Marburg, Germany

Many of the challenges we face today towards achieving a greener economy focus on processes occurring at electrified solid/liquid interfaces. Understanding how the interactions between the solid, the liquid and dissolved species are affected by the applied potential will aid our ability to achieve rational design and targeted optimization of relevant processes. Ab initio molecular dynamic simulations have proven an indispensable tool to gain insights at the scale of atoms and electrons. The talk will present insights into reactions at electrified solid/liquid interfaces enabled by our recent developments of a thermopotentiostat [1,2], and the coupling of DFT calculations with thermodynamic models. The power of these approaches will be demonstrated for two examples: H adsorption on the Pt electrode [3] and Mg surface stability and dissolution.

[1] S. Surendralal, M. Todorova, M. Finnis, and J. Neugebauer, Phys. Rev. Lett. 120, 246801 (2018)

[2] F. Deißenbeck, C. Freysoldt, M. Todorova, J. Neugebauer, and S. Wippermann, Phys. Rev. Lett. 126, 136803 (2021)

[3] S. Surendralal, M. Todorova, and J. Neugebauer, Phys. Rev. Lett. 126,166802 (2021)

O 60.6 Wed 16:30 MA 042

Response Properties of Water at the Electrified Pt(111) Surface

— •Lang Li, Nicolas G. Hörmann, and Karsten Reuter — Fritz-Haber-Institut der MPG, Berlin

Metal-water interfaces play a fundamental role in electrochemistry. An accurate understanding of their properties is required in any attempt to describe electrochemical phenomena such as electrocatalytic reactions or charge transfer processes. In this work, we benchmark the description of electrified Pt(111)/water interfaces based on *ab initio* molecular dynamics simulations at applied potential conditions using density functional theory. We apply the potential by introducing excess electrons that are counterbalanced by partially charged hydrogen atoms. This method is tested with a variety of slab setups and cell sizes. We analyze in detail the structure of the interface as well as the obtained capacitance vs. potential curves and compare with published theoretical and experimental results [1,2]. In addition, we decompose the interfacial water response on Pt(111) into dipolar and chemisorption-related contributions as a function of the introduced electronic excess charge, and introduce a nuanced four-state model of interfacial water with one chemisorbed and three physisorbed states. Our results highlight the response of interfacial water to an applied potential and its importance for understanding overall capacitance, in particular at high electrolyte concentrations [3]. [1] A. Bouzid and A. Pasquarello, J. Phys. Chem. Lett. **9**, 1880 (2018). [2] J.B. Le, Q.Y. Fan, J.Q. Li, and J. Cheng, Sci.Adv. **6**, eabb1219 (2020). [3] L. Li, Y.P. Liu, J.B. Le, and J. Cheng, Cell Rep. Phys. Sci. **3**, 100759 (2022).

O 60.7 Wed 16:45 MA 042

Oxygen Adsorption at the Electrochemical Metal/Water Interface: Au(111) vs. Pt(111)

— •Alexandra M. Dudzinski, Elias Diesen, Karsten Reuter, and Vanessa J. Bukas — Fritz-Haber-Institut der MPG, Berlin

Due to its key role in fuel cell technologies, the oxygen reduction reaction (ORR) has been the subject of extensive studies over the last decades. Dedicated experiments on model single-crystal electrodes have specifically served to establish fundamental trends across transition metal catalysts, e.g., in terms of product selectivity and the effect of applied potential. Such studies revealed that in contrast to Pt-based electrodes, Au shows a distinct preference towards forming hydrogen peroxide (rather than water) with activity that depends strongly on the (absolute) electrode potential. The microscopic origins of this difference are still not well understood and likely lie in details of key elementary reaction steps. Using *ab initio* molecular dynamics, we recently predicted O_2 adsorption as a critical, potential-dependent step of the ORR on the weak-binding Au(111) surface [1]. In this study, we now draw a direct comparison to the more reactive Pt(111) surface and (i) systematically analyze structural and dynamical properties of the two electrochemical interfaces, as well as (ii) investigate O_2 adsorption as a function of potential. Prominently, we find the more reactive Pt(111) to be predominantly covered by specifically adsorbed solvent species under operating ORR conditions, while chemisorbed O_2 shows an almost negligible response to potential.

[1] Dudzinski *et al.*, ACS Catal. **13**, 12074 (2023).

O 60.8 Wed 17:00 MA 042

Interfacial properties of non-planar Platinum slab in contact with water via Deep Potential Molecular Dynamics (DP-MD)

— •Muhammad Saleh[1], Alexander Lozovoi[2], Riccardo Martina[1], Clotilde S. Cucinotta[2], and Marialore Sulpizi[1] — [1]Theoretical Physics of electrified liquid-solid interface, Ruhr-University Bochum, Germany — [2]Department of Chemistry, Imperial College London, UK

One of the crucial aspects that determines the reaction of the metal-liquid is its surface properties, for instance, the dynamics of interfacial interactions between metal-liquid and liquid-liquid species or surface charge distribution. Knowing these mechanisms gives us an insight into the development of catalysts, batteries, or electrochemical reactions in general. However, obtaining the atomistic resolution is experimentally challenging, while most atomistic modeling only focuses on the clean planar surface, which is somewhat inaccurate given that the surface is primarily rough under actual conditions. On the other hand, an adequate representation of a model system requires a sizable number of atoms (i.e., more than 1000 atoms), which increases the computational cost, particularly when employing high-quality approaches. In this work, we adopt the deep neural networks (DNNs) model named deep potential molecular dynamics (DPMD) to reproduce the potential energy surface (PES) of ab initio molecular dynamics (AIMD), enabling an extensive simulation trajectory. As a result, a thorough analysis of the surface characteristics of platinum metal in contact with water is possible.

O 60.9 Wed 17:15 MA 042

Extracting free energy charge transfer paths of solvated ions from molecular dynamic simulations — •Zhenyu Wang, Mira Todorova, Christoph Freysoldt, and Jörg Neugebauer — Max-Planck-Institut für Eisenforschung GmbH, Max-Planck-Str.-1, D-40237, Düsseldorf, Germany

The Marcus theory is indispensable for understanding and predicting electron transfer reactions, which are fundamental in various chemical and biological systems. While solvation shells play a key role in stabilizing ions in solution, a quantitative approach to assess charge transfer intricacies is currently absent. This study introduces a quantitative model to elucidate solvation shell formation precisely. In our model, ions' charges (q_{core}) serve as generalized coordinates, and we derive free energy profiles as functions of q_{core}. We present a systematic workflow for generating solvation configurations around ions with varying charges. Thermodynamic integration aligns the free energy of ions in different charge states. Our approach successfully reproduces Marcus parabolas, revealing a pivotal transition in the solvation mode at $q_{core} \approx 0.5$ e. This model significantly enhances our ability to quantitatively analyze charge transfer complexities, providing fresh insights into solvation shell dynamics. The outcomes of this work holds promise for advancing the understanding of electron transfer reactions in chemical and biological contexts.

O 60.10 Wed 17:30 MA 042

Power of the interface: Understanding (electro-)valorization of biomass-derived chemicals — •Sihang Liu[1], Nitish Govindarajan[2], and Georg Kastlunger[1] — [1]Catalysis Theory Center, Department of Physics, Technical University of Denmark (DTU), 2800 Kgs. Lyngby, Denmark — [2]Materials Science Division, Lawrence Livermore National Laboratory, 7000 East Avenue, Livermore, California 94550, United States

In this contribution, we combine grand-canonical density functional theory (GC-DFT) calculations, microkinetic modeling, ab initio molecular dynamics (AIMD) and experiments to study aqueous-phase furfural adsorption on various transition metal surfaces and understand trends in reactivity of copper for furfural electroreduction. Using AIMD simulations of the metal-water interfaces, we find furfural binding strength in aqueous phase is reduced due to water replacement and reorganization. We then identify the binding energy of OH in vacuum to be a good descriptor to estimate the solvation energy of furfural(Ref1). In the latter study on Cu electrodes, we combine GC-DFT based microkinetic modeling and pH dependent experiments to highlight the predominant role of proton-coupled electron transfer-based pathways and elucidate the possible rate-determining steps towards furfuryl alcohol and 2-methyl furan, respectively(Ref2). Our work improves understandings of interfacial catalysis and chemistry to upgrade furfural and other biomass-derived chemicals. 1. Liu et al. J. Chem. Phys. 2023, 159, 084702 2. Liu et al. EES. Catal., 2023, 1, 539-551

O 61: Focus Session: 2D Transition Metal Carbides, Nitrides and Carbonitrides I (joint session DS/MM/O)

MXenes, two-dimensional transition metal carbides, nitrides and carbonitrides, constitute one of the most rapidly growing class of 2D materials. Discovered in 2010, they have demonstrated exceptional physical, chemical, and electronic properties leading to potential applications in various fields, such as energy storage, catalysis, electromagnetic interference shielding, sensing, and biomedicine. The fundamental physical properties of MXenes are governed by their chemical composition and great research efforts are currently devoted to expanding the range of existing MXenes by tuning their stoichiometry, morphology and surface chemistry as well as adding tailored defects that can bring new functionalities. In this Focus Session, new developments related to MXene synthesis and characterization will be presented. The fundamental physical properties and interfacial processes correlated with MXene surface chemistry, defects and interlayer confinement will be discussed.

Organizers:

Dr. Tristan Petit, Head of the Young Investigator Group Nanoscale Solid-Liquid Interfaces, Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Berlin, Germany

Prof. Vincent Mauchamp, Institut Pprime, CNRS-Université de Poitiers-ISAE ENSMA, Poitiers, France

Time: Wednesday 15:00–17:00 Location: A 053

Introduction Focus Session on MXene - V. Mauchamp & T. Petit

Invited Talk O 61.1 Wed 15:15 A 053

Sustainable synthesis of MXenes and their precursors — •Jesus Gonzalez-Julian[1], Nima Amousa[1], and Filipa Oliveira[2] — [1]Chair of Ceramics, Institute of Mineral Engineering (GHI), RWTH Aachen University, 52074 Aachen, Germany — [2]Department of Inorganic Chemistry, Faculty of Chemical Technology, University of Chemistry and Technology Prague, 166 28 Prague 6, Czech Republic

Synthesis of MXenes is typically performed using HF - or forming insitu HF through the reaction between LiF and HCl, but this chemical etching presents some limitations. HF is highly corrosive and toxic, requiring specific safety regulations, and the chemical etching is highly exothermic, limiting the amount of MXenes that can be produced in the range of grams. These two points are hindering the transfer of MXenes to real applications, despite the excellent properties of these 2D materials. Consequently, new synthesis processes are required to overcome these problems. In this work, we will present a novel sustainable synthesis route for the synthesis of the precursors (MAX phases) and the chemical etching, which is referenced as Molten Salt Shielded Synthesis or MS3. MS3 is carried out at lower temperatures than conventional synthesis routes, in air instead inert atmospheres, and does not require any milling step to obtain fine and loose powders.

O 61.2 Wed 15:45 A 053

Tuning the molten salt etching process by in situ XRD — •Julian T. Müller, Aleksander Gurlo, and Maged F. Bekheet — Technische Universität Berlin, Faculty III Process Sciences, Institute of Material Science and Technology, Chair of Advanced Ceramic Materials, Straße des 17. Juni 135, 10623 Berlin, Germany

MXenes, whose properties are mainly defined by their chemical composition and surface terminations, could be synthesized by etching an A element (A = Al, Si, etc.) from the parent MAX phase using hydrofluoric acid. This acid is of high risk to human health and leads to fluorine terminations on the surface of MXene, which are difficult to alter and may be detrimental for certain applications, e.g. electrodes in different battery systems. Etching via molten salt formation offers an alternative, less harmful way. By sophisticated selection of salts, it is possible to adjust the surface terminations and pave the way for a new generation of tailored MXenes. Gaining insight into the molten salt etching process and tuning its process parameters such as temperature, duration, and precursor ratio are keys to ensuring a qualitative MXene and saving time, energy, and cost. To support this, we are currently establishing an in situ XRD setup at the Advanced Light Source of the Lawrence Berkeley National Laboratory. It will allow measurements under various gas atmospheres up to 1450 °C and 50 bars, including a vapour phase. In the first molten salt etching experiments, we successfully gained knowledge on the etching onset, duration, and phase composition. With this measurement setup, the synthesis of MAX phases and many MXene and salt combinations await to be explored.

O 61.3 Wed 16:00 A 053

Preparation of Magnetic MXenes by Fe intercalation — Tim Salzmann[1], Hanna Pazniak[2], Thierry Ouisse[2], Fabrice Wilhelm[3], Andrei Rogalev[3], Ralf Meckenstock[1], Ivan Tarasov[1], Michael Farle[1], and •Ulf Wiedwald[1] — [1]University of Duisburg-Essen and Center for Nanointegration Duisburg-Essen, Germany — [2]Université Grenoble Alpes, CNRS, Grenoble INP, LMGP, Grenoble, France — [3]European Synchrotron Radiation Facility, Grenoble, France

Yet, the success of generating magnetic MXenes remains very limited since Fe, Co or Ni are incompatible with MAX phase precursors. We present an alternative approach to master magnetic properties of 2D MXenes by intercalating Fe into $\mathrm{Ti_3C_2T_x}$ MXene thin films on $\mathrm{Si/SiO_2}$ in UHV conditions. Annealing of bare MXenes at $T = 1023$ K removes -F, -Cl and -OH. The intercalation of Fe is studied by depositing a 6 nm film on top of the MXenes and subsequent annealing. XRD shows an increase of the interplanar spacing between MXene sheets of 0.16 ± 0.02 nm. XPS and XANES reveal that Fe remains metallic and Fe diffuses 30 nm deep into the multilayers while MXenes keep intact. We study the magnetic properties by VSM and FMR. VSM suggests a new magnetic phase with $M_S = 660 \pm 80$ kA/m and a Curie temperature of 485 K while angular-dependent FMR at 9 GHz and 300 K shows two ferromagnetic and one paramagnetic signals, which we address to the remaining Fe on top and the intercalated Fe in form of quasi 2D disks and isolated Fe ions. Funded by DFG (530103526). Support by CRC/TRR 270 (405553726) is acknowledged.

O 61.4 Wed 16:15 A 053

synthesis and characterization of 2D Mo/Ti solid solutions based MXene for hydrogen evolution reaction in alkaline media. — Lola Loupias[1], Claudia Morais[1], Sophie Morisset[1], Christine Canaff[1], Zheming Li[1], Patrick Chartier[2], Vincent Mauchamp[2], Thierry Cabioc'h[2], Aurélien Habrioux[1], and •Stéphane Célérier[1] — [1]Institut de Chimie des Milieux et Matériaux de Poitiers (IC2MP), Université de Poitiers, CNRS, F-86073 Poitiers, France — [2]Institut Pprime, UPR 3346 CNRS, Université de Poitiers, ISAE-ENSMA, BP 30179, 86962 Futuroscope-Chasseneuil Cedex, France

2D MXenes have gained an ever-increasing attention in various application fields. Their properties can be strongly tuned by modifying the M element in the Mn+1XnTx structure. Among them, Mo-based MXenes are beginning to be successfully explored in many areas. This work focuses on the synthesis and characterization of (Mo,Ti)n+1CnTx MXenes to understand their complex chemistry and to compare them with those of mono-metallic Mo2CTx and Ti3C2Tx. The potential of these materials as HER (hydrogen evolution reaction) catalysts is determined in alkaline medium. It is shown that Mo2Ti2C3Tx MXenes are a credible alternative to Mo2CTx MXenes since the surface properties of both MXenes are similar while their composition is quite different. Indeed, (Mo,Ti)n+1CnTx require lower temperatures and shorter time for the synthesis than for Mo2CTx, a great advantage from an industrial point of view. Finally, some avenues for improving the performance of MXenes for HER will also be described.

O 61.5 Wed 16:30 A 053

MXenes as support for transition metal oxides electrocatalysts for water splitting application — •Axel Zuber[1], Zdeněk Sofer[2], and Michelle Browne[1] — [1]CE-NESD, Helmholtz Zentrum Berlin für Materialien und Energie, Berlin (Federal Republic of Germany) — [2]Department of Inorganic Chemistry, University of Chemistry and Technology, Prague (Czeck Republic)

To reduce the cost of energy conversion and tend towards a greener energy production, research has been trying to use first-row transition metal-based catalysts for water splitting. Despite their good activity for the oxygen evolution reaction, transition metal oxides (TMOs) performance is hindered by their low conductivity and instability under potential. To improve it, recent works have successfully combined them chemically with conductive 2D transition metal carbides and nitrides (MXenes). The rich chemistry, large surface area and conductivity of MXenes make them excellent candidates as electrocatalyst supports, but they are also keen to oxidize in water which induces a loss of these properties. In this study, different MXenes were combined with transition metal oxides and tested as electrodes for the oxygen evolution reaction. The stability of the MXene structure as well as the resulting compound performance for the oxygen evolution reaction were investigated. The electrode materials remained stable and allowed to improve the oxygen evolution reaction overpotential of the transition metal oxide.

O 61.6 Wed 16:45 A 053

Infrared and Raman spectroscopic analysis of functionalized graphene and Mxene layers — •Karsten Hinrichs[1], Mailis Lounasvuori[2], Fatima Akhtar[2], Namrata Sharma[2], Tristan Petit[2], and Jörg Rappich[2] — [1]Leibniz-Institut für Analytische Wissenschaften - ISAS e.V., Application Laboratories Berlin, Schwarzschildstraße 8, 12489 Berlin, Germany — [2]Helmholtz-Zentrum Berlin für Materialien und Energie GmbH 14109 Berlin, Germany

Functionalized 2D-conductive materials like graphene and Mxene layers are interesting templates for catalysis but are also important for building of optical and electro-chemical sensors. Spectroscopic Raman and infrared (IR) ellipsometry analysis can access complementary information on the conductive material but also the ultrathin functional organic layer, respectively. Thereby bands due to molecular vibrations and phonons as well as free carrier absorptions are related to chemical and structural material properties. We acknowledge financial support by the European Union through EFRE 1.8/13.

O 62: Poster: SrTiO$_3$: A Versatile Material from Bulk Quantum Paraelectric to 2D Superconductor (joint session TT/KFM/MA/O)

Strontium titanate (SrTiO$_3$) is a paradigmatic material that plays an important role in various fields of solid-state physics, surface science and catalysis: The pure bulk phase is a wide-band-gap semiconductor that upon cooling becomes a textbook quantum paraelectric. When slightly doped, SrTiO$_3$ turns into a Fermi-liquid-type metal that becomes superconducting at extremely low charge carrier density. SrTiO$_3$-based surfaces and interfaces host un-conventional electronic states such as quasi-two-dimensional electron liquid, magnetism and superconductivity. Despite intensive studies over the past decades, SrTiO$_3$ continues to reveal surprising new phenomena that challenge the established views on this material. To this end achieving light-induced nonequilibrium states and the recent preparation of a 2D oxide based on SrTiO$_3$ opens new playgrounds for research. This Focus Session will present exciting developments in the study of electronic states that are based on the peculiar properties of SrTiO$_3$.

Please note that this Focus Session comprises four parts: Posters are presented within the TT poster session TT58, Wed 15:00-18:00, poster area E. Invited talks are compiled in the session TT62 (Thursday, 9:30 to 12:45, H0104), Contributed talks will be presented in sessions TT72 (Thursday 15:00-18:00, H0104) and TT83 (Fri 9:30-12:30, H0104).

Organizers: Rossitza Pentcheva, University of Duisburg-Essen, Marc Scheffler, University of Stuttgart

Time: Wednesday 15:00–18:00 Location: Poster E

O 62.1 Wed 15:00 Poster E

Optical conductivity of superconducting Nb:SrTiO$_3$ in magnetic fields at GHz frequencies — •Cenk Beydeda[1], Markus Thiemann[1], Martin Dressel[1], Hans Boschker[2], Jochen Mannhart[2], and Marc Scheffler[1] — [1]1. Physikalisches Institut, Universität Stuttgart — [2]Max-Planck-Institut für Festkörperforschung, Stuttgart

Doped SrTiO$_3$ was among the first unconventional superconductors, the application of the BCS theory is questionable due to the small Fermi energy. Here we present the optical conductivity ($1-30\,\mathrm{GHz}$) of superconducting Nb:SrTiO$_3$ in magnetic field. We observe features typical of an s-wave single-gap dirty type II superconductor. We attribute a kink in the magnetic field dependence to 2 distinct superconducting bands. We observe values of the real part of the optical conductivity exceeding the normal state value multiple times for rising magnetic field. Excessive losses at low frequency $hf \ll 2\Delta$ in dependence of temperature are a known feature of superconductivity and a result of coherence effects of the Cooper pairs in the superconducting state (coherence peak). The excessive losses we observe with rising magnetic field are substantially different from the coherence peak, especially in magnetic field dependence and absolute values. As far as we know, excessive losses of this type were not observed in any other superconductor. It is not clear whether Nb:SrTiO$_3$ is the only material that can show excessive losses of this type. We present an interpretation of our data in terms of Caroli-de Gennes-Matricon modes in the vortex state, reproducing the effect of excessive losses qualitatively.

O 62.2 Wed 15:00 Poster E

Ultrafast second harmonic generation spectroscopy of SrTiO$_3$ surfaces and interfaces — Mahendra Kabbinahithlu, Newsha Vesalimahmoud, Tobias Lojewski, Ping Zhou, Katharina Ollefs, and •Andrea Eschenlohr — Faculty of Physics and CENIDE, University Duisburg-Essen, Lotharstr. 1, 47057 Duisburg, Germany

Perovskite oxide heterostructures can exhibit properties at their interfaces that are very different from the bulk, for example a two-dimensional electron gas [1]. These properties emerge from charge carrier localization or charge transfer, which motivates an interface-sensitive analysis of the charge configuration and charge carrier dynamics. Second harmonic generation (SHG) spectroscopy is an interface-sensitive probe in centrosymmetric materials, suitable for the investigation of SrTiO$_3$-based heterostructures [2]. We perform pump-probe SHG spectroscopy with $<$ 30 fs time resolution in the visible wavelength range (1.9-2.5 eV) at SrTiO$_3$(001) surfaces as well as LaTiO$_3$/SrTiO$_3$ heterostructures, and discuss the polarization-, wavelength- and time-dependence of the observed SHG response.

[1] H. Y. Hwang, Y. Iwasa, M. Kawasaki, B. Keimer, N. Nagaosa, Y. Tokura, Nat. Mater. **11**, 103 (2012).

[2] A. Rubano, D. Paparo, Materials **16**, 4337 (2023).

O 62.3 Wed 15:00 Poster E

Low-temperature GHz response of quantum paraelectrics SrTiO$_3$ and KTaO$_3$ — Vincent T. Engl, Nikolaj G. Ebensperger, Cenk Beydeda, Lars Wendel, Marius Tochtermann, Ilenia Neureuther, Ishan Sarvaiya, Martin Dressel, and •Marc Scheffler — 1. Physikalisches Institut, Universität Stuttgart, Stuttgart, Germany

The low-temperature dielectric properties of SrTiO$_3$ and KTaO$_3$ are characteristic of their quantum paraelectric nature: upon cooling, the real part ϵ_1 of the dielectric function strongly increases, but eventually levels off at high values of ≈ 20000 for SrTiO$_3$ and ≈ 4000 for KTaO$_3$. In particular for SrTiO$_3$ it is very demanding to combine such dielectric bulk material with conventional GHz circuitry. We present superconducting coplanar Nb resonators on SrTiO$_3$ and KTaO$_3$ substrates, where in the case of SrTiO$_3$ we employ a distant flip chip geometry. Taking advantage of several resonator modes, we determine the dielectric properties of the two materials at frequencies around 1 GHz and at temperatures down to 25 mK. We thus access regimes of frequency and temperatures, where the dielectric properties of SrTiO$_3$ and KTaO$_3$ have barely been studied.

For the case of SrTiO$_3$, we find an unexpected temperature dependence of the real part ϵ_1 of the dielectric constant: at temperatures below 5 K, where ϵ_1 is expected to vary little upon further cooling, we find a clear maximum around 3 K and a weak minimum around 200 mK. We also observe a strong suppression of microwave losses in both SrTiO$_3$ and KTaO$_3$ for temperatures down to the mK range.

O 62.4 Wed 15:00 Poster E

ferromagnetic two-dimensional electron gas in oxide interfaces — •Yu Chen[1], Maria D'Antuono[1,2], Martando Rath[1], Cinthia Piamonteze[3], Daniele Preziosi[4], Benoit Jouault[5], Daniela Stornaiuolo[1,2], and Marco Salluzzo[1] — [1]CNR-SPIN, Napoli, Italy — [2]Università di Napoli "Federico II", Italy — [3]Photon Science Division, Paul Scherrer Institut, Switzerland — [4]Université de Strasbourg, CNRS, IPCMS UMR, France — [5]Laboratoire Charles Coulomb, UMR 5221, CNRS, Université de Montpellier, France

Interfacial inversion symmetry breaking triggers novel phenomena not observed in bulk materials, such as unconventional superconductivity and magnetism. Here, we report on the realization of ferromagnetic two-dimensional electron gas (2DEG) at (001) and (111) interfaces between LaAlO$_3$, EuTiO$_3$, and SrTiO$_3$. At variance with the octahedral and quasi-octahedral symmetry in bulk SrTiO$_3$ and (001) interface, trigonal crystal field is reconstructed at (111) interface. The experiments of transport, magnetic and x-ray spectroscopy indicate that the filling of Ti $3d$ bands in the EuTiO$_3$ layer and at the interface with SrTiO$_3$ induces an exchange interaction between Eu-$4f^7$ magnetic moments. We observe carrier density-dependent ferromagnetic correlations and anomalous Hall effect, sizable in-plane orbital moment possibly related to Ti-$3d$ electrons occupying bands with the main $3d_{xz,zy}$ and a_{1g} orbital characters at (001) and (111) interfaces, respectively. Our findings show intriguing interplay between ferromagnetism, spin-orbit coupling, and symmetry breaking at oxide 2DEG, serving as a guide for the materials design of advanced spintronics.

O 62.5 Wed 15:00 Poster E

Role of excitonic effects in optical and x-ray absorption spectroscopy of $SrTiO_3$: insights from a combined first principles and many-body theory approach — •V. Begum-Hudde[1], M. E. Gruner[2], and R. Pentcheva[2] — [1]University of Illinois Urbana-Champaign, USA. — [2]University of Duisburg-Essen, Duisburg, Germany.

We present a comprehensive study of the optical[1] and x-ray absorption spectrum[2] (XAS) in the paradigmatic oxide, $SrTiO_3$. Our results demonstrate that inclusion of the quasiparticle effects with single-shot G_0W_0 as well as the electron-hole (e-h), and electron-(core)hole interactions by solving the Bethe-Salpeter Equation (BSE) is integral to accurately describe both the valence and core electron excitations. For the optical spectra, the effect of the exchange-correlation functional is observed to progressively reduce from 1.5 eV variance in the onset of the spectrum in the independent particle picture to 0.3 eV upon inclusion of excitonic corrections. The Ti-$L_{2,3}$ XAS edge is concurrent with experiment w.r.t. the energetic positions of the four-peak structure which is characteristic of Ti octahedral coordination in $SrTiO_3$. We also analyze the origin of prominent peaks in the spectra and identify the orbital character of the relevant contributions by projecting the e-h coupling coefficients from the BSE eigenvectors on the band structure. The spatial distribution of the first bound exciton wave function of the O K edge exhibits an intriguing two-dimensional spread in the x-y plane despite the three-dimensional nature of the material.

[1] Phys. Rev. Mater. **3**, 065004 (2019)
[2] Phys. Rev. Res. **5**, 013199 (2023)

O 62.6 Wed 15:00 Poster E

Boosting the Edelstein effect of two-dimensional electron gases by ferromagnetic exchange — •Gabriel Lazrak[1], Annika Johansson[2], Börge Göbel[2,3], Ingrid Mertig[2,3], Agnès Barthélémy[1], and Manuel Bibès[1] — [1]Laboratoire Albert Fert, Université Paris-Saclay, CNRS, Thales, Palaiseau, FRANCE — [2]Max Planck Institute of Microstructure Physics, Halle, GERMANY — [3]Martin Luther University Halle-Wittenberg, Halle, GERMANY

In this work, we show that making STO 2DEGs ferromagnetic significantly boosts the conversion efficiency of charge and spin currents through direct and inverse Edelstein effects (EE/IEE). Starting from the experimental band structure of non-magnetic SrTiO3 2DEGs, we mimic magnetic exchange coupling by introducing an out-of-plane Zeeman term in a tight-binding model. We then calculate the band structure and spin textures for increasing internal magnetic fields and compute the Edelstein effect using a semiclassical Boltzmann approach. The conversion efficiency initially rises with magnetic field strength, reaching a maximum before declining. This behavior results from the interplay between exchange coupling and the effective Rashba interaction. Our experimental focus is on the 2DEG at the SrTiO3/EuO interface to introduce ferromagnetism into the system.

O 62.7 Wed 15:00 Poster E

Impact of a Si(001) substrate on the electronic reconstruction and two-dimensional electron gas formation at $LaTiO_3/SrTiO_3(001)$ — •Andri Darmawan and Rossitza Pentcheva — Department of Physics, University of Duisburg-Essen

The two-dimensional electron gas (2DEG) formed at oxide interfaces e.g. between the band insulator $SrTiO_3$ and the Mott insulator $LaTiO_3$ has attracted a lot of attention [1]. However, despite the high carrier density at the interface, the carrier mobility is lower compared to semiconductor materials. A strategy to overcome this shortcoming is the integration of the oxide system on a semiconductor substrate [2]. Based on density functional theory calculations with a Hubbard U term we modeled $LaTiO_3/SrTiO_3(001)$ with and without a Si(001) substrate. We explore systematically the sample geometry and the effect of the termination to Si(001) on the electronic reconstruction at the $LaTiO_3/SrTiO_3(001)$ interface. The comparison between the two systems indicates lower effective masses and consequently higher mobility of the 2DEG at $LaTiO_3/SrTiO_3/Si(001)$.

Funding by DFG within CRC1242 and computational time at the Leibniz Supercomputer Center (project pr87ro) are gratefully acknowledged.

[1] A. Ohtomo et al., Nature 419, 378 (2002)
[2] E. N. Jin et al., APL Mater. 2, 116109 (2014)

O 63: Poster DS (joint session DS/MM/O)

Time: Wednesday 17:00–19:00 Location: Poster B

O 63.1 Wed 17:00 Poster B

Tailoring Ti3C2 MXenes towards the Oxygen Evolution Reaction — •Aline Alencar Emerenciano and Michelle Browne — Helmholtz-Zentrum Berlin für Materialien und Energie, Berlin, Germany

Ti3C2 MXenes are promising materials for water splitting due to their high conductivity, hydrophilicity, and good mechanical properties when compared to other 2D-nanomaterials [1]. The possibility of tailoring MXenes in terms of functional groups, flake size/shape, presence of defects, and surface area makes them excellent candidates to be hybridized with transition metal oxides. MXenes/TMO catalysts can provide synergetic properties such as high conductivity in combination with active sites for Oxygen Evolution Reaction (OER). To reduce the mass loading by increasing the intrinsic activity and stability of MXenes, the surface functionalization and surface area must be controlled. In this work, in situ generated HF was utilized to remove Al-elements for the production of Ti3C2. Synthesis protocols utilizing different concentrations of HCl were investigated to evaluate the flake quality in terms of functional groups distribution, flake size towards the existence of multilayered and delaminated nanoflakes, as well as the concentration of pin-roles on their surface. Overall, this work provides a better understanding in how HCl concentration can influence the MXenes contribution in hybrid electrocatalysts considering functional groups distribution and presence of defects.

1.Zhang, C. J. et al. Adv. Mater. 29, 1-9 (2017).

O 63.2 Wed 17:00 Poster B

Chemical functionalisation of CuCo(2)-LDH on V(2)CT(x) for the oxygen evolution reaction — •Bastian Schmiedecke and Michelle Browne — Helmholtz-Zentrum Berlin, Berlin, Germany

MXenes, known for their exceptional surface area and high conductivity, serve as excellent catalyst supports, enhancing electrocatalytic performance in the oxygen evolution reaction (OER). Layered double hydroxide (LDH) materials, though promising for the OER, lack conductivity to ensure easy charge transfer during electrochemical processes. While, research on LDH materials has developed rapidly, there remains a high demand for refining the strategic combination of LDH with two dimensional (2D) materials, such as MXenes, which can significantly improve the water oxidation performance of LDH materials by inducing high conductivity, hydrophilicity and surface area.

This study introduces a hybrid catalyst, CuCo2-LDH grown on delaminated vanadium carbide (V(2)CT(x)) nanosheets to enhance the OER performance. The synthesized CuCo(2)-LDH@V(2)CT(x) electrocatalyst exhibited excellent activity with an overpotential of 289 mV at a catalytic current density of 10 mA cm-2 with a Tafel slope value of 74 mV dec-1. Furthermore, slight performance improvements were observed after 12 h of continuous operation. We propose that the enhanced performance is attributed to the conductivity of V(2)CT(x) and its synergistic interaction with CuCo(2)-LDH, effectively minimizing aggregation, exposing more active sites. This work demonstrates the significant potential of combining LDH-based nanomaterials with V(2)CT(x) MXene for energy conversion applications.

O 63.3 Wed 17:00 Poster B

Analysis of polarization dependent IR spectra of thin films — •Karsten Hinrichs[1], Andreas Furchner[2], Fatima Akhtar[2], Norbert H. Nickel[2], and Jörg Rappich[2] — [1]Leibniz-Institut für Analytische Wissenschaften - ISAS e.V., Application Laboratories Berlin, Schwarzschildstraße 8, 12489 Berlin, Germany — [2]Helmholtz-Zentrum Berlin für Materialien und Energie GmbH 14109 Berlin, Germany

The interpretation of bands in polarization dependent infrared (IR) spectra of thin films is addressed. Several examples such as an isotropic polymer film and functionalized silicon surfaces will be discussed as case studies. It will be shown that the observed vibrational band properties depend on the measurement geometry, the film thickness, the materials dielectric functions, as well as the direction of the probing electromagnetic fields. Comparative Density Functional Perturbation Theory (DPFT) - calculations are used for analysis. The financial support by the Europäischer Fonds für regionale Entwicklung by EFRE 1.8/13 and ProFIT 10185407 as well as the Federal Ministry of Education and Research and the project CatLab (03EW0015A/B) is acknowledged.

O 63.4 Wed 17:00 Poster B

Focused-Ion-Beam induced Defect Emission in Hexagonal Boron Nitride — •Felix Schaumburg, Dominik Kaczmarek, David Plitt, Martin Geller, Günther Prinz, and Axel Lorke — Faculty of Physics, University of Duisburg-Essen and CENIDE, Germany

Defects in the wide bandgap material hexagonal boron nitride (hBN) exhibit single-photon emission at room temperature (RT) [1]. We used the gallium ion beam of a focused ion beam (FIB) to generate ensembles of defects that we associated with boron vacancies having a photoluminescence emission exceeding 800 nm [2]. After irradiation isolated point defects can also be found next to the irradiated areas. We show that our created defects exhibit a bright RT emission, an almost sublinear power dependence commonly associated with a two-level system and an increasing signal, a decreasing full with at half maximum (FWHM), and a minimal shift to lower wavelengths at lower temperatures. Our results show, that we can generate bright emitters in hBN by Ga-ion treatment that are in a wavelength range of up to 800 nm. Our next goals will be contacting these emitters by putting a gate structure on top of it, as well as creating single defects at higher wavelengths. [1] F. Hayee et.al., Nat.Mater.19 (2020) [2] C. Qian et.al., arXiv (2022)

O 63.5 Wed 17:00 Poster B

Optimizing electrical transport and SAW propagation in molybdenum disulfide — •Noah Spitzner[1], Pai Zhao[1], Renrong Liang[2], Chithra Sharma[1], Lars Tiemann[1], and Robert Blick[1] — [1]Center of Hybrid Nanostructures, University of Hamburg, Luruper Chaussee 149, 22761 Hamburg — [2]School of Integrated Circuits, Tsinghua University, 100083 Beijing, China

Few-layered and monolayer MoS$_2$ has gained increasing significance in recent years, due to its large band gap of up to 1.9 eV at the K and K$'$ valley of the hexagonal Brillouin zone. Surface acoustic waves (SAW) and transport measurements allow non destructive probing of the physics governing the material.

In this work we exfoliated MoS$_2$ flakes of a few layers onto a specially tailored substrate with LiNbO$_3$ as piezoelectric top layer. We can pass surface acoustic waves through the MoS$_2$ flake via an interdigitated transducer (IDT) electrode configuration and study the electrical response. To facilitate electron transport at low temperatures, liquid nitrogen physical vapor deposition (LNPVD) was utilized to deposit the contact metals. The cooled deposition reduces Fermi level pinning (FLP) in the contact interface and empowers us to measure longitudinal and transversal voltages with better contact quality.

Under acoustic excitation at 4.2 K, we observed acoustic currents and voltages in MoS$_2$ that depend on the power and frequency of the SAW. Hence, we were able to observe the acoustoelectric and acoustogalvanic effect. We also studied magnetotransport under perpendicular magnetic fields and the weak localization phenomenon.

O 63.6 Wed 17:00 Poster B

Creating realistic carbon nanomembranes using molecular dynamics model simulations — •Levin Mihlan[1], Anna Niggas[2], Filip Vukovic[2], Jürgen Schnack[1], and Richard A. Wilhelm[2] — [1]Universität Bielefeld, Deutschland — [2]TU Wien, Österreich

1nm thin carbon nanomembranes (CNMs) are synthesized from aromatic self-assembled monolayers (SAMs) by electron-induced crosslinking and supposedly of irregular internal structure, which renders standard spectroscopic characterization very difficult [1]. However MD simulations can offer insights into CNMs' internal structure. Recently, it was shown that the neutralisation dynamics of highly charged ions (HCIs) transmitting through thin materials are very sensitive to the material structure [2]. Hence, HCI spectroscopy may, together with accompanying simulations using a time dependent potential [3], be used to characterize CNMs. In order to obtain model membranes whose mechanical and spectral properties , fit to those of manufactured CNMs, a model process starting from a SAM is implemented as an MD simulation. Structures generated this way can be compared to alternative model structures, which are created by incorporating experimental mechanical properties as input parameters. This approach helps to gain a better understanding of the internal structure of CNMs.

[1] Dementyev et al. ChemPhysChem 21.10 1006 (2020)

[2] Wilhelm, Richard A. Surf.Sci.Rep. Vol 77 Issue 4 (2022)

[3] Wilhelm, Richard A; Grande, Pedro L.Commun.Phys.2,89 (2019)

O 63.7 Wed 17:00 Poster B

Rolle of Collective Behavior of Water Molecules in Robust Ferroelectricity in Graphene Nanoribbons — •Igor Stanković[1], M. Awais Aslam[2], and Aleksandar Matković[2] — [1]Scientific Computing Laboratory, Center for the Study of Complex Systems, Institute of Physics Belgrade, University of Belgrade, 11080 Belgrade, Serbia — [2]Institute of Physics, Montanuniversität Leoben, Franz Josef Strasse 18, 8700 Leoben, Austria

An understanding of the water dynamics on the edges of one and two-dimensional structures is scarce. Still, such interactions can be sufficient to perturb local electric environments, therefore offering an opportunity to harness the effect of the local dipole moment of water. Combining experiments on nanoribbon field effect transistors and molecular dynamic simulations, we elucidate a collective behaviour of water within clusters adsorbed on graphene edges. We show that these nanoribbons exhibit significant and persistent remanent fields which can be employed in ferroelectric heterostructures and neuromorphic circuits.

References [1] M. A. Aslam et al, https://doi.org/10.48550 arXiv.2304.09738

O 63.8 Wed 17:00 Poster B

Experimental setup for gas sensing with TMD based field-effect devices — •Axel Printschler[1], Emad Najafidehaghani[1], Antony George[1], Hamid Reza Rasouli[1], David Kaiser[1], Uwe Hübner[2], and Andrey Turchanin[1] — [1]Friedrich Schiller University Jena, Institute of Physical Chemistry, Jena — [2]Leibniz Institute of Photonic Technology (IPHT), Jena

Sensing devices based on 2D transition metal dichalcogenides (TMDs) such as MoS$_2$, WS$_2$, MoSe$_2$, WSe$_2$, *etc.* have attracted significant research interest, as their electronic properties are greatly influenced by variations in the environment, *e.g.*, due to formation of adsorbates. This influence is reflected in changes of the device's electric transport characteristics, which can be precisely measured and used for gas sensing. As toxic gases pose a threat in many fields, there is a high demand in sensitive, selective and flexible gas sensors that also work at low power. Electronic sensors based on TMDs can satisfy this need while being atomically thin and flexible. Here we present an experimental setup and first results on gas sensing with field effect transistors (FETs) fabricated from TMD monolayers grown by chemical vapor deposition (CVD).

O 63.9 Wed 17:00 Poster B

MOCVD synthesis of MoS2 and WS2 combination 2D heterostructures — •Nikolas Dominik, Sebastian Klenk, Florian Herdl, Cormac Ó Coileáin, Tanja Stimpel-Lindner, and Georg S. Duesberg — Institute of Physics, University of the Bundeswehr Munich & SENS Research Center, 85577 Neubiberg, Germany

Two-dimensional (2D) materials (such as MoS2 and WS2) are materials layered at the atomic scale. This gives them exceptional electrical, mechanical and optical properties, and makes them interesting for electronics, photovoltaics and sensing [1]. Van der Waals heterostructures composed of 2D materials expand on the possible range of properties and so have attracted extensive focus due to factors such as smooth heterostructure interfaces, ultrafast carrier transport, and high bandgap tunability [2].

Here we present the synthesis of MoS2/WS2 combination heterostructures via metal-organic chemical vapour deposition (MOCVD) using a high controllable industrial scale multi-precursor system, and show the clearly defined stacked nature of the films produced. We explore the growth parameter space using Raman and X-ray photoemission spectroscopy, and microscopy techniques. We complement our characterisation by examining the influences on the band structure of the layered material.

[1] Q. H. Wang et al., Nature Nanotech 7, 699-712 (2012)

[2] W. Xia et al., Nanoscale 9, 4324-4365 (2017)

O 63.10 Wed 17:00 Poster B

RF-sputtering of Nb and NbN thin films for quantum transport studies — •Peer Heydolph, Vincent Strenzke, Isa Moch, Annika Weber, Lars Tiemann, and Robert Blick — Center for Hybrid Nanostructures (CHyN), Universität Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany

Niobium plays a vital role in research and applications, contributing to advancements in superconductivity, quantum computing, and high-performance electronics. Here, we investigate the impact and interdependence of growth parameters on the properties of radio-frequency (RF)-sputtered niobium (Nb) and niobium nitride (NbN) thin films with a high critical temperature for applications in nanostructures and quantum transport studies at 4.2 Kelvin. We demonstrate that high quality superconducting films can be consistently produced even in a basic sputtering setup. We systematically varied the growth parameters such as the power of the RF-plasma and the flow of argon and nitrogen and survey their effects and interdependence on the properties of the thin films. The quality of the resulting films was characterized via X-ray diffraction (XRD), scanning electron microscopy (SEM) and profilometer measurements to gauge the film thickness. For electrical characterization at various temperatures and magnetic fields we employed a PPMS cryostat. We found that lower flow rates or RF powers do not necessarily lead to a higher critical temperature for NbN, which is in agreement with previous publications. Furthermore, it is crucial to finely tune all parameters and take into account their interdependencies.

O 63.11 Wed 17:00 Poster B

Poly(neutral red) as a Possible Electrode Material for Electrochemical Cells — •Lisa Rohovsky[1], Daniel Holzhacker[1], Tsukasa Yoshida[2], and Derck Schlettwein[1] — [1]Institut für Angewandte Physik, Justus-Liebig-Universität Gießen — [2]Department of Organic Materials Science, Graduate School of Organic Materials Science, Yamagata University

Poly(neutral red) (PNR) is used in sensors owing to its redox characteristics and high electrical conductivity. PNR is also easy to prepare and inexpensive. Therefore, it might also be of interest as an electrode material in electrochemical cells like, e.g., electrochromic cells, batteries, or dye-sensitized solar cells (DSSCs). In the latter case, it could even help to replace platinum as an electrode material, which is expensive and not readily available. In this study, thin films of PNR were prepared by an established electrosynthetic procedure by oxidation of neutral red by cyclic voltammetry (CV) in an aqueous solution.[1] Samples were produced for varied number of oxidative cycles and then analysed for their layer thickness and absorptance. As expected, the layer thickness increased linearly with the number of cycles, as did the absorptance. The reversible electrochemical reduction and reoxidation of aqueous solutions of ferro/ferricyanide in contact to the prepared films was studied as a model electrolyte. Results and implications for the applicability of such PNR electrodes will be discussed. [1]. Y. Harada, D. Kono, P. Stadler, T. Yoshida, SPAST Abstracts ,1(01), First International Conference on Technologies for Smart Green Connected Society 2021.

O 63.12 Wed 17:00 Poster B

Growth of Sc(x)Ga(1-x)N on 6H-SiC by plasma assisted molecular beam epitaxy — •Fabian Ullmann[1,2], Abdul Qadir Shahbaz[1,2], and Stefan Krischok[1,2] — [1]TU Ilmenau, Ehrenbergstraße 29, 98693 Ilmenau — [2]Zentrum für Mikro- und Nanotechnologie, Gustav-Kirchhoff-Straße 7, 98693 Ilmenau

ScGaN can occur in various crystal orientations. The most important are wurtzite and rock salt formation. Depending on the scandium concentration, a phase transition can be found between these orientations. Plasma-assisted molecular beam epitaxy (PAMBE) in combination with reflective high-energy electron diffraction (RHEED) was performed to create layers with different scandium concentrations in ScGaN. To determine the concentration of the grown layers, X-ray photoelectron spectroscopy was used in the same vacuum chamber. In addition, the surfaces were analyzed using atomic force microscopy (AFM, in-situ) and scanning electron microscopy (SEM) to obtain information on the morphology of the surfaces and to confirm the gained crystal orientations X-ray diffraction (XRD) were performed.

O 63.13 Wed 17:00 Poster B

Optical and morphological properties of thin Nb2O5 layers deposited via High Power Impulse Magnetron Sputtering — •Christof Zickenheiner[1], Raul Ramos[2], Viktor Udachin[1], Wolfgang Maus-Friedrichs[1], and José R.R. Bortoleto[2] — [1]Clausthal University of Technology, Clausthal-Zellerfeld, Germany — [2]Instituto de Ciencia e Tecnologia, Sorocaba, Brazil

To address climate challenges and sustainable development, the UN set ambitious goals. Renewable energy, particularly solar, plays a key role. Emerging technologies like perovskite solar cells (PSC) offer cost-effective production, competing with silicon-based counterparts. This study focuses on the electron transport layer in PSC using Nb_2O_5 thin films deposited on glass substrates via HiPIMS from an Nb-target in an Ar/O_2 atmosphere at 25 °C. Pulse duration and oxygen pressure were varied as experimental parameters. Optical emission spectroscopy (OES) revealed the presence of oxygen species within a plasma and excitation of sputtered metal species for the HiPIMS process. The examination of the morphological properties via profilometry and contact angle analysis showed an influence of the experimental parameters on roughness. That is, shorter pulses as well as lower oxygen pressure result in smoother layer growth with a lower deposition rate. The investigation of optical properties via UV/Vis-Spectroscopy showed no significant impact of pulse duration. It became evident that lower oxygen pressure reduces the incorporation of oxygen into the growing layer. This results in a changed phase composition, with the formation of NbO_2, which in turn affects optical and electrical properties.

O 63.14 Wed 17:00 Poster B

An attempt to predict oligomer sputtering using binary collision approximation simulations — •Hans Hofsäss, Felix Junge, and Patrick Kirscht — II. Physikalisches Institut, Universität Göttingen, Germany

The binary collision approximation (BCA) program IMINTDYN [1] allows a prediction of ion solid interactions. For sputtering of carbon and SiO2 experimental sputter yields are significantly higher than yields from BCA simulations. SDTrimSP simulations [2] reproduce experimental sputter yields by adjusting the surface binding energies. For O atoms 1 eV instead of the elemental sublimation energy of 2.58 eV and for carbon 4.5 eV instead of 7.4 eV is used. For sputtering of carbon it was shown [3] that sputtering of oligomers and clusters is relevant. We introduce a model to simulate oligomer sputtering using the IMINTDYN program based solely on thermodynamic formation enthalpies. In particular sputtering of O2 and SiO dimers and carbon oligomers is energetically favorable. To predict the oligomer sputter fraction, we use Boltzmann factors based on the ratios of oligomer and monomer formation enthalpies. We show that we can quantitatively predict the carbon and SiO2 experimental sputter yields.

[1] H. Hofsäss, A. Stegmaier, Nucl. Instr. Meth B 517 (2022) 49

[2] A. Mutzke, R. Schneider, W. Eckstein, R. Dohmen, K. Schmid, U. von Toussaint, G. Bandelow, SDTrimSP Version 6.00, MPI Plasma Physics, report IPP 2019-02 (2019)

[3] E.Oyarzabal, R.P. Doerner, M. Shimada, G.R. Tynan, J. Appl. Phys. 104 (2008) 043304

O 63.15 Wed 17:00 Poster B

Growth of Antimony thin films on c-plane Sapphire — •Jonathan Spelsberg, Alexander Fuhrich, and Martin Salinga — Institut für Materialphysik, Universität Münster, Wilhelm-Klemm-Str. 10, 48149 Münster

Antimony has been demonstrated to function as a single-element phase change material for electronic memory applications [1]. Confinement into nanostructures is crucial for stabilizing the amorphous phase of this material that is otherwise known for its fast crystallization kinetics. Yet, the exact influence of interfacing dielectrics on the physical properties of Antimony requires a fundamental investigation. To this end, samples with a well-controlled interface are required. Here, we report about the growth of Antimony thin films on c-plane Sapphire by molecular beam epitaxy. Based on characterization with RHEED and AFM, we demonstrate the impact of substrate preparation and process temperatures on the growth of 3 nm to 30 nm thick antimony films. Moreover, we discuss the relevance of Antimony cluster size and the effective use of a cracker effusion cell.

[1] M.Salinga et al., Monatomic phase change memory, Nature Materials 17, p. 681-685 (2018)

O 63.16 Wed 17:00 Poster B

Novel nanofabrication facility for ultra-clean samples — •Alexander Fuhrich and Martin Salinga — Institut für Materialphysik, Universität Münster, Wilhelm-Klemm-Str. 10, 48149 Münster

We present the capabilities of a novel nanofabrication facility dedicated to the fabrication of nanostructures under ultra-clean conditions. In addition to a UHV cluster for MBE growth, the system comprises a lithography unit in an inert argon atmosphere. Air- and water-sensitive samples, like materials for memristive switches, can be structured using thermal scanning probe lithography with a lateral resolution down to less than 30 nm. Samples can be analyzed in-situ using RT-STM and RHEED. A self-sufficient UHV suitcase is used to characterize samples in other setups, such as femtosecond-laser pump probe spectroscopy, without ever exposing our samples to air.

O 63.17 Wed 17:00 Poster B

Preparation and Characterization of Mixed Electrodes of WO_3 and MoO_3 as Electrochromic Coatings — •Smail Mekhilef, Thi Hai Quyen Nguyen, and Derck Schlettwein — Institut für Angewandte Physik, Justus-Liebig-Universität Gießen

Electrochromic coatings can modify the transmittance for ambient light. As smart windows for buildings, relevant contributions can be expected for the energy-efficiency of buildings. Sputter- deposited WO_3 is typically used. Sustainability can be increased if low-temperature processes are established that would, further, allow the use of low-impact and low-weight polymer substrates. For large-scale applications, one may want to avoid W as a critical element and, at least partly, replace it by more abundant Mo. In this study, we extended established wet-chemical methods to prepare porous thin film electrodes of WO_3 [1] towards the processing of mixed oxides of W and Mo. Thin films of different W/Mo were prepared and characterized by scanning electron microscopy, atomic force microscopy and, mainly, by spectroelectrochemical measurements in contact to an inert organic electrolyte at either constant or swept electrode potential and the results will be discussed. [1]. T. H. Q. Nguyen, F. Eberheim, S. Göbel, P. Cop, M. Eckert, T. P. Schneider, L. Gümbel, B. M. Smarsly, D. Schlettwein, Enhancing the Spectroelectrochemical Performance of WO_3 Films by Use of Structure-Directing Agents during Film Growth, Appl. Sci. 2022, 12, 2327.

O 63.18 Wed 17:00 Poster B

Deposition of reduced ceria thin films by reactive magnetron sputtering for the development of a resistive gas sensor — •Paul-G. Nitsch[1], Markus Ratzke[1], Emilia Pozarowska[2], Jan I. Flege[2], Carlos Alvarado Chavarin[3], Christian Wenger[3], and Inga A. Fischer[1] — [1]Experimentalphysik und funktionale Materialien, BTU-CS, Cottbus, Germany — [2]Angewandte Physik und Halbleiterspektroskopie, BTU-CS, Cottbus, Germany — [3]IHP - Leibniz-Institut für innovative Mikroelektronik, Frankfurt (Oder), Germany

The use of cerium oxide for hydrogen sensing is limited by the low electrical conductivity of layers deposited from a ceria target. To increase the electrical conductivity, partially reduced cerium oxide layers were obtained from a metallic cerium target by reactive magnetron sputtering. The proportions of the oxidation states Ce3+, present in reduced species, and Ce4+, present in fully oxidized species, were determined by ex-situ XPS. For electrical characterization, films were deposited on planarized tungsten finger electrodes. IV curves were measured over several days to investigate possible influences of oxygen and humidity on electrical conductivity. The morphological stability of the layers under ambient conditions was investigated by microscopical methods. The XPS results show a significant amount of Ce3+ in the layers. The electrical conductivity of as-grown samples is several orders of magnitude higher than that of samples grown from a ceria target. However, the conductivity decreases over time, indicating an oxidation of the layers. The surface morphology of the samples was found to be changing drastically within days, leading to partial delamination.

O 63.19 Wed 17:00 Poster B

Modification of the optical and electrical properties of AZO thin films for variety of applications — •Maria Stefanova[1], Dimitrina Petrova[1,2], Blagovest Napoleonov[1], Stefani Bogoeva[1], Vladimira Videva[1,3], Velichka Strijkova[1], Vera Marinova[1], and Dimitre Dimitrov[1,4] — [1]Institute of Optical Materials and Technologies-BAS Sofia, Bulgaria — [2]South-West University "Neofit Rilski", Blagoevgrad, Bulgaria — [3]Sofia University, Sofia, Bulgaria — [4]Institute of Solid State Physics, Bulgarian Academy of Sciences, Sofia, Bulgaria

Here we report on the deposition of Aluminium Doped Zinc Oxide (AZO) thin films on sapphire substrates (AZO/Sapphire) by using ALD method and the influence of postdeposition UV-Ozone treatment on the films' properties. XRD revealed a polycrystalline wurtzite structure. The influence of UV-Ozone treatment on surface morphology, electrical and optical properties of AZO/Sapphire was investigated. It was found that UV-Ozone treatment improves the electrical and optical properties while did not cause significant changes to the polycrystalline structure and surface morphology of the AZO films which opens potential for various practical applications.

Acknowledgements This work is supported by the Bulgarian National Science Fund under the grant number KP-06-N-68/1 and the Research equipment of distributed research infrastructure INFRAMAT (part of Bulgarian National roadmap for research infrastructures) supported by Bulgarian Ministry of Education and Science.

O 63.20 Wed 17:00 Poster B

Transient Photoluminescence of Lead Halide Perovskites Beyond Lifetimes: Non-excitonic Geminate Pairs and Carrier Transport — •Hannes Hempel[1], Martin Stolterfoht[2], and Thomas Unold[1] — [1]Helmholtz Zentrum Berlin, Germany — [2]Chinese University of Hong Kong, China

Transient photoluminescence (trPL) is the standard technique to quantify the lifetimes of photogenerated charge carriers in energy conversion materials. However, trPL decays are sensitive to processes other than only carrier recombination. We show that the initial trPL transients of lead halide perovskite thin films on glass are dominated by charge transport, namely hot carrier transport, spread-out of non-excitonic gemmate pairs, and redistribution. Including these processes in the modeling of trPL yields the doping and intrinsic carrier concentration, the mobility and diffusion coefficient, the hot carrier transport length, and the radiative and non-radiative lifetimes. Further, we calibrate trPL to absolute photon numbers, which allows quantifying the transient quasi-Fermi-level splitting and reveals fundamental energy losses in photo absorbers such as the increase of entropy by loss of geminate correlation and redistribution. The presented analysis is crucial for the appropriate interpretation of trPL and yields almost all optoelectronic properties relevant for application as a photo-absorber in solar cells.

O 63.21 Wed 17:00 Poster B

Ultrafast Lattice Dynamics in Epitaxially Grown Bismuth Thin Films — •Timo Veslin[1], Felix Hoff[1], Jonathan Frank[1], Abdur Rehman Jalil[3], Julian Mertens[1], and Matthias Wuttig[1,2,3] — [1]I.Institute of Physics (IA) RWTH Aachen University — [2]Jülich-Aachen Research Alliance (JARA FIT and JARA HPC) — [3]PGI 10 (Green IT), Forschungszentrum Jülich GmbH

Femtosecond optical pump probe measurements are carried out in order to detect changes in the reflectivity of the material response of epitaxially grown bismuth thin films on sub-ps timescales. Examination of reflectivity changes due to the coherent phonon response provides insight into the ultrafast lattice dynamics and relaxation of bismuth thin films. The pronounced thickness dependent behavior of bismuth is analysed by coherent phonons within the framework of displacive and impulsive excitation models. Here, we show how the competition of phonon softening due to different laser fluencies and phonon hardening from confinement effects can be understood. Raman measurements are carried out to help to disentangle both effects. XRD measurements are shown to help to understand the structural change of the lattice structure. These combined measurements will help to understand confinement and fluency effects in a single model and shows how to tune bonding properties in this unconventional class of materials.

O 63.22 Wed 17:00 Poster B

Atomic-Scale Insights Into The Interlayer Characteristics of Thin-Layered Materials Using Ultra-high Vacuum Tip-Enhanced Raman Spectroscopy — •Soumyajit Rajak and Nan Jiang — University of Illinois Chicago, Chicago, USA

Optoelectronic properties of molecular thin films are controlled by the local nanostructures of a molecular arrangement. Probing the effect of the local environment of nanostructures is challenging because the spatial resolution of conventional optical spectroscopic techniques is limited by the diffraction limit of light. Coupling light with plasmonic nano-objects creates highly localized surface plasmons (LSPs), which allows us to break the diffraction limit. Herein we present a combined topographical and optical analysis of different surface-sensitive arrangements of molecules and 2D material heterostructures using angstrom-scale resolution scanning tunneling microscopy (STM) and ultra-high vacuum tip-enhanced Raman spectroscopy (UHV-TERS). TERS uses the apex of an STM tip made of a plasmonic metal to couple light to the near field. The Raman modes of the nanostructure underneath this tip are enhanced by the nano-confined surface plasmons which allows us to obtain chemical information with Angstrom scale spatial resolution. STM images combined with localized surface plasmon resonance-enhanced Raman signals reveal different adsorbate configurations of single molecule entities and a fundamental view of interfacial interactions. The atomic scale insights obtained into the local environment enable precise control over the fabrication of nanostructures with tailored optoelectronic properties.

O 63.23 Wed 17:00 Poster B

In-situ Study of Surface Band-Bending in c-ZnO and its Effect on the Excitonic Dielectric Function — •Luis Rosillo Orozco[1], Kurt Hingerl[1], and Christoph Cobet[1,2] — [1]Zentrum für Oberflächen und Nanoanalytik, Johannes Kepler Universität, Linz, Austria — [2]Linz School of Education, Johannes Kepler Universität, Linz, Austria

Semiconductors in air and in vacuum often have a band-bending near the surface caused by surface states capturing bulk charges or simply due to natural polarization in the case of polar materials, as is the case of Zinc Oxide. Nevertheless, when semiconductors are in contact with an electrolyte we can intentionally produce a surface dipole and create a space charge region (SCR) by adding another electrode in the solution and applying a voltage between the two.

Space charge regions have a big impact on the optical and electrical properties of semiconductors and, of course, semiconductor devices. Therefore, it is of high interest to understand the effects caused by them.

We present in-situ spectroscopic ellipsometry (SE) combined with electrochemical techniques to study the response of the discrete excitons and exciton-phonon complexes (EPC) to the inner electric fields produced near the semiconductor surface. Using mono-chromatic transients we are able to identify the flat-band potential. A semi-empirical optical model is developed to study the contribution of the surface band-bending to the total dielectic function for a range between 3.2 e.V. to 3.6 e.V.

O 63.24 Wed 17:00 Poster B

In situ optical tracking of oxidation state changes of NiFe alloys by Reflectance Anisotropy Spectroscopy — •Sandhya Chandola[1], Karuppasamy Dharmaraj[2], Jörg Rappich[1], Norbert Esser[3,4], and Sonya Calnan[2] — [1]Young Investigator Group Nanoscale Solid-Liquid Interfaces (CE-NSLI), Hahn-Meitner-Platz 1, 14109 Berlin, Germany — [2]Kompetenzzentrum Photovoltaik Berlin, Schwarzschildstr. 3, 12489 Berlin, Germany — [3]Institut für Festkörperphysik, TU Berlin, Hardenbergstr. 36, 10623 Berlin, Germany — [4]Leibniz-Institut für Analytische Wissenschaften ISAS e.V., Schwarzschildstr. 8, 12489 Berlin, Germany

The oxygen evolution reaction (OER) is a key reaction involved in water splitting and has attracted increasing attention for hydrogen generation for clean energy uses. Nickel/iron (NiFe)-based compounds have been known as active OER catalysts for decades, and there has been increasing interest in developing NiFe-based materials for higher activity and stability.

Reflection Anisotropy Spectroscopy (RAS) in the visible spectral range is a polarisation sensitive optical spectroscopy technique used to study morphological and electronic structure changes of surfaces and thin films in-situ. It achieves high sensitivities in the detection of ultrathin layer structures down to the sub-nanometer scale and can follow the optical changes of the surface during catalytic reactions. By combining in-situ RAS and cyclic voltammetry (CV), the changes in both the redox state and structural phase during OER activity of NiFe alloys can be optically tracked.

O 63.25 Wed 17:00 Poster B

Triptycene as a versatile building block for self-assembled monolayers — Takanori Fukushima[1], Manfred Buck[2], Egbert Zojer[3], and •Michael Zharnikov[4] — [1]Tokyo Institute of Technology, Yokohama 226-8503, Japan — [2]EaStCHEM School of Chemistry, University of St Andrews, St Andrews KY16 9ST, UK — [3]Institute of Solid State Physics, NAWI Graz, Graz University of Technology, 8010 Graz, Austria — [4]Angewandte Physikalische Chemie, Universität Heidelberg, 69120 Heidelberg, Germany

When employing self-assembled monolayers (SAMs) for tuning surface- and interface-properties, a strong binding to the substrate, structural uniformity, and the ability to align functional groups and to control their density are desirable. To achieve these goals, tripod systems bearing multiple bonding sites have been developed as an alternative to conventional monodentate systems. A bonding of all three sites has, however, hardly been achieved with the consequence that structural uniformity and orientational order in tripodal SAMs are usually quite poor. To resolve that problem, we designed a series of triptycene-based molecules decorated with three anchoring groups, which can be assembled on different substrates. Depending on the character of the anchoring groups, well-defined tripodal SAMs could be prepared on Au(111), Ag(111), and indium tin oxide. Either unsubstituted or differently substituted triptycene-based molecules were assembled in context of different issues, viz. (i) homogeneous tripodal assembly, (ii) polymorphism and hidden chirality, (iii) on-surface click chemistry, (iv) multiple pathways in charge transfer, and (v) nanofabrication.

O 63.26 Wed 17:00 Poster B

A new design concept for SAMs of N-heterocyclic carbenes — Mateusz Wróbel[1], Daria M. Cegiełka[1], Andika Asyuda[2], Krzysztof Kozieł[3], •Michael Zharnikov[2], and Piotr Cyganik[1] — [1]Smoluchowski Institute of Physics, Jagiellonian University, 30-348 Krakow, Poland — [2]Angewandte Physikalische Chemie, Universität Heidelberg, 69120 Heidelberg, Germany — [3]Faculty of Chemistry, Jagiellonian University, 30-387 Krakow, Poland

Self-assembled monolayers (SAMs) of N-heterocyclic carbenes (NHCs) on metal substrates are currently one of the most promising systems in context of molecular engineering of surfaces and interfaces. Interest in these systems is mainly driven by their assumingly higher thermal stability compared to thiolate SAMs most broadly used at the moment. Most of the NHC SAMs utilize imidazolium as an anchoring group for linking molecules to the metal substrate via carbene C atom. It is well established in the literature that upright-oriented and stable NHC SAMs can only be built when using bulky side groups attached to nitrogen heteroatoms in imidazolium moiety, which, however, reduce significantly the packing density. In contrast, combining several complementary experimental techniques, we show that aromatic monolayers exhibiting at least double surface density, upright molecular orientation, and ultra-high thermal stability compared to the NHC SAMs reported before can be readily fabricated on the basis of NHCs with small methyl side groups in combination with simple solution-based preparation procedure. These parameters are crucial for numerous applications, including molecular and organic electronics.

O 63.27 Wed 17:00 Poster B

Interaction study between thin films of polyvinyl acetate and (plasma-treated) aluminum — •Sascha Zimmermann[1], Philipp Moritz[1], Oliver Höfft[1], Lienhard Wegewitz[1], Wolfgang Maus-Friedrichs[1], and Sebastian Dahle[2] — [1]Clausthal University of Technology, Germany — [2]University of Ljubljana, Slovenia

Composite materials made of wood and aluminum are becoming increasingly popular due to the combination of their properties. However, in order to exploit these advantages, the two materials must be bonded with sufficient stability. While the adhesive polyvinyl acetate (PVAc) exhibits high adhesive strength on wood, its bonding capabilities with aluminum are limited. The surface of the aluminum must therefore be modified. In this work, the aluminum surface is treated with a dielectric barrier discharge plasma in air with the aim of enabling molecular interactions. Various analytical techniques, including X-ray photoelectron spectroscopy (XPS), Reflection Absorption InfraRed Spectroscopy (RAIRS) and Atomic Force Microscopy (AFM), are employed to analyse the chemical interactions between aluminum and PVAc. This involves producing nm-thin films using spin coating. For untreated aluminum, no interactions were detected. AFM revealed the inability to deposit nanometer-thin films due to the lack of interactions, leading to the formation of PVAC-clusters. Plasma-treated aluminum led to a stronger chemical shift in the XPS spectra indicating hydrogen bonding. This finding was confirmed by RAIRS, which shows bands associated with hydrogen bonding.

O 63.28 Wed 17:00 Poster B

Spectroscopical properties investigation of pyrene based molecules in perspective of singlet fission — •Sruthy Asa Rajan, Sergey Bagnich, and Anna Köhler — Universität Bayreuth, Bayreuth, Germany

Photovoltaics plays a vital role in renewable energy. Theoretically, convectional silicon solar cells' efficiency is limited to 33% known as the Shockley-Queisser limit, which is limited by some practical and fundamental losses like thermalisation loss. Singlet fission (SF) is a carrier multiplication process, which has the potential to overcome Shockley-Queisser limit. In an organic semiconductor, chromophore in its singlet excited state shares its energy with a nearby ground-state chromophore to form two triplet excitons. Recent studies shown that some pyrene derivatives exhibit SF. A study has shown that bridged SF chromophore forms generated triplet pair and hence leading to formation of long-lived triplets.

We are investigating the impact on absorption, photo-luminescence, lifetime decay, time-resolved spectroscopy and quantum yield of different N-substitution groups on para Diketopyrrollopyrrole (PDPP) based pyrene derivatives, which have a comparable triplet energy to silicon bandgap. We will focus more on the changes in morphology of film of these compounds by varying the conditions of film preparation, such solvents, annealing effect, etc. We hypothesis that certain orientation of these pyrene-base molecules can trigger singlet fission.

O 63.29 Wed 17:00 Poster B

Understanding the disparate interactions of thin polymer films with natively oxidized metal surfaces and metal oxide crystals — •Friedrich Bürger, Philipp Moritz, Lienhard Wegewitz, and Wolfgang Maus-Friedrichs — Clausthal Centre of Material Technology, Clausthal University of Technology, Agricolastr. 2, 38678 Clausthal-Zellerfeld

In the Collaborative Research Centre 1368 “oxygen free production” the molecular interactions between thin films of commonly used polymers, poly(ethyl cyanoacrylate) (PECA) and poly(methyl methacrylate) (PMMA), and different metal oxides have been characterized. In the context of this project, certain interactions became apparent, e. g. hydrogen bonding or ionic interactions that were observed on natively oxidized samples representing very thin oxide films. In contrast, these interactions were absent on a TiO_2 single crystal. To investigate if this effect is reproducible and inherent to metal oxide single crystals, the interactions of PECA and different metal oxide single crystals are determined using X-Ray Photoelectron Spectroscopy (XPS), Ultraviolet Photoelectron Spectroscopy (UPS) and Metastable Induced Electron Spectroscopy (MIES). These results are compared to the interactions of natively oxidized metal samples with thin polymer films. Assuming reproducibility, a root cause for the disparate interactions is proposed.

O 63.30 Wed 17:00 Poster B

Influence of processing atmospheres on curing and bonding of cyanoacrylate adhesives — •Philipp Moritz, Oliver Höfft, Lienhard Wegewitz, and Wolfgang Maus-Friedrichs — Technical University Clausthal, Clausthal-Zellerfeld, Germany

Cyanoacrylates are fast-curing adhesives that are often used in joining technology to bond components together quickly. The curing and strength of the bond are often improved with numerous additives and surface pre-treatments. However, the surrounding process atmosphere is an aspect that has hardly been used to control curing and adhesive interactions.
To investigate the interactions and curing, thin films of cyanoacrylate are deposited on natively oxidized copper substrates. The surrounding atmosphere is varied between (i) air atmosphere, (ii) argon and (iii) an oxygen-free environment (O_2 partial pressure $< 10^{-20}$ mbar). The curing and underlying molecular interactions at the interface between cyanoacrylate and oxidized copper are investigated using spectroscopic and microscopic methods.
Curing in argon and the O_2-free atmosphere is significantly slower than in air. Nevertheless, strong interactions occur in an oxygen-free atmosphere and air, e.g. hydrogen bonds and ionic interactions. In argon, on the other hand, no molecular interactions are observed.
Funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) – Project-ID 394563137 – SFB 1368.

O 63.31 Wed 17:00 Poster B

The influence of PDMS residues on the mobility of molecules deposited onto Si/SiO2 wafers — •Erik von der Oelsnitz[1,2], Tim Völzer[1,2], Julian Schröer[1], Tobias Korn[1,2], and Stefan Lochbrunner[1,2] — [1]Institute of Physics, University of Rostock, Germany — [2]Department ”Life, Light & Matter”, University of Rostock, Germany

The exfoliation and stamping of flakes of 2D materials onto a substrate is one of the most used methods for the preparation of transition metal dichalcogenide (TMDC) monolayers. However, it was found that during the transfer of the flake, impurities and in particular residues of the common stamp material polydimethyl siloxane (PDMS) are transferred to the substrate. This could have a major impact when analyzing TMDCs, especially in TMDC/molecule hybrid structures. In order to examine the effect of these impurities, dye molecules were evaporated onto a Si/SiO2 wafer and then measured in a fluorescence lifetime microscope (FLIM). Here, in a certain area, the dye coverage is removed by laser-induced photodegradation and the diffusion of the molecules into this depleted area is examined. Three wafers are compared, each representing a specific step of the preparation process. The FLIM measurements showed that the molecules on the bare wafer diffuse into the depleted area the fastest, while they take significantly longer on the samples that came into contact with PDMS. This shows that the surface properties of exfoliated 2D materials are significantly influenced by the contact with PDMS, which in turn has a major effect on the mobility and thus the dynamics of deposited molecules.

O 64: Laureate of the Gaede Prize 2024

Time: Wednesday 17:30–18:00 Location: MA 004

Prize Talk O 64.1 Wed 17:30 MA 004

Molecular spin switches on surfaces — •Manuel Gruber — University of Duisburg-Essen, Duisburg, Germany — Laureate of the Gaede-Prize 2024

Magnetic molecules on surfaces have attracted considerable interest, in particular, in view of potential (quantum) technological applications. The ligands around the metal center essentially determine the electronic configuration of the ion, which is closely interconnected with the electronic, optical, and *magnetic* properties of the molecule. Molecules with multiple stable ligands configurations are particularly interesting as they exhibit a palette of physical properties.

We will report on different strategies to controllably modify the magnetic properties of molecules. Spin-crossover complexes exhibit two stable configurations with different ligand-field strengths. We will present the local and remote reversible spin-state switching of such complexes on a metal surface via electron injection [1]. For a second class of system, a ligand is displaced relative to the metal center to change the symmetry of the ligand field, and thereby the spin [2]. In a last example, the orbital moment of a dinuclear complexes is modified by addressing the peripheral ligands [3].

[1] Johannsen *et al.*, ACS Nano **15**, 11770 (2021), [2] Köbke *et al.*, Nat. Nanotechnol. 15, 18 (2020), [3]*Li *et al.*, ACS Nano **17**, 10608 (2023)

O 65: Poster: Molecular Nanostructures on Surfaces

Time: Wednesday 18:00–20:00 Location: Poster C

O 65.1 Wed 18:00 Poster C

On surface synthesis of a Dicaesium Tetraphenylporphyrin Complex: A combined TPR, XPS, STM and DFT study — •Kassandra Zoltner, Leonard Neuhaus, Florian Münster, Lukas Heuplick, Jan Herritsch, and J. Michael Gottfried — Department of Chemistry, University of Marburg, 35043 Marburg, Germany

Tetrapyrroles such as porphyrins and their metal complexes exhibit interesting chemical and optoelectronic properties leading to numerous potential applications in photosensor technology and organic electronics. While the surface chemistry of transition metal-porphyrin complexes has been extensively studied, there is a growing interest in exploring complexes with alkali metals. This study focuses on the on-surface reaction of tetraphenylporphyrin (H_2TPP) with caesium in the multilayer and monolayer regime on Ag(111). The reaction product was conclusively identified by temperature programed desorption mass spectrometry (TPD-MS) as Cs_2TPP, where both aminic hydrogens were replaced by caesium ions. The data also indicate that Cs_2TPP is thermally stable up to 700 K. Upon annealing up to 400 K the N1s X-ray photoelectron spectrum revealed a complete metalation to Cs_2TPP in the monolayer, whereas in the multilayer a partial conversion is already observed at room temperature. DFT calculations indicate that Cs_2TPP exhibits a bipyramidal structure with Cs ions on both sides of the molecular plane. The conducted study sheds light on the on-surface coordination chemistry of heavy alkali metal tetrapyrrole complexes potentially leading to further improvement and advancements in modern technologies.

O 65.2 Wed 18:00 Poster C

Step reconstruction from metal-molecule interaction — •Julie Teerink, Aleš Cahlík, Danyang Liu, Carolina A. Marques, and Fabian D. Natterer — Department of Physics, University of Zurich, Winterthurerstrasse 190, CH-8057, Switzerland

The model system for metal-molecule interaction PTCDA/Ag(111) shows diverse structural phases and strong charge-transfer. Co-adsorption of PTCDA and Ag-adatoms leads to substantial mass transfer and restructuring of the substrate surface. The PTCDA molecules embed themselves into Ag step edges, which strongly curve and form isolated Ag step-spirals or Ag islands, depending on the presence of substrate screw dislocations. These islands and step-spirals show 2D and 1D quantum confinement of the Ag surface state. To investigate the relationship between molecule length and step curvature, we co-deposit Ag-adatoms with NTCDA, a molecule with the same oxygen-carbon bonds as PTCDA but with a smaller carbon backbone.

O 65.3 Wed 18:00 Poster C

Synthesis and Characterization of Pentadecacene by Tip-manipulation with a Scanning Probe Microscope — •Grigori Pasko[1], Zilin Ruan[1], Tim Naumann[1], Jakob Schramm[2], John Bauer[3], Holger F. Bettinger[3], Ralf Tonner-Zech[2], and J. Michael Gottfried[1] — [1]University of Marburg, 35043 Marburg, Germany — [2]University of Leipzig, 04103 Leipzig, Germany — [3]University of Tübingen, 72076 Tübingen, Germany

Carbon nanomaterials exhibit versatile electronic properties that potentially can be exploited in the field of organic semiconductors. The acene series is an interesting class of compounds consisting of linearly fused polycyclic aromatic hydrocarbons (PAHs). The description as a closed-shell system becomes inadequate for long acenes as their (poly-)radical character increases. The resulting enhanced reactivity makes higher acenes hardly accessible for solution chemistry, so that novel synthetic procedures are required. Here, we demonstrate the on-surface synthesis of pentadecacene (15ac), the longest acene known to date, via atom-manipulation-induced C-C-bond dissociation of a trietheno-bridged precursor on the Au(111) surface. Scanning tunneling microscopy/spectroscopy (STM/STS) and atomic force microscopy (AFM) were carried out at temperatures below 5 K to investigate the electronic properties. 15ac has an antiferromagnetic singlet (S=0) ground state characterized by a singlet-triplet gap of 124 meV and a transport gap of 1.11 eV. Spin localization and Kondo-screening was observed for Au-complexes of 15ac with an uneven number of C-Au-bonds which reveals the S=1/2 ground state of such complexes.

O 65.4 Wed 18:00 Poster C

On-surface reactions of functionalized p-terphenyl on Cu(111): kinetics of the single reaction steps involved — •Mohit Jain[1], Tamam Bohamud[1], Daniel Kohrs[2], Nathaniel Ukah[2], Hermann A. Wegner[2], and Michael Dürr[1] — [1]Institut für Angewandte Physik and Zentrum für Materialforschung, Justus-Liebig-Universität Giessen, Germany — [2]Institut für Organische Chemie and Zentrum für Materialforschung, Justus-Liebig-Universität Giessen, Germany

For the commonly employed coupling types in on-surface synthesis, the different intermediates formed in the course of the reactions have been investigated in great detail over the recent years. For this, the intermediates are typically induced at variable surface temperatures and are then analysed at low temperature.

In contrast to this, we study the kinetics of the on-surface reaction of 3,3"-dibromo-p-terphenyl on Cu(111) when following the reaction at constant temperature (300 K) with time. Whereas the intermediates are first dominated by long, chain-like structures, they reorganize with increasing time into small 3-membered ring structures. The process includes, a.o., breakage and formation of the involved metal-organic complexes, but trans-to-cis isomerization of the two conformers on the surface was observed to be the slowest reaction step. An almost complete phase change from a mixed phase of chain- and ring-like structures to a phase of regularly oriented, 3-membered ring structures was thus observed only on longer time scales.

O 65.5 Wed 18:00 Poster C

Theoretical investigation of dibromopyrene on sodium chloride coated copper substrate — •Florian Alexander Pfeiffer[1], André Schirmeisen[2], Daniel Ebeling[2], and Simone Sanna[1] — [1]Insitute for Theoretical Physics, Justus Liebig University Gießen, Germany — [2]Insitute for Applied Physics, Justus Liebig University Gießen, Germany

Organic 2D materials as molecular electronics are of great interest for various applications. Increasingly sophisticated methods of on-surface manipulation extend the scope of possible structure modifications to tune the electronic properties of such nanoarchitectures.

Halogenated organic precursors such as dibromopyrene (DBP) make the building blocks for assembly. A sodium chloride bilayer helps to electronically decouple the metallic surface (here Cu (111)) from the adsorbate, increasing mobility and simplifying manipulation.

Density Functional Theory (DFT) allows for bottom-up ab initio investigations of the system, while experiments reveal insights top-down, yielding mutual benefits. The DFT code of the Vienna Ab initio Simulation Package (VASP) was utilized to calculate adsorption geometry and energy with Potential Energy Surfaces (PES). Diffusion pathways and their energy barriers could be determined using the Nudged Elastic Band (NEB) method.

For further comparability with experimental results simulations of Scanning Tunneling Microscopy (STM) and Atomic Force Microscopy (AFM), utilizing the Probe Particle Model [1], have been calculated.

[1] P. Hapala et al., Phys. Rev. B 90, 085421 (2014).

O 65.6 Wed 18:00 Poster C

Photoinduced increase of the local molecular coverage on a surface — •Christophe Nacci and Leonhard Grill — Institute of Chemistry, University of Graz, Graz, Austria

Ullmann coupling is one of the preferred reactions to synthesize covalent molecular architectures on surfaces [1]. The dissociation of specific substituents carried by the molecular precursors as well as the formation of new bonds can be triggered by different external stimuli, for instance heat, light and electric current. While the thermal control of on-surface chemical reactions is a well-established approach, only few studies report the on-surface polymerization by light.

Here, light-induced chemical reactions of an anthracene derivative on a surface [2] were investigated by combining low-temperature scanning tunneling microscopy and X-ray photoemission spectroscopy. Special emphasis is on the spatial distribution of the products, studied by position-dependent measurements. Polymerization takes place only in a limited portion of the surface, i.e., at the areas hit by the most intense laser spot. A significant increase of the local molecular coverage is observed and mainly ascribed to the local interplay between thermally induced diffusion of the adsorbed species and the reduced mobility of the grown oligomers. Moreover, despite long UV irradiations, debromination of the adsorbed species never progresses to completion within the experiment time.

[1] L. Grill and S. Hecht, Nature Chem. 12, 115 (2020) [2] C. Nacci et al., J. Phys. Chem. C 125, 22554 (2021)

O 65.7 Wed 18:00 Poster C

Electrospray Ionization Ion Beam Deposition of Organic Molecules — •Florian Münster, Lukas Heuplick, Grigori Pasko, Jan Herritsch, and J. Michael Gottfried — Department of Chemistry, University of Marburg, 35043 Marburg, Germany

When studying large molecules on surfaces, the deposition process can often be a limiting factor. While small molecules can usually be thermally evaporated using a Knudsen cell, the vapor deposition of larger molecules may be hindered by their low vapor pressure, especially if the molecules have labile functional groups. One approach to overcome these preparation-related limitations is electrospray ionization ion beam deposition (ESI-IBD). While this technique is becoming increasingly popular, for example in the preparation of large biomolecules, the precise chemical state of the deposited molecules is often unknown. For this study, we chose meso-tetraphenylporphyrin as a well-known model system to investigate the chemical state of the molecule after deposition via ESI-IBD. X-ray photoelectron spectroscopy (XPS) indicates the presence of an N-protonated species. By utilizing scanning tunneling microscopy (STM), we observed self-assembled islands composed of multiple distinguishable species, in agreement with the existence of N-protonated porphyrin molecules.

O 65.8 Wed 18:00 Poster C

Theoretical investigation of 3,3"-dibrom-p-Terphenyl on copper substrate — •Kevin Eberheim[1], Simone Sanna[1], and Michael Dürr[2] — [1]Institut for Theoretical Physics, Justus-Liebig-University Gießen, Germany — [2]Institut for Applied Physics, Justus-Liebig-University Gießen, Germany

Selectivity is a key parameter for building customized organic nanostructures via bottom-up approaches. Therefore, strategies are needed that allow connecting molecular entities at a specific stage of the assembly process in a chemoselective manner. Studying the mechanisms of such reactions is the key to apply these transformations for the buildup of organic nanostructures on surfaces. Especially, the knowledge about the precise adsorption geometry of intermediates at different stages during the reaction process and their interactions with surface atoms or adatoms is of fundamental importance, since often catalytic processes are involved. With first-principles simulations we can determine the orientation and position of the 3,3"-dibrom-p-Terphenyl as well as adsorbed mono/diradicals and the halogens. For further comparability with experimental results simulations of Scanning Tunneling Microscopy (STM) and Atomic Force Microscopy (AFM) have been calculated.

O 65.9 Wed 18:00 Poster C

Adsorption Behavior of N Heterocyclic Carbene with Thiophene on Au (111) — •Natasha Khera[1], Pranjit Das[1], Kwan Ho Au-Yeung[1], Suchetana Sarkar[1], Soyoung Park[2], Franziska Lissel[2], and Francesca Moresco[1] — [1]Center for Advancing Electronics Dresden, TU Dresden, 01062 Dresden, Germany — [2]Leibniz-Institut für Polymerforschung Dresden e.V., 01069 Dresden, Germany, and Faculty of Chemistry and Food Chemistry, TU Dresden, 01062 Dresden, Germany

This research delves into the adsorption and rotation characteristics of N-Heterocyclic Carbene with Thiophene adsorbed on an Au (111) surface, employing Low Temperature Scanning Tunneling Microscopy in Ultra High Vacuum (LT-UHV STM) conditions. The molecule under investigation displays two distinct chiralities and maintains a flat/planar configuration. The molecule demonstrates rotation (chirality independent) around sulfer upon voltage pulses with the STM tip.

O 65.10 Wed 18:00 Poster C

Control of single-molecule motion on Ag(111) surface — •Donato Civita, Julia Lanz, and Leonhard Grill — Department of Physical Chemistry, University of Graz, Austria

The motion of molecules adsorbed on surfaces is fundamental for molecular collisions and reactions at surfaces, for instance in heterogeneous catalysis, on-surface polymerization or the bottom-up construction functional nanostructures. On atomically flat surfaces, the motion of adsorbed molecules can be controlled by scanning tunnelling microscopy (STM) manipulation. However, this technique was so far restricted to few nanometres of dislocation distances and limited control on the dislocation pathway. The study of single dibromo-terfluorene (DBTF) molecules on a Ag(111) surface has revealed surprisingly large distances of more than 100 nm that can be achieved with STM manipulation with picometre precision [1]. The motion of single DBTF molecules is strictly confined to one atomic row of the flat surface, rendering this system highly suitable for the study of molecular dynamics.

Here, we show that the large spatial extension of DBTF motion, and its one-dimensional confinement allow the direct measurement of the molecule traveling time with microsecond precision. Thus, we use this direct measurement to study the molecular velocity in different surface areas and in dependence of the local surroundings that appear to substantially affect the molecular dynamics.

[1] D. Civita, M. Kolmer, G. J. Simpson, A.-P. Li, S. Hecht, L. Grill, Control of long-distance motion of single molecules on a surface, Science, Vol. 370, Issue 6519, pp. 957-960 (2020).

O 65.11 Wed 18:00 Poster C

Monitoring of molecular configurations during manipulation with a scanning probe microscope — •Joshua Scheidt[1,2], Jonas Lederer[2], Hadi H. Arefi[1], Mario Infantino[1], Alexander Diener[1], F. Stefan Tautz[1], Klaus-Robert Müller[2], and Christian Wagner[1] — [1]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, Jülich, Germany — [2]Fakultät IV - Elektrotechnik und Informatik, Technische Universität Berlin, Berlin, Germany

A bold vision in nanofabrication is the assembly of functional molecular structures using a scanning probe microscope (SPM). This approach requires continuous monitoring of the molecular configuration during manipulation. Until now, this has been impossible because the SPM tip cannot simultaneously act as an actuator and an imaging probe. We implement configuration monitoring using a machine learning model trained on DFT calculated data. We model the manipulation as a pre-computed Partially Observable Markov Decision Process (POMDP) in the form of a finite-state automaton, and use a particle filter to approximate the actual configuration in real time in the lab. This is enabled by a virtual reality interface in which we directly control the manipulation process and receive immediate feedback. Our proof-of-principle investigations are based on SPM manipulations of a PTCDA (3,4,9,10-perylene-tetracarboxylic dianhydride) molecule on the Au(111) surface.

O 65.12 Wed 18:00 Poster C

On-Surface Synthesis of Carbon Nanoribbons with Nonalternant Topologies — •Dong Han[1], Konstantin Y. Amsharov[2,3], and J. Michael Gottfried[1] — [1]Department of Chemistry, Philipps-Universität Marburg, Marburg, Germany — [2]Institute of Chemistry, Organic Chemistry, Martin-Luther-Universität Halle-Wittenberg, Halle, Germany — [3]Department of Chemistry and Pharmacy, Institute of Organic Chemistry II, Friedrich-Alexander-Universität Erlangen-Nürnberg, Erlangen, Germany

The graphene-based nanostructures embedded with nonhexagonal rings potentially exhibit exotic (opto-)electronic properties. However, their experimental realization remains rather challenging. Here we report the on-surface synthesis of carbon nanoribbons with several nonalternant topologies, including zigzag-shaped nanoribbon bearing 5-6-7 membered rings (dominant product) and porous nanoribbon, by lateral fusion of polyindeno[2,1-a]fluorene polymer chains on Au(111). The yield of nanoribbons is enhanced by successive molecular deposition to increase the coverage of polymer chains. The polymer chains originate from the Ullmann coupling of halogenated precursors and subsequent cyclodefluorination. The reaction species are scrutinized by scanning probe microscopy (SPM). X-ray photoelectron spectroscopy (XPS) is utilized to track the reaction process. This work demonstrates the versatility of lateral fusion in fabricating unusual carbon nanomaterials.

O 65.13 Wed 18:00 Poster C

Surface Chemical Bond and Molecular Topology of Polycyclic Aromatic Systems: Pyrene vs. Acepleiadylene — •Lukas Ruppenthal[1], Florian Münster[1], Neuhaus Leonard[1], Tim Naumann[1], Jon H. Both[1], Jan Herritsch[1], Zilin Ruan[1], Pengcai Liu[2], Xing-Yu Chen[2], Jiawen Cao[2], Jakob Schramm[3], Ralf Tonner-Zech[3], Xiao-Ye Wang[2], and J. Michael Gottfried[1] — [1]University of Marburg, Germany — [2]Nankai University, China — [3]University of Leipzig, Germany

Metal/organic interfaces have a large impact on the performance of organic (opto-)electronic devices. Therefore, the detailed understanding of their chemical, electronic and geometric structure is important for the further technological development. Many common organic semiconductors contain π-electron systems with alternant topologies, whereas non-alternant alternatives have only recently found increasing attention due to their unusual electronic properties. Here, we compare the alternant polycyclic aromatic molecule pyrene with its non-alternant isomer acepleiadylene regarding their interaction with the Cu(111) surface, using PES, NEXAFS, TPD, LT-STM, nc-AFM and DFT. We find that the non-alternant isomer shows increased metal/molecule-interaction and adsorption energy as well as a lower adsorption distance due to its reduced HOMO-LUMO gap, which brings the LUMO energetically closer to the Fermi energy of the metal, causing stronger hybridization with electronic states of the metal surface.

O 65.14 Wed 18:00 Poster C

Visualising the steps in a chemical reaction: On-surface reactivity of brominated tetraphenyl porphyrin on Cu(111) — •Ailish Gray, Michael Clarke, Matthew Edmondson, and Alex Saywell — School of Physics & Astronomy, University of Nottingham, UK

Scanning probe microscopy techniques have been extensively employed to provide details of on-surface reactions [1] and can provide significant insight into the effects of substrate and molecular chemistry upon the reaction products of on-surface synthesis.[2] The Ullmann-type coupling of brominated tetraphenyl porphyrin (BrxTPP) on Au(111) [3] is a seminal work, demonstrating molecular characterisation of on-surface covalent coupling. However, on Cu(111) the reaction progress is different,[4] and for various substrates a variety of reactions (including; ring-closing, metalorganic coordination, and self-metalation) may occur. Here, we detail a low-temperature ultra-high vacuum scanning tunnelling microscopy (STM) study of BrxTPP (x=0-4) on Cu(111). We observe multiple steps in an on-surface reaction; debromination, formation of metal-organic frameworks, ring opening, self-metalation, and covalent coupling. Systematic, temperature controlled, investigation reveals step-wise evolution of the reaction, with the intermediate reaction stages probed using STM imaging and manipulation.

[1] A. Sweetman, N.R. Champness, A. Saywell, Chem. Soc. Rev., 2020, 49, 4189 [2] L. Grill, S. Hecht, Nat. Chem. 2020, 12, 115 [3] L. Grill et.al, Nat. NanoTech., 2007, 2, 687 [4] C.M. Doyle et. al, Chem. Commun. Camb., 2011, 28, 12134

O 65.15 Wed 18:00 Poster C

On-surface degradation reactions of Pb(II)-tetraphenylporphyrin — •Cong Guo, Jan Herritsch, Lukas J. Heuplick, Stefan R. Kachel, Mark Hutter, and J. Michael Gottfried — Department of Chemistry, University of Marburg, 35043 Marburg, Germany

Metal-organic compounds of heavy main group elements are promising building blocks for the assembly of novel (opto-)electronic surface structures. However, dynamic processes of these compounds at surfaces or interfaces (i.e., chemical reactions or structural adaption upon adsorption) are largely unexplored, but can be crucial for the devices' performance. To fill this knowledge gap, we investigated the on-surface chemistry of lead(II) tetraphenylporphyrin (Pb(TPP)). On Cu(111), Pb(TPP) undergoes spontaneous transmetalation above 380 K, resulting in an exchange of the incorporated Pb ion by a Cu adatom.[1] However, a different reactivity was found for Pb(TPP) on Au(111). Here, a spontaneous demetalation of Pb(TPP) and formation of a free-base porphyrin was observed by XPS, STM, and TPD. Temperature-dependent XPS confirms the demetalation and reveals cyclodehydrogenation side-reactions of the porphyrin ligand starting above 450 K. At 720 K, re-metalation of the porphyrin with substrate Au atoms results in Au(II)-porphyrin. The observed de- and trans-metalation reactions are practically relevant, because they alter the electronic and chemical properties of the metal-organic interface substantially. [1] J. Herritsch et al., Nanoscale 13, Nanoscale, 2021,13, 13241-13248 (2021).

O 65.16 Wed 18:00 Poster C

Molecular materials for carbon capture and storage: On-surface characterisation of self-assembly and thermal-stability of metal phthalocyanines — •Joseph Straw, Matthew Edmondson, Liv Warwick, James N. O'Shea, and Alex Saywell — School of Physics & Astronomy, University of Nottingham, UK

The successful capture and storage of carbon- and nitrogen-containing greenhouse gases (e.g. CO2 and NOx - produced by industrial processes) is of significant environmental importance; materials enabling gas sequestration are a route to reducing the effects of climate change. Molecular based materials, containing units which can ligate and trap gas molecules, may provide a way of selectivity and efficiency removing pollutants. Porphyrins and phthalocyanines (Pcs), both of which can be functionalised with reactive metal atoms, have been extensively studied in ultra-high vacuum (UHV) conditions.[1] Importantly, the ligation of gaseous species to the metal centres of such molecules has been shown to be feasible. [2] Here we study the formation of molecular arrays of iron phthalocyanine (FePc) on Au(111) under systematically controlled thermal conditions. We employ UHV scanning tunnelling microscopy (UHV-STM) and X-ray photoelectron spectroscopy (XPS) to characterise the morphology and chemistry of the on-surface synthesised structures and investigate thermal stability of the Pc macrocycle and extended molecular islands.

[1] J. M. Gottfried, Surf. Sci. Rep. 70, 259 (2015) [2] E. Vesselli, J. Phys. Mater. 3, 022002 (2020)

O 65.17 Wed 18:00 Poster C

On-surface synthesis of nanographenes on proximitized superconducting substrates. — •Katerina Vaxevani[1], Dongfei Wang[1], Stefano Trivini[1], Jon Ortuzar[1], and Jose Ignacio Pascual[1,2] — [1]CIC nanoGUNE-BRTA, 20018 Donostia-San Sebastian, Spain — [2]Ikerbasque, Basque Foundation for Science, 48013 Bilbao, Spain

Exploring molecular magnetism on superconducting substrates has the advantage over atomic spins of selectively tuning the exchange interaction with the underlying substrate. However, most elemental superconductors have highly reactive surfaces which result in the decomposition of the molecular species upon adsorption. To tackle this problem, we fabricated a metallic proximitized superconducting platform to achieve on-surface synthesis of molecular nanostructures while maintaining the superconducting properties of the substrate. First, we focused on the growth of Ag on a Nb(110) single crystal, where we find 20nm-high superconducting Ag(111) islands. There, we deposited the 2OS radical molecules to study the interaction of the molecular spin with the substrate. Additionally, we realized on-surface synthesis of the triangulene chains, previously reported only for an Au(111) pristine surface. A clear magnetic fingerprint, manifested as in-gap Yu-shiba-rusinov states, is found in some triangulene units probably because of intrinsic defects in the formation of the chains, while regular formed chains show no magnetic signal. Finally, we show our next approach for the ideal platform to study spin physics of extended pi-conjugated molecular systems.

O 66: Poster: Nanostructures at Surfaces

Time: Wednesday 18:00–20:00 Location: Poster C

O 66.1 Wed 18:00 Poster C

Direct electron beam patterning of electro-optically active PEDOT:PSS for switchable metasurfaces — •Dominik Ludescher[1], Siddharth Doshi[2,3], Julian Karst[1], Moritz Floess[1], Johan Carlström[3], Bohan Li[3], Nofar Mintz Hemed[2], Yi-Shiou Duh[3], Nicholas A. Melosh[2], Mario Hentschel[1], Mark Brongersma[3] und Harald Giessen[1] — [1]4th Physics Institute and Research Center SCoPE, University of Stuttgart, Germany — [2]Department of Materials Science and Engineering, Stanford University, Stanford, CA 94305, United States — [3]Geballe Laboratory for Advanced Materials, Stanford University, 476 Lomita Mall, Stanford, CA 94305, USA

Conducting polymers, exemplified by PEDOT:PSS, exhibit distinctive electronic and polymeric attributes. When subjected to CMOS-compatible voltages, PEDOT:PSS transitions between insulating and metallic states via an intrinsic electrochemical redox reaction. Consequently, these materials are suited perfectly for AR/VR applications, advanced display technologies, and dynamic sensors. Harnessing the full potential, however, demands a robust foundation in fabrication techniques. Historically, electron-beam lithography was utilized to generate a resistant etch mask, albeit with fabrication complexities. To circumvent some of these challenges, we present a novel and alternative fabrication method. This concept is based on the electron-beam induced water solubility modulation due to crosslinking between the PEDOT:PSS polymer chains. With this approach nano-wire structures with a width of down to 250 nm can be achieved.

O 66.2 Wed 18:00 Poster C

Enhancing Chiroptical Sensitivity: Exploring Chirality with Second Harmonic Generation-Circular Dichroism Spectroscopy — •Charitini Panagiotopoulou, Kevin Liang, Changseop Jeong, Ueli Heiz, and Aras Kartouzian — Chair of Physical Chemistry, School of Natural Sciences & Catalysis Research Center, Technische Universität München, Lichtenbergstr. 4, 85748 Garching, Germany

Chirality holds significance in a variety of molecules, playing a fundamental role in chemical, biological, and material processes. Circular Dichroism (CD) spectroscopy, utilizing circularly polarized light, discloses the differential in absorption between right and left circularly polarized light. Chiral molecules, featuring non-superimposable mirror images, necessitate CD for rapid, non-destructive analysis of their structural and electronic properties. Our study introduces a method to enhance CD sensitivity; Second Harmonic Generation (SHG) spectroscopy, known as SHG-CD spectroscopy. This approach uses the nonlinear response of SHG, which is particularly effective in non-centrosymmetric environments like thin film surfaces. Under UHV conditions, we investigate the structural aspects of chiral molecules on thin films and the chirality transfer to achiral counterparts, making a wide range of applications possible, from fundamental studies of surface properties to practical applications in fields such as materials science, biophysics, and catalysis.

O 66.3 Wed 18:00 Poster C

Measuring the local surface potential of ionic liquids layers upon charge deposition — •Mirco Wendt[1,2], Regina Lange[2], Jens Berdermann[1], Ingo Barke[2], and Sylvia Speller[2] — [1]Institute for Solar-Terrestrial Physics, German Aerospace Center (DLR), Kalkhorstweg 53, 17235 Neustrelitz — [2]Institute of Physics, University of Rostock, Albert-Einstein-Str. 25 18059 Rostock

The interaction of ionic liquids (IL) with surfaces at different potentials is extensively studied with respect to their potential application in double layer capacitors[1,2]. The surface potential of the liquid itself, however, is rarely investigated[3], although it was shown that electrostatic screening in IL does not adhere to well established models for low concentration electrolytes[4]. We show how the local surface potential of 1-Butyl-1-methyl-pyrrolidinium- dicyanamide (BMP DCA) can be measured in ultra-high vacuum, using a q-Plus-Sensor in a Kelvin-Probe-like configuration. We address implications and assets of this approach and discuss its application to BMP DCA thin films on floating gold islands on top of glass. To modify the native surface potential, patches of this film were exposed to the electron beam of a scanning electron microscope and show both temporary and permanent contrast differences upon prolonged exposure. Studying the local surface potential of such patches and its evolution should allow for a better understanding of mechanisms involved.

[1] Thangavel, et al., J. Power Sources 2018 [2] Han, et al., Adv. Mater. Interfaces 2020 [3] Zhang, et al., Acta Phys. Chim. Sin. 2016 [4] Gebbie, et al., Proc. Natl. Acad. Sci. 2013

O 66.4 Wed 18:00 Poster C

Plasma Treatment of Electrode Surfaces — •Joshua Nicolai Maruschtschyn[1], Timo Wagner[1], Nicolas Wöhrl[1], Vineetha Vinayakumar[2], Christian Marcks[3], Anna Mechler[3], Doris Segets[2], and Axel Lorke[1] — [1]Faculty of Physics and CENIDE, University Duisburg-Essen, Germany — [2]Particle Science and Technology (IVG-PST) and CENIDE, University of Duisburg-Essen — [3]AVT.ERT, RWTH Aachen University

In the planned transition toward a CO2 neutral economy and industry, hydrogen plays a significant part. In the production of hydrogen catalysis is of great importance, necessitating better and cheaper catalysts. To make high-performance electrodes cheaper, they will need to be produced in series. To establish this the H2Giga project was started. One promising method here in is the plasma treatment of the electrodes, with which various material properties can be enhanced, such as increasing the surface area or improving the adhesion of catalyst particle coatings. The method is viable for scaling and roll to roll production. The experimental setup and procedure used for the plasma treatment is discussed. Various electrode materials are treated, such as nickel, nickel with catalyst particle coatings, copper and others. The results are investigated using a scanning electron microscope, electrochemical tests and adhesion tests. The results are presented and possible future steps are discussed.

O 66.5 Wed 18:00 Poster C

Direct two-photon laser structuring of electrically switchable PEDOT:PSS for nanophotonics — •Pavel Ruchka, Dominik Ludescher, Leander Siegle, Mario Hentschel, and Harald Giessen — 4th Physics Institute and Research Center SCoPE, University of Stuttgart, 70569 Stuttgart, Germany

Switchable conducting polymers, such as PEDOT:PSS, have recently seen a growing interest, as they promise new possibilities for compact integrated devices in augmented/virtual reality applications, nanophotonics, and other fields of optics. Typically, such materials are structured by imposing and etching resist-masks, which can be tedious, costly, and time-consuming. Here, we present a new method to fabricate switchable micro- and nano-structures from PEDOT:PSS using direct laser writing by two-photon lithography. We study the resolution that can be achieved with this technique and demonstrate electrical switching of the optical properties of the obtained structures. As an example, a simple grating with sub-micrometer linewidth is produced and switched, resulting in an electrically switchable nanophotonic beam-steering device, which can be benchmarked to already existing fabrication methods.

O 66.6 Wed 18:00 Poster C

Design of femtosecond nanostructured stainless steel orthopedic implants, magnetron sputtered with Cu and optimized for calcium phosphates crystals growth for application in orthopedics — •Liliya Angelova[1], Albena Daskalova[1], Maja Sikiric[2], and Tihomir Car[2] — [1]Institute of Electronics, Bulgarian Academy of Sciences, 1784 Sofia, Bulgaria — [2]Ruder Bošković Institute, 10 000 Zagreb, Croatia

Nowadays, the engineering of load-bearing bone tissue after severe trauma still relies on metal-based permanent implants. Such artificial tissues are usually applied in the body and come into direct contact with the recipient's cells, whose adhesion directly affects the acceptance or rejection of the implant by the patient. The present study aims to create nano rough texture employing ultra-short femtosecond laser (fs)-induced periodicity in the form of LIPSS on the surface of a mechanically stable stainless steel implant model, that is additionally functionalized by magnetron sputtered Cu thin layer, giving the as created implants stable antimicrobial surface interface. Calcium phosphate (CaP) crystal growth was additionally performed due to the well-known high mechanical stability and load-bearing properties that CaPs give to the bone connective tissue, as well as for the strong interface bond that they make between the artificial implant and the surrounding bone tissue, stabilizing in that way the implanted structure in the body. The optimization of the bioactive properties of the as-created antimicrobial hybrid topographical design has the potential for subsequent practical application in bone tissue engineering.

O 66.7 Wed 18:00 Poster C

Dewetting of Thin Gold Films on Flat and Nanostructured Polystyrene Surfaces — •Felix Lohmeyer, Daniel Kool, Philipp Hodges, and Jörg K. N. Lindner — Nanopatterning - Nanoanalysis - Photonic Materials, Department of Physics, Paderborn University, Germany

Limited knowledge exists regarding the wetting and dewetting behavior of ultra-thin metal films on polymer surfaces, although this understanding is crucial for advancing nanotechnology processes. We show that dewetting of thin metal films occurs below the polymer's melting point making it valuable for patterning purposes. We present preliminary results for 7 nm thin gold films deposited on polystyrene after thermal annealing. Different polymer substrates, including planar polystyrene films, a phase-separated PS-b-PMMA block-copolymer with a fingerprint nanostructure, and a nanostructured polystyrene surface obtained by selectively removing the PMMA matrix from an ordered block-copolymer film, are explored.

Dewetting degree is examined as a function of annealing time and temperature. The fraction of Au-covered surface is larger on fingerprint-patterned PS-b-PMMA surfaces than on pure planar PS films. On nanostructured PS films, gold decomposes upon annealing into rods, both on top of the PS fins and in the pattern valleys. These results illuminate the potential of dewetting on polymers for nanotechnology purposes.

O 66.8 Wed 18:00 Poster C

Photoinduced dynamics of plasmonic HfN nanodots and thin films — •Marc Herzog[1], Maximilian Mattern[1], Alexander von Reppert[1], Steffen Zeuschner[1], Matthias Rössle[1], Florin Boariu[1], Jan-Etienne Pudell[2], Angel Rodriguez-Fernandez[2], Wonhyuk Jo[2], Anders Madsen[2], Sven Askes[3], Andrea Baldi[3], and Matias Bargheer[1,4] — [1]Institut für Physik, Universität Potsdam, Germany — [2]European XFEL, Germany — [3]Vrije Universiteit Amsterdam, Netherlands — [4]HZB Berlin, Germany

Metallic nanoparticles can be exploited to focus light energy onto the nm scale where it may trigger specific processes such as chemical reactions. The "gold standard" in nanoplasmonics, i.e. Au nanoparticles, suffers from the superior electron heat conductivity which quickly spreads the absorbed nanofocused energy away from the hot spots. Hafnium nitride (HfN) is a very robust material and its plasmonic properties and large electron-phonon coupling facilitate a light-generated long-lived nanofocusing of heat rendering it a very promising material in plasmon-assisted catalysis.

The knowledge of the local temperature in nanoparticles is critical for a thorough understanding of light-driven nanoscale processes, however, its experimental determination is by no means straightforward. Using ultrafast x-ray diffraction, we compare the photoinduced coherent and incoherent expansion dynamics of HfN thin films and substrate-supported nanodots. We thereby shed light on the morphology dependence of nanooptical and thermoelastic properties of HfN nanostructures.

O 66.9 Wed 18:00 Poster C

An intermediate morphology in the patterning of the crystalline Ge(001) surface induced by ion irradiation — •Denise J. Erb[1], Daniel A. Pearson[2], Tomas Skeren[3], Martin Engler[1], R. Mark Bradley[4], and Stefan Facsko[1] — [1]Helmholtz-Zentrum Dresden-Rossendorf, Germany — [2]Pennsylvania State University, USA — [3]Czech Technical University in Prague, Czech Republic — [4]Colorado State University, USA

We investigate the self-organized nanopattern formation of the Ge(001) surface induced by ion beam bombardment at elevated surface temperatures. Two previously-observed kinds of topographies are seen, i.e., anisotropic patterns of rectangular pyramids and isotropic patterns of shallow round basins [1]. In addition, we unexpectedly observe an intermediate type of pattern, in which isolated rectangular pyramids stand above a landscape of shallow basins. The observed morphology depends on the chosen ion energy and flux and surface temperature. To model the observed pattern morphologies, we extend the equation of motion to include a second order correction term resulting from the curvature dependence of the sputter yield from a patterned surface [2]. This term produces the isolated peaks in the intermediate patterns, while the formation of spike singularities is averted by the Ehrlich-Schwoebel effect. For a range of parameter values, the resulting continuum model of the surface dynamics produces patterns that are remarkably similar to the experimentally observed morphologies (manuscript in review). [1] X. Ou et al., Phys. Rev. Lett. 111 (2013) [2] R.M. Bradley and G. Hobler, J. Appl. Phys. 129 (2021)

O 66.10 Wed 18:00 Poster C

Argon ion induced nanostructuring on Titanium surfaces — •Jens Bauer, Hannaneh Hosseini, Gregor Dornberg, Frank Frost, and Andriy Lotnyk — Leibniz-Institut für Oberflächenmodifizierung, Permoserstraße 15, D-04318 Leipzig, Germany

A flexible processing tool to fabricate different kinds of nanostructured surfaces in form of ordered fields of nano-dot or nano-ripple structures is ion-beam erosion by low-energetic ions. A broadband argon ion beam at 1.5 keV is used to investigate the nanostructure formation on polished, polycrystalline cp-Ti samples. The morphology is analyzed by white light interferometry (WLI), atomic force microscopy (AFM), scanning electron microscopy (SEM), and transmission electron microscopy (TEM). Electron back-scatter diffraction (EBSD) experiments are performed to analyze the local crystal structure. Under normal ion incidence conditions the ripple morphology is found to be correlated to the crystallographic orientation of the titanium grain structure. Ordered nano-ripples with saw-tooth cross-section are formed with a structure period of 20 – 350 nm and a height of 4 – 50 nm. At low structure periods < 25 nm the ripple morphology tends to unclench resulting in chains of ordered nano-dot arrays. The structure evolution is investigated in the fluence range of 0.3 – 2.0 x 10^{19} $1/\mathrm{cm}^2$. The ripple formation on Ti is observed to be time independent indicating a convergent structuring process at normal ion incidence.

O 66.11 Wed 18:00 Poster C

Defect formation in 2D materials by irradiation with highly charged ions — •Lucia Skopinski, Yossarian Liebsch, Stephan Sleziona, Leon Daniel, Lars Breuer, and Marika Schleberger — Fakultät für Physik and CENIDE, Universität Duisburg-Essen, Duisburg, Germany

Two-dimensional (2D) materials, in particular transition metal dichalcogenides, are known for their unique properties. Under bombardment with highly charged ions (HCIs), these materials are modified by targeted defects, allowing tuning of desired properties. Our study addresses the fundamental mechanisms governing the interaction between HCIs and 2D-MoS_2 on different substrates. Each HCI possesses both kinetic and potential energy due to its velocity and charge state, leading to changes such as sputtering of particles upon impact with the material surface. However, a full understanding of the underlying mechanisms is still under discussion.

An in-depth analysis of the emission of secondary ions and atoms aims to unravel the importance of potential and kinetic energy for the underlying nuanced processes. The measured distributions allow a distinction between sputtering driven by the potential and the kinetic energy of the primary ion. The low velocities of the emitted particles suggest an interaction mechanism related to electron-phonon coupling. Furthermore, a comparative study of STEM measurements for pores in 2D-MoS_2 irradiated on a substrate and free-standing provides insight into the influence of the substrate on the modification of the 2D material.

O 66.12 Wed 18:00 Poster C

Microscopic and Magnetic Analysis of Iron Oxide Core-Shell Nanoparticles with Variable Core/Shell Ratio — •Aladin Ullrich, Michael Kühn, and Manfred Albrecht — Universität Augsburg, Universitätsstr. 1, 86159 Augsburg

Cubic iron oxide nanoparticles of about 15 nm were prepared by thermal decomposition of an iron oleate precursor in 1-Octadecene [1]. From scanning transmission electron microscopy and electron energy loss spectroscopy (EELS) measurements a core-shell composition of the particles was found. The particles show a wustite like structure in the core and a spinel like structure in the shell that is indicated from signatures in the EELS spectra [2]. Both, the position of the Fe $L_{2,3}$ white lines and the oxygen fingerprint structure were investigated. By successive oxidation of the particles under a controlled atmosphere, the core/shell ratio could be adjusted up to the complete disappearance of the core. The magnetic properties of this antiferromagnetic core - ferrimagnetic shell system were investigated for a sample series with core/shell ratios from about 0.3 to zero. The change of the magnetic properties like the blocking temperature, the coercivity, and the exchange bias field was investigated. Magnetic memory measurements following different protocols show clear memory effects and, hence, indicate the presence of a super spin glass state in the samples.

[1] A. Ullrich, M. M. Rahman, A. Azhar, M. Kühn, M. Albrecht, Nanoprticle Research **24**, 183 (2022). [2] A. Ullrich, M. M. Rahman, P. Longo, S. Horn, Scientific Reports **9**, 19264 (2019).

O 66.13 Wed 18:00 Poster C

Plasmonic nanoparticles on silicon: photoemission characteristics and control — •Waqas Pervez, Kevin Oldenburg, Sylvia Speller, and Ingo Barke — Institute of Physics, University of Rostock

Laser excitation of localized plasmons in nanoparticles can give rise to a vastly enhanced photoemission yield [1, 2]. Here we study detailed characteristics of the photoelectron emission process of size-selected nanoparticles with diameters around 10 nm, deposited from the gas phase on silicon. To this end we employ femtosecond laser excitation in a photoemission electron microscope (PEEM). The emission intensity is investigated as a function of space, emission angles, laser wavelength and polarization. We discuss how these parameters can be used to control the electron emission from individual particles. Furthermore, we propose experiments based on a two-color setup which should enable various excitation schemes and reveal phase effects leading to anisotropic electron emission, in analogy to the case of free particles [3].

[1] M. Rohmer et al., Phys. Stat. Solidi B 247, 1132 (2010)

[2] K. Oldenburg et al., J. Phys. Chem. C 123, 1379 (2019)

[3] J. Passig et al., Nat. Commun. 8, 1181 (2017)

O 66.14 Wed 18:00 Poster C

Al-based anodes with high reversible capacity for Li-ion battery — •Kangzhe Cao[1,2], Huiqiao Liu[2], Jiahui Ma[2], and Yong Lei[1] — [1]Fachgebiet Angewandte Nanophysik, Institut für Physik & IMN MacroNano, Technische Universität Ilmenau, 98693 Ilmenau, Germany — [2]College of Chemistry and Chemical Engineering, Xinyang Normal University, Xinyang 464000, China

Aluminum (Al) can alloy with Li at a lithiation potential of ˜0.2 V to form LiAl, offering a theoretical capacity of 993 mAh g-1. Moreover, the merits of high electronic conductivity, abundant resources, and environmental benignity endow the Al anode competitive for Li-ion batteries (LIBs). However, the natural Al2O3 passivation layer leads to a large voltage dip (0.1 V), which would interrupt the lithiation process and result in a limited capacity. Meanwhile, the volume expansion (97 %) always makes the electrode in an unstable structure. Herein, we demonstrate a neat avenue to replace the inactive Al2O3 layer with active Sn at room temperature and further encapsulate them into the carbon-based matrix for LIBs. Not any acid or tedious process is needed. Benefiting from the elimination of Al2O3 and the cooperation of carbon matrix, the as-prepared Al-based anodes (Al@Sn@GO pellets and Al@C-Sn pellets) exhibit little voltage dip (0.03 V), higher reversible capacities, and longer cycling life compared to the commercial Al pellet electrode. Considering the low-cost, energy-saving, and facile scalable preparation of the Al-based anodes, our work presents an attractive vision for the development of the practical Al-based anode.

O 66.15 Wed 18:00 Poster C

Optimization of incommensurate organic/inorganic interface structures to study superlubricity — •Lukas Hörmann[1], Johannes J. Cartus[2], and Oliver T. Hofmann[2] — [1]University of Warwick, Coventry, UK — [2]Graz University of Technology, Graz, Austria

Friction is a significant source of energy loss in mechanical devices. One way to reduce this loss is by achieving superlubricity * extremely low friction. Conventional wisdom suggests that incommensurate interface structures facilitate superlubricity. Accurately describing friction necessitates precise first-principles modelling of the interface structure, which is particularly challenging for organic/metal interfaces due to their tunability and propensity for incommensurate structures. However, simulations of incommensurate structures require large system sizes making such calculations intractable. We address this challenge by developing a machine-learned interatomic potential capable of accurately determining energies and forces for structures containing thousands to tens of thousands of atoms. With this approach, we quantify the breakdown of low-friction states in incommensurate structures caused by static distortion waves. Furthermore, we extract design principles to engineer incommensurate interface systems that suppress the formation of static distortion waves, enabling lower friction coefficients.

O 66.16 Wed 18:00 Poster C

Modification of Single-Walled Carbon Nanotubes Using MeV Heavy Ions — •Ayman Sherif El-Said[1], Rene Heller[2], and Stefan Facsko[2] — [1]Physics Department and Interdisciplinary Research Center for Advanced Materials, KFUPM, Dhahran 31261, Saudi Arabia — [2]Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf (HZDR), 01328 Dresden, Germany

Highly energetic heavy ions are used efficiently for tailoring the properties of various nanomaterials [1]. Among the recently investigated advanced materials, the carbon-based ones have attracted a lot of interests due to their promising properties for a wide variety of technological applications [2]. Here, we focus on the structural modifications of single-walled carbon nanotubes (SWCNTs) by irradiation with MeV iodine ions from the tandem accelerators at the Ion Beam Center of HZDR. The ion irradiation leads to fragmentation of the nanotubes into broken segments after surpassing of a specific ion-fluence threshold [3]. Moreover, the Raman spectroscopic measurements show that the disorder parameter increases as a function of ion fluence. Different approaches are discussed for the explanation of the mechanisms responsible for the observed ion-induced changes in SWCNTs.

[1] F. Aumayr, S. Facsko, A.S. El-Said, C. Trautmann, M. Schleberger, J. Phys.: Condens. Matter 23, 393001 (2011).

[2] F.L. Michael et al., Science 339, 535 (2013).

[3] A.S. El-Said, S. Rao, S. Akhmadaliev, S. Facsko, Phys. Rev. Applied 13, 044073 (2020).

O 66.17 Wed 18:00 Poster C

Surface engineering via an ultrathin alumina membrane for constructing stable sodium metal anodes — •Jiajia Qiu, Changfan Xu, Yu Duan, Huaping Zhao, and Yong Lei — Fachgebiet Angewandte Nanophysik, Institut für Physik & IMN MacroNano, Technische Universität Ilmenau, 98693 Ilmenau, Germany

Despite the high theoretical specific capacities (1166 mAh g-1) and low redox potentials (-2.71 V vs. normal hydrogen electrode) of Na metals, unstable solid electrolyte interphase (SEI) remains a major bottleneck due to the mossy or dendritic growth of Na in the repetitive stripping and plating process of sodium metal batteries. Owing to chemical and electrochemical stability during electrochemical plating and stripping, the Al2O3 coatings enable reduced Na dendrite growth. Simultaneously, the ultrathin alumina membrane (UTAM) network can change the traditional nucleation and growth modes from the origin, which can provide a porous 3D host to accommodate the large volume change of the Na anode. As a result, the UTAM network significantly improved Coulombic efficiency while avoiding short-circuit risks. This novel design may bring in new opportunities for next-generation high-performance Na metal batteries.

O 66.18 Wed 18:00 Poster C

Confinement and band dispersion of excited surface electrons in a two-dimensional porous network on a metallic surface — •Lukas Kau, Martin Mitkov, Ralf Hemm, Lyu Lu, Martin Anstett, Martin Aeschlimann, and Benjamin Stadtmüller — University of Kaiserslautern-Landau and Research Center OPTIMAS, Erwin-Schrödinger Str. 46, 67663 Kaiserslautern, Germany

Two-dimensional metal-organic networks (MONs) on noble-metal surfaces are highly tuneable porous architectures for designing the electronic properties of surfaces by quantum confinement. In this study, we investigate the electron confinement and the corresponding band structure of the excited surface electrons, i.e. the image potential state (IPS), in a self-assembled Cu-coordinated T4PT porous network on a Cu(111) surface. Using two-photon photoemission spectroscopy, we are able to map the band dispersion of the IPS, which reveals a remarkably flat band dispersion and a large effective band mass of approximately $2m_0$. Compared to the occupied Shockley electrons, the excited electrons in the IPS exhibit a strong confinement in the Cu-T4PT porous network [1]. This substantially different confinement of the Shockley surface and the IPS electrons is rooted in the vertical potential landscape of the network structure. Finally, we provide a first glimpse into the hot electron dynamics of the IPS.

[1] Lyu, Lu, et al. arXiv preprint arXiv:2307.06814 (2023)

O 66.19 Wed 18:00 Poster C

Structural Characterisation via X-ray Standing Wave Anal-

ysis: Reactions of Tetraphenyl Porphyrin — •Eleanor S. Frampton[1], Chris J. Judd[2], Matthew Edmondson[2], David A. Duncan[3], Robert G. Jones[2], and Alex Saywell[2] — [1]MAX IV Laboratory, Lund, Sweden — [2]School of Physics, University of Nottingham, UK — [3]Diamond Light Source, UK

Surface-confined reactions offer alternative synthetic pathways to solution-based chemistry and can be studied with a range of techniques to gain deep insights into the on-surface processes taking place during the chemical reaction. By employing a variety of analysis methods a route to understanding and characterizing the mechanistic details of these processes becomes available. This insight provides the tools required to potentially control the selectivity and efficiency of these reactions as well as to design systems with specific functionality.

Here I will focus on how the NIXSW technique can be utilized in combination with STM and XPS. The example system of tetraphenyl porphyrin (TPP) on Au(111) is used to describe how the combination of these methods can lead to a detailed model of an on-surface system, including adsorption geometries of individual molecules. This system consists of three distinct phases, resulting from consecutive on-surface reactions. The chemical specificity of XPS and NIXSW afford us insight into the chemical and structural changes exhibited by the nitrogen atoms within the TPP molecule at each reaction stage. [1,2]

[1] Woodruff, D. P. (2005), Rep. Prog. Phys., 68(4), 743 [2] Frampton, E. S....& Saywell, A. (2023), Inorg. Chim. Act., 558, 121718

O 66.20 Wed 18:00 Poster C

Imaging charge carriers generation process in 2D semiconducting perovskite material — •Dmitrii Sychev[1,4], Andrei Mitrofanov[1,2], Khrystyna Rymsha[1], Karina Zakirova[1], Petr Formanek[1], Darius Pohl[3], Eva Bittrich[1], Quinn Besford[1], Brigitte Voit[1,2], and Ilka Hermes[1] — [1]Leibniz-Institut für Polymerforschung Dresden e.V., Hohe Str. 6, 01069 Dresden, Germany — [2]Chair of Organic Chemistry of Polymers, Technische Universität Dresden, 01069 Dresden, Germany — [3]3Dresden Center for Nanoanalysis (DCN), Center for Advancing Electronics Dresden (cfaed), Technische Universität Dresden, 01069 Dresden, Germany — [4]Technische Universität Dresden, Helmholtztraße 10, Dresden 01062

Organic-inorganic halide perovskites have found their application as photodetectors. The transition from bulk to 2D material enables a greater variety of possible compositions as well as their functionalities.

Here, we investigated novel thin (nanometer scale) particles of halide perovskite semiconducting 2D material for their potential application as photodetectors. Kelvin probe force microscopy (KPFM) is well suited for this purpose, revealing local contact potential. KPFM measurements were performed under 405 and 458 nm illumination. The measurements show a significant decrease in the surface potential of the particles under light. This photoresponse demonstrates the suitability of the material for photodetection applications.

O 66.21 Wed 18:00 Poster C

Collective Radial Breathing Modes in Homogeneous Nanotube Bundles — •Charlotte Berrezueta-Palacios[1], Dekel Nakar[2], Anna Wroblewska[3], Oisín Garrity[1], Han Li[4,5], Nitzan Shadmi[2], Benjamin S. Flavel[4], Ernesto Joselevich[2], Stephanie Reich[1], and Georgy Gordeev[1,6] — [1]Department of Physics, Freie Universität Berlin, Germany. — [2]Department of Molecular Chemistry and Materials Science, Weizmann Institute of Science, Israel — [3]Faculty of Physics, Warsaw University of Technology, Poland — [4]Institute of Nanotechnology, Karlsruhe Institute of Technology, Germany. — [5]Department of Mechanical and Materials Engineering, University of Turku, Finland — [6]Department of Physics and Materials Science, University of Luxembourg, Luxembourg.

We present a Raman study of the collective vibrations arising from the homogeneous bundling of single-walled carbon nanotubes and analyze the dependence of their vibrational coupling on the tube diameter using two systems, single-walled carbon nanotube coils and a monochiral CNT film. We report on two breathing-like modes for quasi-infinite bundles, compared to the single radial breathing mode characteristic of isolated tubes. The exciton-phonon coupling in these modes is probed with resonant Raman spectroscopy, revealing the same resonance energy for both breathing-like peaks. Our experimental findings align well with previously reported theoretical studies, demonstrating a $1/d$ scaling for all modes, as well as confirming the relative shift of the modes dependent on intertube interaction.

O 66.22 Wed 18:00 Poster C

MBD+C: how to include metallic character (Type C non-additivity) into atom-based dispersion energy schemes — •Alberto Ambrosetti[1] and John Dobson[2] — [1]Università degli Studi di Padova (Italy) — [2]Griffith University (Australia)

Dispersion (van der Waals, vdW) interactions in low-dimensional metals are known to exhibit anomalous ”Type -C non-additivity” [Int. J. Quantum Chem. 114, 1157 (2014)] resulting in behavior that is missed by popular atom-based schemes for dispersion energy calculations. For example, the vdW interaction energy between parallel metallic nanotubes at separation D falls off as approximately D^{-2}, whereas current atom-based schemes predict D^{-5}. Here we show how to include Type C effects efficiently within atom-based schemes such as ”Many Body Dispersion” (MBD) and "universal MBD" (uMBD). We apply our technique to calculate the van der Waals interaction between parallel metallic chains of gold atoms.

O 67: Poster: Oxide, Insulator and Semiconductor Surfaces

Time: Wednesday 18:00–20:00 Location: Poster C

O 67.1 Wed 18:00 Poster C

DFT study of the adsorption of small molecules on a vicinal ZnO surface — •Marlene Sell and Bernd Meyer — Interdisciplinary Center for Molecular Materials and Computer Chemistry Center, FAU Erlangen-Nürnberg, Germany

Understanding the adsorption of reactants on the surface of a catalyst is not only of interest for exploring reaction mechanisms, but the adsorption itself can also alter the structure of the catalyst surface.

In this study, the adsorption of carbon dioxide, formaldehyde, and formic acid, three important intermediates in the synthesis of methanol from syngas, on the vicinal ZnO(10$\bar{1}$4) surface was investigated using density-functional theory (DFT). Four different structures of the surface were considered: two previously proposed truncated bulk terminations, both include 2-fold coordinated surface atoms, and two new structural model, which are found to be lower in energy due to the elimination of the 2-fold coordinated atoms by a shift of atomic rows. For each of the three molecules and four surface models, the energetically most favorable structure was determined and characterized by calculation of the XPS core level shifts.

In addition, the stabilization of the different surface models by a successively increasing number of water molecules was compared. The resulting adsorption energies show that once water molecules adsorb on the undercoordinated Zn surface atoms, the bulk-truncated surface structure becomes energetically more favorable than the structure with shifted atomic rows.

O 67.2 Wed 18:00 Poster C

A structural study of CO ligation to Cu adsorbed on Fe3O4 (001) — •Fulden Eratam[1], Benedikt Klein[1,2], Lena Puntscher[3], Matthew Stoodley[1,2], Alexander Generalov[4], Alexei Preobrajenski[4], Tien-Lin Lee[1], Gareth Parkinson[3], and David A. Duncan[1] — [1]Diamond Light Source, Didcot, UK — [2]University of Warwick, Coventry, UK — [3]TU Wien, Vienna, Austria — [4]MAX IV, Lund, Sweden

Single atom catalysis (SAC) utilizes atomically dispersed single species of metals to serve as isolated active sites on a support and offers a promising tool for maximizing catalytic activity, selectivity and stability. Iron oxides are a popular support for such catalysts (e.g. [Angew. Int. Ed. 132 (2020), 12968]), where X-ray absorption near edge spectroscopy (XANES) is typically used to identify the coordination state of the SAC. The (001) surface of single crystal magnetite (Fe3O4) has been shown to coordinate a high density of single metal atoms [Science 346 (2014), 1215]. We have previously demonstrated that CO coordination pulls these atoms out of the surface by 0.2 Å [J. Chem. Phys. 152 (2020), 051102]. The current work highlights the effect of one such CO distortion on the XANES data and presents Cu L-edge XANES of Cu single metal adatoms adsorbed on Fe3O4(001) before and after CO coordination. In the absence of CO, it is found that Cu adatoms produce a XANE spectrum very similar to that of Cu2O. Meanwhile, CO coordination is found to produce a significant enhancement in the intensity of a pre-edge feature, resolved at an energy range typical of CuO.

O 67.3 Wed 18:00 Poster C

Enhanced Infrared Reflectance-Absorbance Spectroscopy on Magnetite Surfaces via Platinum Nanoparticle Encapsulation — Eric Sauter[1], •Lachlan Caulfield[1], Christof Wöll[1], Johanna Reich[2], Sebastian Kaiser[2], and Barbara A.J. Lechner[2] — [1]Karlsruhe Institute for Technology — [2]Technical University Munich

This study delves into the surface science of magnetite (Fe3O4) using ultra-high vacuum (UHV) techniques, concentrating on platinum nanoparticles' (Pt NPs) encapsulation and its impact on Infrared Reflectance-Absorbance Spectroscopy (IRRAS) bands. Magnetite, widely explored for diverse applications, possesses unique surface properties that can be tailored for enhanced functionality. Pt NPs, known for catalytic prowess, were precisely deposited on the magnetite surface in UHV conditions, ensuring a uniform distribution. Advanced surface science techniques characterized resulting morphology and composition. Surface ligand infrared spectroscopy (SLIR) revealed significant enhancements in CO-specific infrared bands' intensity after Pt NP encapsulation, signifying a notable influence. The study elucidates mechanisms driving this enhancement, emphasizing Pt NPs' role in influencing vibrational modes on the magnetite surface. This research advances understanding of magnetite surface modification and systematically explores enhancement effects on infrared bands from Pt NP encapsulation. Implications extend to designing and optimizing catalytic and sensing applications, where surface reactivity and spectroscopic sensitivity are critical.

O 67.4 Wed 18:00 Poster C

AAO template-guided preparation of ordered ZnO nanopore arrays — •Yu Duan[1,2], Jiajia Qiu[1], Mo Sha[1], Huaping Zhao[1], and Yong Lei[1] — [1]Fachgebiet Angewandte Nanophysik, Institut für Physik & IMN MacroNano, Technische Universität Ilmenau, 98693 Ilmenau, Germany — [2]Center for Advanced Thin Films and Devices, School of Materials and Energy, Southwest University, Chongqing, 400715, China

The nanostructural design of zinc oxide (ZnO) has been of great interest as it can effectively affect its photoelectric conversion efficiency, photocatalytic activity, electrochemical activity, and other properties. In this work, we employed an anodic aluminum oxide (AAO) template-guided approach. The structural design and replication of the AAO template enabled the controlled growth of ZnO nanostructures with precise porous morphology. As a result, highly ordered ZnO nanopore arrays were prepared with tunable pore size and height. This unique array of ZnO nanopores may have potential applications in the fields of photovoltaic conversion, gas sensing, and water decomposition. In addition, the present synthesis strategy also has the potential for precise structural design of other metal and oxide materials.

O 67.5 Wed 18:00 Poster C

Initial stages of epitaxial growth of Fe_3O_4 films on $SrTiO_3$(111) — •Luis Witte, Tobias Pollenske, and Joachim Wollschläger — Institute of Physics, University of Osnabrück, Barbarastrasse 7, 49076 Osnabrück, Germany

Magnetite films hold promising perspectives in spintronics as, e.g., MRAM (Magnetic Random Access Memory) devices. MRAMs, based on magnetic tunnel junctions, promise non-volatile storage with improved energy efficiency and extended battery life for electronic devices. The ferromagnetism-controlled spin polarization of electrons in magnetite enables precise switching operations. Specific adjustments to the layer properties can be achieved by selectively varying the deposition temperature.

In this study, ultrathin Fe_3O_4 layers were deposited on $SrTiO_3$ (111) using reactive molecular beam epitaxy evaporating in $5 \cdot 10^{-6}$mbar O_2 with deposition temperatures from 200°C to 600°C. The focus of this work is on the temperature-dependent growth modes of magnetite during the initial stages of growth. LEED and XPS measurements were conducted after each step to gain insights into the structural and chemical compositions of the magnetite layers within the first monolayers. Upon completion of the growth process, additional XRR and (HE)XRD measurements were performed to obtain information about the structure and final thickness of the film.

O 67.6 Wed 18:00 Poster C

Polaron formation on $BaTiO_3$ surfaces induced by oxygen vacancies — •Andreas Ziegler and Bernd Meyer — Interdisciplinary Center for Molecular Materials and Computer Chemistry Center, FAU Erlangen-Nürnberg, Germany

$BaTiO_3$ is widely used as substrate and recently found interest in catalysis for oxidative coupling reactions. For these applications it is important to understand its surface properties in an oxygen-deficient state. While the localization of electrons due to oxygen vacancies has been studied intensely for bulk $BaTiO_3$, excess electrons on the surfaces of $BaTiO_3$ have not been elucidated yet.

We applied spin-polarized density functional theory (DFT) with an on-site Hubbard+U correction to determine the behavior of defect electrons created by oxygen vacancies on the Ba- and Ti-terminated $BaTiO_3$(001) surface. By applying atomic distortions before geometry optimization we achieved different localization/delocalization pattern of the two defect electrons. We find that localization of both electrons is energetically always more favorable than solutions with delocalized electrons. On the Ba-terminated surface, in the lowest-energy configuration, one electron is localized within the oxygen vacancy on the surface and the other on a Ti ion in the subsurface layer. This is in contrast to the bulk, where both defect electrons preferentially localize on neighboring Ti sites. On the Ti-terminated surface, strong surface relaxations occur, hindering an electron to localize in the vacancy and allowing localization only on two specific Ti sites.

O 67.7 Wed 18:00 Poster C

In situ X-ray photoelectron spectroscopy study of atomic layer deposited ceria on SiO_2: substrate influence on the reaction mechanism during the early stages of growth — •Max Gertig, Carlos Morales, Karsten Henkel, and Jan Ingo Flege — Applied Physics and Semiconductor Spectroscopy, Brandenburg University of Technology Cottbus-Senftenberg, Konrad-Zuse-Straße 1, 03046 Cottbus, Germany

Atomic layer deposition (ALD) is known to produce amorphous and defect-rich films in a layer-by-layer fashion, which can potentially give rise to unexpected material properties. In particular, ultrathin films (few monolayers) will show the highest complexity, as the substrate-material interaction will play a major role during deposition. Therefore, it is crucial to understand the early stages of growth of the ALD process to control and potentially tailor this interfacial interaction. Applying a surface science approach combined with complementary ex-situ characterization, we have studied by in-situ X-ray photoelectron spectroscopy (XPS) the early stages of ceria (CeO_x) growth on SiO_2 substrates deposited by thermal-ALD using $Ce(thd)_4/O_3$. Interestingly, an initial mixture of Ce^{3+} and Ce^{4+} was observed, although only Ce^{4+} may be expected considering the used precursor and oxidant. This fact, together with a deviation from the ideal layer-by-layer growth and a higher growth rate during the first cycles, indicates a significant influence of the substrate of the ALD reaction mechanism as well as a correlation between morphology and ceria oxidation state.

O 67.8 Wed 18:00 Poster C

Adsorption of methanol on Si(001) revisited: alternative reaction pathways at increased surface temperature — •Sophie Göbel[1], Timo Glaser[1], Gustav Nolte[1], Denys Jockel[1], Christian Länger[1], Julian Heep[1], Hendrik Weiske[2], Jan-Niklas Luy[2], Ralf Tonner-Zech[2], and Michael Dürr[1] — [1]Institut für Angewandte Physik and Zentrum für Materialwissenschaften, Justus-Liebig-Universität Giessen, Germany — [2]Wilhelm-Ostwald-Institut für Physikalische und Theoretische Chemie, Universität Leipzig, Germany

Molecular electronics is seen as a possibility to advance semiconductor technology beyond Moore's law. With silicon as the technologically most important substrate, understanding the interaction between organic molecules and silicon surfaces is crucial for tailoring the properties of the required organic/inorganic interfaces. While low energy pathways have been extensively studied in the past, our research focuses on the question if channels with higher barriers can be addressed at higher surface temperatures. In this context, we studied the binding of methanol, the simplest alcohol, on Si(001) using X-ray photoelectron spectroscopy, scanning tunneling microscopy, and DFT calculations. Exploring this in principle simple system reveals a multitude of possible reaction channels spanning a wide range of energy barriers. However, experimentally we find the initial methanol adsorption always to proceed via O-H dissociation and further reaction channels to be operative only in the further course of the reaction.

O 67.9 Wed 18:00 Poster C

Preparation and characterization of P- and In-rich AlInP(100) by XPS and in-situ RAS — •Mohammad Amin Zare Pour, Sahar Shekarabi, Agnieszka Paszuk, and Thomas Hannappel — Grundlagen von Energiematerialien, Institut für Physik, Technische Universität Ilmenau

n-AlInP(100) is commonly used for a selective transport of electrons in so-called window layers in high-efficiency III-V multijunction solar or photoelectrochemical (PEC) cells. A fundamental understanding of water interaction with the window layer is crucial for PEC applications as well as for atomic layer deposition of passivation layers such as TiO2. In this study we focus on the preparation of different surface reconstructions of AlInP(100) by metalorganic chemical vapour deposition (MOCVD) utilizing in-situ reflection anisotropy spectroscopy (RAS) and benchmarking of the in-situ signals by X-ray photoelectron spectroscopy (XPS) and low energy electron diffraction (LEED). Our findings show that annealing the AlInP(100) surface at 310 °C leads to a P-rich surface reconstruction displayed by a (2x1)-like LEED pattern, while annealing it at 440 °C forms an In-rich surface reconstruction with a diffuse (2x4) LEED pattern. XPS results show the presence of P-P dimers on the surface of P-rich AlInP, while In-In bonds were resolved on In-rich surfaces. We determine a band bending of 1.0 eV and 1.7 eV toward the surface in the band alignment of P- and In-rich AlInP surfaces, respectively, indicating that In-rich surface has more surface states.

O 67.10 Wed 18:00 Poster C

Structure and Stability of Al-alloyed β-Ga_2O_3(100) surfaces — •Ming-Chao Kao[1,4], Lukas Paul Schewe[2], Arub Akhtar[3], Jana Rehm[3], Saud Bin Anooz[3], Zbigniew Galazka[3], Andreas Popp[3], Jan Ingo Flege[2], Vedran Vonk[1], and Andreas Stierle[1,4] — [1]Centre for X-ray and Nanoscience, Deutsches Elektronen-Synchrotron, Hamburg, Germany — [2]Applied Physics and Semiconductor Spectroscopy, Brandenburg University of Technology Cottbus, Germany — [3]Leibniz-Institut für Kristallzüchtung , Berlin, Germany — [4]Department of Physics, University of Hamburg

β-Ga_2O_3 (GaO) holds great promise in high-power applications. GaO is predicted to outperform the leading technologies based on SiC and GaN due to a three times higher calculated critical field strength. It has been identified that alloying with Al results in an almost linear increase in the bandgap between Ga_2O_3 (4.85eV) and Al_2O_3 (7eV). A critical issue in the bandgap engineering of GaO is maintaining reasonable electron mobility, which is low for the pure material $(150cm^2/V/S)$. This project aims to explore GaO by Al alloying, which allows tunable wide bandgaps for engineering material properties and at the same time maintains excellent crystal quality. PXRD and their Rietveld refinement on several AlGaO crystals indicate a preferential ordering of Al and Ga over octahedral and tetrahedral sites. The results of an SXRD study using synchrotron radiation addressing the surface structure of β-$Al_xGa_{2-x}O_3$(100) substrates with $x = 0 - 0.25$. Our results indicate that the GaO(100) surface is nearly bulk terminated and remains smooth up to 800°C and 10^{-5}mbar of molecular oxygen.

O 68: Poster: Scanning Probe Techniques: Method Development

Time: Wednesday 18:00–20:00 Location: Poster C

O 68.1 Wed 18:00 Poster C

GXSM4 - an update — Percy Zahl[1] and •Thorsten Wagner[2] — [1]Center for Functional Nanomaterials, Brookhaven National Laboratory, USA — [2]Institute of Experimental Physics, Johannes Kepler University Linz, Austria

GXSM is an open-source software project with a history of more than 25 years[1−−3] and a worldwide user community[4] that provides a state-of-the-art control software for all types of scanning probe techniques (STM, AFM, ...). The fourth generation of this versatile tool is based on the latest GNOME/linux desktop and a digital signal processor (DSP). The build-in General Vector Probe (GVP) enables freely programmable spectroscopy (e.g. IV in case of STM, FZ in case of NC-AFM, ...) and manipulation/probing. Up to three arbitrary wave forms can directly control the coarse motion of the SPM (Besocke/Beetle, Koala, ...). The embedded Python interface can perform even more complex and automated tasks. Thus, it is only a small step to achieve AI-based image recognition and processing (via OpenCV) and automated scanning control.[5] We will summarize the recent achievments and give an outlook on future plans (e.g. RedPitaya/FPGA based hardware), but we are also happy to discuss issues and technical requirements from the community.

[1] P. Zahl et al. in Rev. Sci. Instrum. 74, 1222–1227 (2003)
[2] P. Zahl et al. in J. Vac. Sci. Technol. B28, C4E39 (2010)
[3] P. Zahl et al. in Imaging and Microscopy, 17, 38-41 (2015)
[4] gxsm.sf.net and github.com/pyzahl/Gxsm4
[5] Arias et. al. in J. Phys. Chem. A 127, 5959–6134 (2023)

O 68.2 Wed 18:00 Poster C

Enhancing AFM image analysis through machine learning with style translation and data augmentation — •Jie Huang[1], Niko Oinonen[1], Lauri Kurki[1], and Adam S. Foster[1,2] — [1]Department of Applied Physics, Aalto University, Helsinki, Finland — [2]WPI Nano Life Science Institute (WPI-NanoLSI), Kanazawa University, Kakuma- machi, Kanazawa, Japan

Atomic Force Microscopy (AFM) is critical for atomic-scale nanostructure characterization. Simulations, especially using Particle Probe AFM (PPAFM), provide a cost-effective means for rapid image generation. Leveraging state-of-the-art machine learning models and substantial PPAFM-generated datasets, properties like molecular structures, electrostatic force potential, and molecular graphs can be accurately predicted using AFM images from simulations or experiments. However, transferring model performance from PPAFM to real AFM images poses challenges due to the subtle variations in real experimental data compared to the seemingly flawless nature of simulations. Our study explores Cycle GANs for style translation to augment data and improve the predictive accuracy of machine learning models in surface property analysis. Focused on mitigating the gap between simulated PPAFM and authentic AFM images, we optimize hyperparameters, showcasing the method's effectiveness through paired data comparisons. This research promises valuable insights, providing a novel approach to enhance machine learning model efficiency in the absence of abundant experimental data.

O 68.3 Wed 18:00 Poster C

On the origin and elimination of cross coupling between tunneling current and excitation in scanning probe experiments that utilize the qPlus sensor — Michael Schelchshorn, •Fabian Stilp, Marco Weiss, and Franz J. Giessibl — University of Regensburg, Institute of Experimental and Applied Physics, Universitätsstrasse 31, D-93040 Regensburg, Germany

The qPlus sensor allows for the simultaneous operation of scanning tunneling microscopy (STM) and atomic force microscopy (AFM). When operating a combined qPlus sensor STM/AFM at large tunneling currents, a hitherto unexplained tunneling current-induced cross coupling can occur, which has already been observed decades ago. Here, we study this phenomenon both theoretically and experimentally; its origin is voltage drops on the order of μV that lead to an excitation or a damping of the oscillation, depending on the sign of the current. Ideally, the voltage drops would be phase-shifted by $\pi/2$ with respect to a proper phase angle for driving and would, thus, not be a problem. However, intrinsic RC components in the current wiring lead to a phase shift that does enable drive or damping. Our theoretical model fully describes the experimental findings, and we also propose a way to prevent current-induced excitation or damping.

O 68.4 Wed 18:00 Poster C

Sample preparation for STM analysis — •Caroline Hommel, Lukas Spree, Luciano Colazzo, and Andreas Heinrich — Center for Quantum Nanoscience, 52 Ewhayeodae-gil, Daehyeon-dong, 03760 Seoul, South Korea

The process of sample preparation within ultra-high vacuum (UHV) settings crucially impacts the success of scanning tunneling microscopy (STM) experiments. While the sublimation of atoms and thermally stable, volatile molecules provides a pristine avenue to introduce analytes onto UHV surfaces, challenges arise when compounds lack volatility or thermal stability. In such instances, alternative methods become essential. Studies highlight solvent-based preparations like dropcasting as potentially beneficial, though they necessitate preparation outside the UHV chamber. An alternative, the electro spray deposition technique, proves effective by enabling molecules to traverse multiple pumping stages into UHV environments out of a solvent.

This technique's success hinges upon several critical parameters, including distance to the sample, solvent type, concentration, pressure, and duration. We investigate these parameters using functionalized fullerenes. Transitioning from bulk sample characterization of these intriguing compounds to investigating thin layers or individual molecules demands innovative methodologies to regulate interactions with the underlying substrate and effectively orientate the encapsulated species.

O 68.5 Wed 18:00 Poster C

Molecular behaviour during SPM tip functionalization for Scanning Quantum Dot Microscopy — •Paul Laubrock, Tim Dierker, and Philipp Rahe — Universität Osnabrück

Scanning quantum dot microscopy (SQDM) [1] is a variant of scanning probe microscopy (SPM) that enables the quantitative mapping of the electrostatic potential at the atomic scale [2]. A prerequisite for SQDM is the SPM tip functionalization with a (molecular) quantum dot. We developed an instructive flowchart for efficient preparation of functionalized SQDM tips based on scanning tunneling microscopy (STM) manipulation experiments at 5K with 3,4,9,10-perylene-tetracarboxylic-dianhydride (PTCDA) molecules on Ag(111). Here we discuss experimental observations regarding the molecular behaviour in the tip-sample gap. In particular, we investigate an apparent instability in the STM signals appearing with a particular imaging contrast after lateral manipulation of a single PTCDA molecule. We interpret this and further observations in the context of dynamic bond formation/breaking in the surface-molecule-tip system.

[1] C. Wagner et al., PRL 115, 026101 (2015)

[2] C. Wagner et al., Nat. Mater. 18, 853 (2019)

O 68.6 Wed 18:00 Poster C

High-resolution characterization of single-step motion in piezoelectric friction-inertia walkers — •Felix Huber, Susanne Baumann, and Sebastian Loth — University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany

High precision positioners are a prerequisite for the realization and automation of cutting edge research experiments. Piezoelectric friction-inertia walkers offer the combination of high accuracy movement on the millimeter scale with a resolution on the nanometer scale and are essential for scanning probe microscopy (SPM) applications. The behavior and reliability of nanopositioners based on the friction-inertia principle strongly depends on the respective use of contact surface materials.

Here, we use optical interferometry to monitor the single step motion of a linear piezoelectric friction-inertia walker on the nanometer scale. We test different combinations of metallic and ceramic contact surface materials as well as contact surface geometries and identify material combinations that offer large step sizes or reliable single-step motion which can be implemented easily in home-built SPM setups.

O 68.7 Wed 18:00 Poster C

Automated tip preparation for electron spin resonance measurements in STM — •Lovis Hardeweg, Cathrin Haab, Wantong Huang, Paul Greule, Máté Stark, Christoph Sügers, Wolfgang Wernsdorfer, and Philip Willke — Physikalisches Institut (PHI), Karlsruhe Institute of Technology, Karlsruhe, Germany

The development of scanning tunnelling microscopy (STM) combined with electron spin resonance (ESR) is a powerful technique to image and investigate spins on surfaces [S. Baumann et al. Science 350.6259 (2015), pp. 417-420]. However, it relies on spin-polarized tips and their preparation often requires time-consuming and repetitive work. Here, we present a multi-step routine based on supervised machine learning that autonomously creates ESR-active tips. It includes locating magnetic Fe atoms on MgO/Ag, MgO-layer thickness determination, tip evaluation/conditioning, as well as drift correction. Besides, the number of MgO monolayers on Ag is automatically determined and the individual Fe atoms are picked up onto the tip. Finally, the routine evaluates the ESR capabilities of the tip.

O 68.8 Wed 18:00 Poster C

Development and implementation of data model for scanning tunneling microscopy and scanning tunneling spectroscopy — •Yichen Jin[1], Rubel Mozumder[1], Yan Wang[2], Jürgen P. Rabe[1,3], Heiko B. Weber[4], Tamás Haraszti[5], Sabine Maier[4], Carlos-Andres Palma[2,3], Sandor Brockhauser[1], and Claudia Draxl[1] — [1]Humboldt-Universität zu Berlin, Berlin, Germany — [2]Institute of Physics, Chinese Academy of Sciences, Beijing, PR China — [3]Department of Physics & IRIS Adlershof, Humboldt-Universität zu Berlin, Berlin, Germany — [4]Department of Physics, Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany — [5]Leibniz-Institut für Interaktive Materialien e.V., RWTH, Germany

Since the first realization of scanning tunneling microscopy (STM) and scanning tunneling spectroscopy (STS), a wide range of applications has been developed by using the technique of quantum tunneling effect. The community aims at organize experimental data along FAIR principles, thereby developing or using a standard vocabulary of metadata. NeXus allows to generate such a sort of data modelling and with that we have developed specific data model called NX_sts for STM/STS experiment. Our work focuses not only on storing data and metadata from STM/STS experiments in terms of the NX_sts vocabulary but also supports handling data-analysis results. In this presentation, we will show how metadata can be converted into the NeXus data format. We will detail the basic software architecture of reader that transforms metadata file into STM/STS specific data model which in terns follows NeXus data format concepts.

O 68.9 Wed 18:00 Poster C

Towards light-wave scanning tunneling microscopy in strong magnetic fields — Victoria Ruckerbauer, •Felix Giselbrecht, Andreas Rank, Christoph Rohrer, Leo Ringer, Michael Betz, Christian Meineke, Jakob Schlosser, Rupert Huber, and Jascha Repp — Department of Physics, University of Regensburg, Regensburg, 93040, Germany

In light-wave-driven scanning tunneling microscopy (LW-STM) the tunneling junction is biased by an ultrashort laser pulse instead of having a static voltage [1]. This method enables combined Ångström and femtosecond spatiotemporal resolution as demonstrated by the direct observation of the ultrafast motion of a molecular orbital [2]. Combining this development with a tunable magnetic field would allow to follow spin dynamics – spin precession, for example – in molecules and other atomic structures with single-electron sensitivity. To this end, a novel LW-STM is being developed with an external magnetic field to resolve single-spin dynamics with atomic spatial and ultrafast temporal resolution. We will discuss the instrumental challenges of this development and present the resulting instrument design including the STM head, the laser source as well as the solution to introduce the laser transient from outside the vacuum system to the STM junction. [1] Cocker *et al.*, Nature Photonics **7**, 620 (2013), [2] Cocker *et al.*, Nature **539**, 263 (2016).

O 68.10 Wed 18:00 Poster C

Non-contact AFM experiments with a diamond tip equipped needle sensor: From field emission to tunneling — •Stefan Schulte[1,2], Sven Just[1,3], Victor I. Kleshch[4], F. Stefan Tautz[1,3], and Ruslan Temirov[1,2] — [1]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, Germany — [2]II. Physikalisches Institut, Universität zu Köln, Cologne, Germany — [3]Fundamentals of Future Information Technology, Jülich Aachen Research Alliance (JARA), Jülich, Germany — [4]Moscow, Russia

A length extension quartz resonator (needle sensor) equipped with a CVD-grown single crystal diamond needle on the tip is characterized in a room-temperature AFM setup at UHV conditions. The particular setup allows for simultaneous AFM, field emission and tunneling experiments. The diamond tip needle sensor could be a platform to enable the measurements of electrostatic surface potential in scanning quantum dot microscopy (Wagner et al., Phys. Rev. Lett., 2015). We focus on the transition from a non-conductive state of the diamond to a surface-conductive state, which occurs when the surface of the diamond needle is transformed into amorphous carbon at elevated field emission currents. The current-voltages characteristics of the diamond needle are recorded at different distances ranging from the attractive AFM regime to several nm above the surface. Simultaneously recorded AFM signals of the resonant behavior of the sensor reveal the specific transformation processes. Finally, the conductive diamond tip enables tunneling experiments at comparably low bias voltages.

O 68.11 Wed 18:00 Poster C

Development of a Fast In Situ Scanning Tunneling Microscope for Studies of Electrochemical Interfaces — •Knud Schröter, Fabian Schröfel, Matthias Greve, Karsten Tarhouni, and Olaf Magnussen — Institut für Experimentelle und Angewandte Physik, Kiel

The atomic-scale understanding of processes at the interface between solid electrodes and liquid electrolytes is of high importance for electrochemical energy storage and conversion. Electrochemical scanning tunneling microscopy (ECSTM) is a key technique for the investigation of these interfaces and as such, it has seen widespread use. However, image acquisition in conventional ECSTM is a rather slow process, requiring tens of seconds or minutes per image. To help understand the precise reaction mechanisms of atomic and molecular species at solid-liquid interfaces, their movement and interactions need to be resolved.

For this, high-speed STMs capable of operating at rates >10 images per second are necessary. However, this technique has not been widely employed, mainly because of the instrumental requirements.

In this contribution, we present a new high-speed ECSTM developed and built in our group based on a Nanonis SPM controller. The instrument includes a novel scanner design with two independent piezo stacks for slow wide-range motion and local high-speed motion and a custom high-bandwidth preamplifier integrated into the scan head as close as possible to the tip. We will show first results on the implementation of the fast imaging mode.

O 68.12 Wed 18:00 Poster C

Automated STM imaging and spectroscopy: porphyrin molecular arrays as an exemplar — •Matthew Edmondson and Alex Saywell — School of Physics & Astronomy, University of Nottingham, UK

Materials analysis via scanning probe microscopy (SPM) is often labour intensive and not standardised to the level of the procedures employed within characterisation techniques such as nuclear magnetic resonance (NMR) and mass spectrometry. While SPM approaches provide impressive atomic/sub-molecular spatial resolution, [1] and allow characterisation of electronic states, a key challenge is to obtain statistically sufficient measurements over the panoply of features present on the material under study. Molecular overlayers, specifically the often-studied porphyrin-based assemblies on metals [2], provide a test-platform for the automated spatial and electronic characterisation of a molecule-substrate system. Porphyrin molecules may undergo various on-surface process (e.g. ring-closing, metalorganic coordination, and self-metalation) with a resultant change in electronic states potentially offering a 'fingerprint' of the chemical and structural changes.

Here, we present details of a semi-automated approach to characterising a molecule-substrate system. Custom SPM imaging and spectroscopy routines have been developed to interface with commercial control electronics and provide a route towards automated data collection.

[1] A. Sweetman, N.R. Champness, A. Saywell, Chem. Soc. Rev., 2020, 49, 4189 [2] Gottfried, J. M., Surf. Sci. Rep., 2015, 70, 259

O 69: Poster: Spin Phenomena in Chiral Molecular Systems

Time: Wednesday 18:00–20:00 Location: Poster C

O 69.1 Wed 18:00 Poster C

Self-assembly of the chiral donor-acceptor molecule DCzDCN on Cu(100) — Robert Ranecki, •Stefan Lach, and Christiane Ziegler — RPTU Kaiserslautern - Landau and Research Center OPTIMAS, Erwin-Schrödinger-Str. 56, 67663 Kaiserslautern, Germany

The twisted donor-acceptor molecule DCzDCN (5-(4,6-diphenyl-1,3,5-triazine-2-yl)benzene-1,3-dinitrile) shows an interesting self-assembly behavior on Cu(100). This D-A molecule is of particular interest as a system that uses reverse intersystem crossing (rISC) to massively increase the fluorescence yield. The orientation of the molecules with the donor part towards the substrate results in a chiral resolution at the interface, forming only homochiral phases. It ranges from tetramers in the submonolayer region to 1D structures near monolayer coverage, with a slight excess of the M-(-) dimer-determined phases. Using a multi-method approach with STM/STS and photoelectron spectroscopy, the topography and the molecular electronic properties of the DCzDCN/Cu(100) system are studied globally and locally. The appearance of hybrid interface states (HIS) will be discussed [2].

[1] Y. J. Cho et al. Adv. Mater. 26, 4050 (2014) [2] R. Ranecki et al. arXiv:2310.19534 [cond-mat.mtrl-sci]

O 69.2 Wed 18:00 Poster C

Multi-scale simulation of structural motifs in assemblies of helical molecules — Kevin Preis[1], •Sibylle Gemming[1], Jeffrey Kelling[1], and Florian S. Günther[2] — [1]Instute of Physics, TU Chemnitz, Chemnitz, Germany. — [2]UNESP, Rio Claro, Brazil.

Helical polypeptide molecules sparked interest as candidate materials for studying spintronic effects on the nano-scale. As charge carriers are transmitted through assemblies of helical molecules, spin polarization of the transmitted current has been detected by scanning tunneling spectroscopy and attributed to a polarization dependent tunneling resistance or a spin-selective propagation along the backbone of the helical molecule. For single molecules there is, however, debate, whether the helical symmetry break alone or also the adsorption-induced surface polarization for films on metallic substrates decisively influences the polarization direction [1]. The present simulations, address potentially beneficial effects of aggregate formation from helical molecules depending on the type of helicity, the orientation on the surface and the type and strength of the molecule-surface interaction. For polyalanine (PA) we studied the lateral*ordering between PA homo- and heterochiral pairs by van-der-Waals-corrected DFT-based tight binding calculations. Denser packing geometries can be reached by heterochiral PA pairs. Furthermore, coarse-grained classical potentials were derived from the DFTB data, and the different PA phases seen in STM were also successfully obtained from Monte-Carlo simulations. [1] Aragones et al., small 2017, 13, 1602519.

O 69.3 Wed 18:00 Poster C

Pinning of magnetic domains and skyrmions by adsorption of chiral polypeptides — Yael Kapon[1], •Theo Balland[2], Fabian Kammerbauer[2], Mathias Kläui[2], Shira Yochelis[1], Mathias Kläui[2], and Yossi Paltiel[1] — [1]Institute of Applied Physics, Faculty of Sciences, The Hebrew University of Jerusalem,Jerusalem 9190401, Israel — [2]Institute of Physics, Johannes Gutenberg University, Staudingerweg 7, 55128 Mainz, Germany

Spin currents are of intrinsic interest to the magnetism community due to its ability to influence magnetic states. A high degree of spin polarization can be achieved by chirality-induced spin selectivity (CISS) [1]. Here we investigate the impact of adsorbed chiral polypeptides on the pinning of magnetic domains [2]. The examined structure comprises chiral polypeptide α-helix polyalanine adsorbed on a Ta(5)/CoFeB(0.9)/MgO(2)/Ta(2)/Au(5) thin film, exhibiting perpendicular magnetic anisotropy, along with a Ti(2)/Ni(80)/Au(5) thin film. Utilizing Magneto-Optic Kerr Effect microscopy, the magnetic structures are visualized, and the surface magnetization is quantified. Magnetic imaging of domain walls unveils distinctions between chiral and achiral molecules adsorbed on the surface. Comparative analysis of the in-plane magnetization curves of Ti/Ni/Au/chiral molecules enables the observation of the rotation of the easy axis of magnetization. This rotation is attributed to the alignment of magnetization with the spatial orientation (tilt angle) of the molecules [2].

[1] R. Naaman et al., Nat. Rev. Chem. 3, 250-260 (2019)

[2] Y. Kapon et al., J. Chem. Phys. 159, 064701 (2023)

O 69.4 Wed 18:00 Poster C

Theory for phonon-mediated chirality-induced spin selectivity — •Namgee Cho, Thibaut Lacroix, James Lim, Susana F. Huelga, and Martin B. Plenio — Institut für Theoretische Physik, Universität Ulm, D-89081 Ulm, Germany

Chirality-induced spin selectivity (CISS) is an intriguing phenomenon where the chirality of organic molecules induces significant spin selectivity in various electronic processes, such as bound electron transport, photoelectron transmission and chemical reactions. Although the CISS effects have been demonstrated in various experiments, the underlying mechanism is still unknown. Recently, the influence of electron-phonon interaction on CISS has attracted considerable interest. However, the previous theoretical studies have considered simplified models where phonon degrees of freedom are treated classically or approximately described by a single quantum harmonic oscillator. In this work, we consider a pseudo-mode theory where multiple quantum harmonic oscillators under Lindblad noise effectively describe non-Markovian phonon baths inducing fluctuations in electronic parameters of chiral molecules. We investigate how the electron-phonon interaction leads to CISS effects in charge separation and how the degree of spin polarization depends on environmental parameters.

O 69.5 Wed 18:00 Poster C

Chiral-Induced Spin Selectivity and Non-equilibrium Spin Accumulation in Molecules and Interfaces: A First-Principles Study — •Sumit Naskar[1], Vladimiro Mujica[2], and Carmen Herrmann[1] — [1]Institute for Inorganic and Applied Chemistry HARBOR Bldg. 610 Luruper Chaussee 149, 22761 Hamburg, Germany — [2]Arizona State University, School of Molecular Sciences, PO Box 871604, Tempe, Arizona 85287-1604, USA

Electrons moving through chiral molecules are selected according to their spin orientation and the helicity of the molecule, an effect known as chiral-induced spin selectivity (CISS)[1-2]. To help elucidate the underlying mechanism, a non-equilibrium Green's function method, combined with a Landauer approach and density functional theory, is applied to carbon helices contacted by gold electrodes, resulting in spin polarization in the non-equilibrium electronic structure of the junctions[3]. While this spin polarization is small calculated with a pure exchange-correlation functional, its sign changes with the direction of the current and with the handedness of the molecule[3]. Previous studies as well as our results suggest that computationally more expensive hybrid functionals may lead to considerably larger spin polarization in the electronic structure[4]. Thus non-equilibrium spin accumulation could be a key component in understanding the CISS mechanism.

References: [1] Goehler B. et. al., Science 2011, 6019, 894.; [2] Naaman R. et al., J. Phys. Chem. Lett. 2020, 11, 3660. [3] Naskar S. et al., J. Phys. Chem. Lett. 2023, 14, 694. [4] Zoellner M. et al., J. Chem. Theo. Comput. 2020, 16, 7357

O 70: Poster: Spins on Surfaces & Surface Magnetism

Time: Wednesday 18:00–20:00 Location: Poster C

O 70.1 Wed 18:00 Poster C

Weakly coupled S=1/2 molecules on a superconductor — •Vivek Krishnakumar Rajathilakam[1], Nicolaj Betz[1], Lukas Arnhold[1], Matteo Briganti[2], Lorenzo Poggini[4], Jaisa Fernandez Soarez[5], Luana C. De Camargo[5], Giulia Serrano[3], Matteo Mannini[2], Federico Totti[2], Roberta Sessoli[2], Susanne Baumann[1], and Sebastian Loth[1] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]Department of Chemistry Ugo Schiff, University of Florence, Italy — [3]Department of Industrial Engineering, University of Florence, Italy — [4]Institute of Chemistry of Organometallic Compounds - CNR, Florence, Italy — [5]Department of Chemistry, Federal University of Paraná, Brazil

Sandwich complexes hosting a transition metal ion can show magnetic properties of great interest for quantum information processing [1]. Here we study an S=1/2 organometallic sandwich CpTi(cot), with very long coherence times in frozen solution [2]. We use a Scanning Tunneling Microscope (STM) at 50 mK to measure the properties of individual CpTi(cot) molecules adsorbed on a superconducting vanadium surface V(100) with oxygen reconstruction. It shows no Yu-Shiba-Rusinov states, but prominent S=1/2 spin excitations can be traced up to 15 T. They show spin-spin interactions for molecules with separation up to 1.4 nm. These results show a promising step towards using molecules on superconductors for molecular qubit applications. [1] AM. Ariciu, et al. Nat Commun 10, 3330 (2019) [2] L.C. de Camargo, et al. Angew. Chem., Int. Ed., 60, 2588 (2021)

O 70.2 Wed 18:00 Poster C

Controlling Yu-Shiba-Rusinov States with a Molecular Switch — •Arnab Banerjee, Niklas Ide, Richard Berndt, and Alexander Weismann — Instituts für Experimentelle und Angewandte Physik, CAU Kiel

Tin phthalocyanine (SnPc) is a non-planar diamagnetic molecule. On metal surfaces, it is geometrically bistable and transitions may be induced by electron injection. We studied SnPc on superconducting Pb(100) using low-temperature scanning tunneling spectroscopy. Yu-Shiba-Rusinov (YSR) states are observed upon assembly of the molecules into supramolecular arrays indicating the presence of a molecular spin. Moreover, the effect of geometrical switching on the YSR state is demonstrated.

O 70.3 Wed 18:00 Poster C

Quantum simulator to emulate lower dimensional physics and chemistry — Emil Sierda, Xiaochun Huang, Danis Badrtdinov, Brian Kiraly, Elze Knol, •Anna M. H. Krieg, Gerrit C. Groenenboom, Mikhail I. Katsnelson, Malte Rösner, Daniel Wegner, and Alexander A. Khajetoorians — Institute for Molecules and Materials, Radboud University, Nijmegen, The Netherlands

Designing materials with tailored physical and chemical properties requires a quantitative understanding of interacting quantum systems. In order to provide predictability, a promising route is to create bottom-up platforms, where the electronic properties of individual and interacting atoms can be emulated in a tunable manner. Here, we present a quantum simulator based on patterned Cs ions embedded in a 2DEG on the surface of semiconducting InSb(110). We use this platform to emulate the structure and orbital landscape of planar organic molecules. Using STM/STS and ab initio calculations, we show that an artificial atom can be derived from localized states of patterned Cs ions. The resultant potential can be used to couple artificial atoms, leading to bonding and anti-bonding states as well as to different orbital symmetries. Based on these artificial orbitals, we emulate molecular orbitals of known organic molecules based on various atomic structures. In a different limit where Cs atoms are much closer, this quantum simulator can also be used to probe the effect of e-e interactions. Our experimental data suggest that dense structures exhibit many-body effects which can be extended to complex quantum states based on arbitrary lattices.

O 70.4 Wed 18:00 Poster C

Non-contact layer stabilization of azafullerene radicals: route towards high-spin-density surfaces — Yuri Tanuma[1,2], Gregor Kladnik[3,4], Luca Schio[4], •Marion van Midden Mavrič[1], Bastien Anézo[5], Erik Zupanič[1], Gregor Bavdek[4,6], Ruben Canton-Vitoria[7], Luca Floreano[4], Nikos Tagmatarchis[7], Hermann A. Wegner[8,9], Alberto Morgante[4,10], Chris Ewels[5], Dean Cvetko[1,3,4], and Denis Arčon[1,3] — [1]Jožef Stefan Institute, Ljubljana, Slovenia — [2]CAREM, Hokkaido University, Sapporo, Japan — [3]Faculty of Mathematics and Physics, University of Ljubljana, Slovenia — [4]CNR-IOM. Trieste, Italy — [5]IMN, Nantes, France — [6]Faculty of Education, University of Ljubljana, Slovenia — [7]Theoretical and Physical Chemistry Institute, Athens, Greece — [8]Institute of Organic Chemistry, Giessen, Germany — [9]Center for Materials research, Giessen, Germany — [10]Physics department, University of Trieste, Italy

Organic radicals offer a promising qubit platform. Despite them usually being unstable, as their unpaired electrons are highly reactive, they can be stabilized via weak interactions with the surface.

We study $C_{59}N^{\bullet}$ deposited on Au(111) in UHV at molecular coverage up to 2ML. Using NEXAFS and STM we show that single azafullerenes are indeed deposited on the surface. Together with DFT calculations, NEXAFS/XPS allows us to identify the spectroscopic fingerprint of the molecular radical state. The binding of $C_{59}N^{\bullet}$ changes as a function of coverage. We identify a novel intermediate high-spin-density state between 1ML and 2ML.

O 70.5 Wed 18:00 Poster C

Manipulating Quantum States: Indirect Readout of $^{163}\mathrm{DyPc}_2$ Single-Molecule Magnets via Spin Cascade on Au(111) using Scanning Tunneling Microscopy — •Simon Gerber[1], Hongyan Chen[1], Philip Schmid[1], Vera Schmeiser[1], Svetlana Klyatskaya[1], Eufemio Moreno-Pineda[2], Mario Ruben[1,3], and

Wulf Wulfhekel[1] — [1]Karlsruher Institute of Technology — [2]Universidad de Panamá — [3]Centre Européen de Sciences Quantiques, Straßbourg

We present an innovative method to indirectly read the nuclear spin of ^{163}DyPc$_2$ (I=5/2) single-molecule magnets on Au(111) using milli-Kelvin scanning tunneling microscopy. The Kondo effect, initiated by an unpaired electron in the π-radical on the exposed Pc ligand, induces a spin cascade[1,2], revealing discrete signal jumps on the order of minutes. These jumps, which may be correlated with nuclear spin flips, suggest possibilities for manipulation with an RF source. First results of RF-induced transitions are presented. This study focuses on the interplay between local 4f electrons, unpaired electrons and nuclear spins in rare earth-based systems. We demonstrate that reading the nuclear spin of ^{163}DyPc$_2$ using spin-polarised scanning tunneling microscopy (Sp-STM) does not need a magnetic field. This research makes a significant contribution to the understanding and manipulation of quantum states in single molecule magnets.

[1] R. Vincent et al,. Nature **488**, 357 (2012); S. Thiele et al., Science **344**, 1135 (2014) [2] T. Frauhammer, H. Chen et al., Phys. Rev. Lett. **127**, 123201 (2021)

O 70.6 Wed 18:00 Poster C

Mn on 2H-NbSe2 - From Single Magnetic Atoms to Spin Chains — •Margarete Huisinga[1], Lisa M. Rütten[1], Kai Rossnagel[2,3], and Katharina J. Franke[1] — [1]Fachbereich Physik, Freie Universität Berlin, Germany — [2]Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, Germany — [3]Ruprecht Haensel Laboratory, Deutsches Elektronen- Synchrotron DESY, Hamburg, Germany

On superconductors the interaction of unpaired electron spins of magnetic atoms with the Cooper pairs gives rise to so called Yu-Shiba-Rusinov (YSR) states. These states can be probed on the atomic scale using a scanning tunneling microscope (STM). YSR states are influenced by the environment of the atom on the surface such as its position on the lattice or a charge density wave (CDW).

Here, we investigate Mn atoms on 2H-NbSe2. Using the tip of a STM we build chains of Mn atoms. Interestingly, densely packed Mn chains exhibit inelastic excitations outside the superconducting gap along with YSR states. These inelastic excitations depend on the parity of the number of atoms in the chain, and vary in intensity and energy along the chain. We investigate the evolution of the inelastic excitations and YSR states with increasing chain length.

O 70.7 Wed 18:00 Poster C

Controlling hybridization in Yu-Shiba-Rusinov dimers through a charge-density wave — •Lisa M. Rütten[1], Eva Liebhaber[1], Kai Rossnagel[2,3], and Katharina J. Franke[1] — [1]Fachbereich Physik, Freie Universität Berlin, Germany — [2]Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, Germany — [3]Ruprecht Haensel Laboratory, Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany

Unpaired adatom spins on superconductors interact with the Cooper pairs of the substrate and induce Yu-Shiba-Rusinov (YSR) states inside the superconducting gap. These states can be probed by scanning tunneling spectroscopy at the single-atom scale. In quasi-two-dimensional materials, the YSR wave functions extend over several nanometers. If the YSR wave functions of two impurities overlap they can hybridize which is observed as a splitting of YSR resonances in tunneling spectra. Additionally, the YSR states are influenced by the presence of a charge-density wave (CDW). Fe atoms on $2H$-NbSe$_2$ have been shown to exhibit long-range YSR wave functions, where the energies, at which the YSR resonances are observed, depend strongly on the atoms position with respect to the CDW.

Here, we use lateral manipulation to arrange Fe atoms on $2H$-NbSe$_2$. While an isolated atom hardly influences the CDW, a set of atoms may impact its incommensurate pattern. We build a dimer of Fe atoms and then rearrange additional atoms around it to modify the CDW. We show that the hybridization strength can be switched as the underlying CDW is changed.

O 70.8 Wed 18:00 Poster C

Chiral hybridization of Yu-Shiba-Rusinov states in Fe clusters on Pb(111) — •Bharti Mahendru[1], Léa Meynier[1], Lisa M. Rütten[1], Harald Schmid[1], Felix von Oppen[1,2], and Katharina J. Franke[1] — [1]Fachbereich Physik, Freie Universität Berlin, Berlin, Germany — [2]Dahlem Center for Complex Quantum Systems and Fachbereich Physik, Freie Universität Berlin, Berlin, Germany

Unpaired electron spins exchange coupled to a superconductor give rise to bound states inside the superconducting energy gap, so-called Yu-Shiba-Rusinov (YSR) states. Scanning tunneling microscopy and spectroscopy (STM/STS) are powerful tools to probe the excited state around the Fermi level of such systems at the atomic scale. Previously, it has been shown that the crystal field splits the d-levels of Mn atoms on Pb surfaces, which lead to distinct YSR states inheriting the symmetry of the spin-carrying orbital. Here, we investigate self-assembled clusters of Fe on Pb(111) using STM/STS. Single Fe atoms, although having four unpaired electrons in the d-levels in gas phase, do not show YSR states on Pb(111) surface. In contrast, we observe different clusters, all showing in-gap states. These YSR states exhibit different symmetries that are related to the structure of the cluster on the Pb(111) surface. Interestingly, one type of the cluster exhibits spatially chiral YSR wavefunctions in differential conductance (dI/dV) maps. Using a simple model, we find that the origin of this chirality results from the adsorption of Fe atoms in the three-fold symmetric hollow sites of the substrate and the extent of the YSR wavefunctions.

O 70.9 Wed 18:00 Poster C

Self-Assembly of Magnetic Atom-Molecule Complexes on a Surface — Wantong Huang[1], Paul Greule[1], Máté Stark[1], Joris Van Slageren[2], Benedetta Casu[3], Christoph Sürgers[1], Wolfgang Wernsdorfer[1], Giorgio Sangiovanni[4], Christoph Wolf[5], and •Philip Willke[1] — [1]Physikalisches Institut, Karlsruhe Institute of Technology, Karlsruhe, Germany — [2]Institute of Physical Chemistry and Center for Integrated Quantum Science and Technology IQST, University of Stuttgart, Pfaffenwaldring 55, 70569 Stuttgart, Germany — [3]Institute of Physical and Theoretical Chemistry, University of Tübingen, Auf der Morgenstelle 18, 72076 Tübingen, Germany. — [4]Institut für Theoretische Physik und Astrophysik, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — [5]Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul, Republic of Korea.

Both single atomic as well as molecular spin centers are prime candidates for atomic-scale quantum information processing and often individually addressed and manipulated using STM. Here, we demonstrate a reliable self-assembly of magnetic complexes consisting of individual iron (Fe) atoms and molecules on a silver substrate. We employ two types of molecules, bis(dibenzoylmethane) copper(II) [Cu(dbm)2] and iron phthalocyanine (FePc), and show that Fe atoms always preferentially attach underneath the benzene ring ligand of the molecules. In both cases, a molecule can host up to two Fe atoms. In addition, we observe a change in the magnetic properties of the attached Fe atoms in spectroscopy, revealing a distinct Kondo signature at the Fe site.

O 70.10 Wed 18:00 Poster C

Exploring the magnetic field dependence of emulated molecular orbitals on Cs/InSb(110) — •Hermann Osterhage, Niels P. E. van Mullekom, Emil Sierda, Kira Junghans, Julian H. Strik, Daniel Wegner, and Alexander A. Khajetoorians — Institute for Molecules and Materials, Radboud University, Nijmegen, The Netherlands

Quantum simulators can be used to emulate custom-made Hamiltonians that are mathematically hard to solve in a physical system. In a recent realization of a quantum simulator, molecular orbitals were emulated by positioning individual Cs atoms on an InSb(110) surface [1]. It was previously shown that this material system also exhibits Landau quantization in the presence of an out-of-plane magnetic field [2]. On this platform, artificial atoms can be realized as localized electronic states created from ring structures of Cs atoms, assembled using scanning tunneling microscopy. Coupling these artificial atoms in different geometries results in artificial molecular structures that emulate planar organic molecules with different symmetries [1]. The resulting structures show electronic states of multi-orbital, as well as hybrid orbital character. Here, we explore the role of magnetic field on the emulated molecular states.

[1] E. Sierda *et al.*, Science 380, 1048 (2023).

[2] K. Hashimoto *et al.*, Phys. Rev. Lett. 101, 256802 (2008).

O 70.11 Wed 18:00 Poster C

Simulation of the nuclear spin state switching driven by ESR in STM — •Hester Vennema[1], Rik Broekhoven[1], Evert Stolte[1], Esther Teng[1], Jinwon Lee[1], Philip Willke[2], and Sander Otte[1] — [1]Delft University of Technology, Delft, The Netherlands — [2]Karlsruhe Institute of Technology, Karlsruhe, Germany

The spin of a single nucleus is a prime candidate for quantum information applications due to its weak coupling to the environment and subsequently longer coherence times. Using the high energy resolution of in combination with the high spatial resolution of STM, it is possible to measure the hyperfine interaction between the nucleus and an electron for single atoms on a surface. A recent effort has been proven successful to resolve the coherent dynamics of a hyperfine driven interaction between nucleus and electron spin.

In our latest experiment, we have observed nuclear spin switching in real time of a 49Ti isotope (I = 7/2) on MgO/Ag(100). The electron spin is continuously driven by a constant RF voltage and interacts with the nucleus by hyperfine interaction. In this work, we demonstrate a model to gain insight in the seen dynamics. Particularly, we are interested in the effect on the nuclear spin dwell time when driving the electron spin slightly off resonance. Combining open system dynamics for two coupled spins, and an intuitive simulation of the experiment, we gain insight in the interaction of the electron and the nucleus. This could lead to understanding dominant relaxation processes and improving conditions to increase the lifetime of the nuclear spin.

O 70.12 Wed 18:00 Poster C

Magnetic adatoms on ultrathin films of the hexagonal insulator ZnO — •Henrik Lichtl[1], Lukas Arnhold[1], Leon Rullkoetter[1], Nicolaj Betz[1], Christoph Wolf[2], Susanne Baumann[1], and Sebastian Loth[1] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]Center for Quantum Nanoscience, Institute for Basic Science, Seoul, South Korea

Different thin insulating layers can be used to tailor the electronic properties of adsorbed species. We use a double layer of zinc oxide (ZnO), a hexagonal wide bandgap semiconductor[1], to mitigate electron scattering between the Ag (111) substrate and Co atoms deposited on the ZnO surface. Ultrathin zinc oxide (ZnO) forms a honeycomb lattice and a Moiré superlattice on Ag(111). Scanning tunneling microscopy (STM) and density-functional theory (DFT) show that the Co atoms adsorb in the hollow sites of the ZnO hexagons. The atoms can be manipulated laterally with the STM tip. We find different Co entities that range from individual atoms to small nanostructures that feature distinct shape- and magnetic field-dependent spin excitations. This study lays the foundation for further exploration and manipulation of spins on ZnO towards design of frustrated atomic-scale magnetic systems.

[1] A. Shiotari, et al., J. Phys. Chem. C 2014, 118, 47, 27428-27435

O 70.13 Wed 18:00 Poster C

Coherent control of artificial atom-molecule hybrids — •Máté Stark[1], Wantong Huang[1], Paul Greule[1], Daria Sostina[2], Constantin Weeber[1], Jose Gálvez[3], Christoph Sürgers[1], Christoph Wolf[3], Wolfgang Wernsdorfer[1], and Philip Willke[1] — [1]Physikalisches Institut (PHI), Karlsruhe Institute of Technology, Karlsruhe, Germany — [2]Institute of Quantum Materials and Technologies (IQMT), Karlsruhe Institute of Technology, Karlsruhe, Germany — [3]Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul, Republic of Korea

By harnessing the atomic precision of Scanning Tunneling Microscopy (STM) in tandem with Electron Spin Resonance (ESR), we show in this work that artificially built atom-molecule hybrids extend the scope of future building blocks towards more versatile qubit structures. Specifically, our focus is on a magnetic system comprising an iron phthalocyanine (FePc) molecule and an Fe atom, resulting in a mixed spin(1/2,1) antiferromagnetically coupled Heisenberg system. This chemical complex shows properties attributed to a spin-1/2 system, which we characterize by driving it into coherently controlled ESR. By using both pump-probe measurements and coherent control techniques, we find a remarkable enhancement in the relaxation time of the hybrid, reaching approximately 1us. This significantly surpasses the values of pristine FePc [1,2] and we attribute this enhancement to the correlation between the two spins in the dimer as well as to enhanced potential scattering. [1] Zhang, X., et al., Nat. Chem. 14 ,59-65 (2022); [2] Willke, P., et al., ACS nano 15, 17859-17965 (2021)

O 70.14 Wed 18:00 Poster C

Spin and charge control of topological end states in chiral graphene nanoribbons — •Leonard Edens[1], Francisco Romero[1], Manuel Vilas-Varela[2], Giuliana Beretta[3], Sofia Sanz[4], Thomas Frederikssen[4], Diego Peña[2], and Nacho Pascual[1] — [1]CIC nanoGUNE, Spain — [2]CiQUS and Universidade de Santiago de Compostela, Spain — [3]Politecnico di Torino, Italy — [4]DIPC, Spain

Chiral graphene nanoribbons can exhibit radical end states due to nontrivial electronic topology. A localized spin fingerprint however remains unobserved as electron transfer to or from the underlying catalyst metal due to work function mismatch causes deviation from half filling, quenching spin. Gold alloyed with rare-earth metals still catalyses on-surface synthesis while exhibiting a lower work function. We show here that (3,2,8)-chiral nanoribbons can be synthesized with atomic precision on the ferromagnetic $GdAu_2$ intermetallic surface, and do not undergo significant charge transfer. By tunneling spectroscopy, we furthermore observe that the occupancy of the two topological end states can be reversibly switched from singly to doubly occupied by lateral manipulation of the nanoribbon across the substrate's moiré lattice. In the doublet case, low-energy features are interpreted as Kondo-screened spin-flip excitations of the π-radical state acted upon by the Gd 4f-mediated exchange bias.

O 70.15 Wed 18:00 Poster C

Paramagnetic resonance of individual organic molecules driven by quantum spin torque — Stepan Kovarik[1], Richard Schlitz[1,2], Aishwarya Vishwakarma[1], •Dominic Ruckert[1], Pietro Gambardella[1], and Sebastian Stepanow[1] — [1]ETH Zürich, Zurich, Switzerland — [2]Department of Physics, University of Konstanz, Germany

It is well-established that time-dependent electromagnetic fields can excite the spin states of a quantum system. In electron paramagnetic resonance (EPR), an oscillating magnetic field B_1 with RF frequency allows coherent control over the spin state of single atoms or molecules.

However, our research shows an alternative mechanism for initiating spin resonance on a single pentacene molecule locally probed in STM.

Here, the mechanism for driving spin resonance is mediated by spin-polarized transport through a locally probed single molecule on the surface. A spin-polarized current oscillating with RF frequency can drive the precession of the localized magnetic moment as theoretically predicted by Shakirov et al. [1]. This process provides a distinct and complementary perspective to the B_1-driven mechanism [2].

Our findings allow the current induced spin initialization of a single spin and thereby hold promise for advancements in molecular-scale electronics and spintronics

O 70.16 Wed 18:00 Poster C

Aging in the self-induced spin glass Nd(0001) — Lorena Niggli[1], •Julian H. Strik[1], Anders Bergman[2], Mikhail I. Katsnelson[1], Daniel Wegner[1], and Alexander A. Khajetoorians[1] — [1]Institute for Molecules and Materials, Radboud University, Nijmegen, The Netherlands — [2]Department of Physics and Astronomy, Uppsala University, Uppsala, Sweden

Elemental neodymium has been shown to be a self-induced spin glass, where glassy behaviour stems solely from the frustrated nature of the magnetic interactions [1]. This contrasts with traditional spin glasses, where the presence of disorder is essential toward realizing glassy behaviour. The magnetic state of Nd(0001) is characterized by a lack of long range order, but exhibits local non-collinear order (Q-states). Upon increasing the temperature, neodymium displays an unusual magnetic phase transition from a self-induced spin glass to a long-range ordered multi-Q phase [2]. Here, we explore the aging behaviour of Nd(0001) in its self-induced spin glass state using spin-polarized scanning tunneling microscopy in varying magnetic fields and variable temperature. We explore how the favourability of the Q-states evolves as we age the system and relate these changes to the preferred structure of the ordered phase. These observations indicate that neodymium may be a multi-well system, which deviates from the traditional energy landscape expected of spin glass systems, thus providing a new platform to study aging dynamics as well as dynamic heterogeneity.

[1] U. Kamber et al., Science 368 (2020).

[2] B. Verlhac et al., Nat. Phys. 18 (2022).

O 71: Poster: Plasmonics and Nanooptics

Time: Wednesday 18:00–20:00 Location: Poster D

O 71.1 Wed 18:00 Poster D

Light-driven Microdrones — •Carsten Büchner, Jin Qin, and Bert Hecht — Nano-Optics & Biophotonics Group, Experimental Physics 5, University of Würzburg, Germany

The precise manoeuvring of microscopically small objects is a major component of today's microrobotics research and promises a wide range of applications in areas such as micromanufacturing or medicine. Most microrobots use magnetic, acoustic or chemical methods to generate the required propulsion. Here, we demonstrate the fabrication and steering of microdrones that are controlled solely by optical forces in unfocussed light fields. The architecture of the microdrones with a lateral size of approximately 2 um and a mass of 2 pg is comparable to recreational drones and can be realised with two or four motors. The nanomotors, which are individually addressable chiral plasmonic nanoantennas, resonantly scatter the circular polarization components of the driving light into well-defined directions, thus achieving thrust exclusively by photon recoil forces. Two overlapping unfocused light fields of 830 and 980 nm wavelength are used to address the motors. This allows the microdrone to operate in 2D in all three degrees of freedom in a fluid environment. With the help of electro-optic modulators, the properties of the light fields can be changed both quickly and precisely, thus varying the movement direction of the drone. By implementing optical tweezers in the drone body, the drones are capable of transporting and releasing nano-sized cargos like nanodiamonds. Biological applications such as the manipulation of red blood cells, yeast cells or bacterias are also conceivable and are being investigated.

O 71.2 Wed 18:00 Poster D

Plasmonic lightning-rod effect — •Vlastimil Křápek, Rostislav Řepa, Michael Foltýn, Tomáš Šikola, and Michal Horák — Brno University of Technology, Czechia

Field enhancement is one of the key targets in plasmonics [1,2,3]. Intuitively, it is expected that plasmonic antennas (PAs) produce a strong electric field near their sharp features. However, there are other factors besides local curvature contributing to the field enhancement, including the evanescent nature of surface plasmon polaritons or the magnitude of the induced charge for a specific excitation.

In our communication, we isolate the contribution of the local curvature of a PA to the magnitude of the electric field of its dipole plasmon resonance. We design a set of rod-like PAs of identical length with two cylindrical terminations, one with a fixed radius and one with a variable radius. The PAs are investigated by electron energy loss spectroscopy (EELS) and electromagnetic simulations. At the fixed-radius end, we observe the identical loss intensity and field enhancement for all the PAs. This ensures that the variations in the response at the variable-radius end can be attributed purely to the local curvature. Finally, we present an intuitive phenomenological model for the induced electric field based on an effective induced point charge.

[1] M. Hrtoň *et al.*, Phys. Rev. Applied **13**, 054045 (2020).

[2] V. Křápek *et al.*, Nanophotonics **9**, 623 (2020).

[3] O. Bitton *et al.*, Nat. Commun. **11**, 487 (2020).

O 71.3 Wed 18:00 Poster D

Control and measurement of optical fields in a random photonic media — •Diptabrata Paul and Frank Cichos — Peter Debye Institute for Soft Matter Physics, Universität Leipzig, 04103 Leipzig, Germany

The ubiquitous presence of disordered photonic structures in nature has inspired investigation of how wave propagation in complex media could be exploited to control the scattering, transport, and localization of light in matter. However, volumetric scattering in the complex media leads to a universal interference pattern called speckles, which poses a significant challenge in controlling and probing phase and amplitude of light after passing through the medium. To address this, herein, we propose a method by which modulation of incident optical fields using a digital micromirror device (DMD) could achieve modification of optical fields in the random photonic media. Real time imaging of the speckles along with an iterative feedback algorithm sequentially switches the DMD mirror states to on or off positions to achieve incident field binary amplitude modulation and hence desired modification of fields in the medium. On the other hand, we employ a diffuser that could be used for sensing the thermo-optical modulation of an optical field. Specifically, we infer the local perturbation of the wave front by comparing the speckle pattern between a reference and a thermo-optically modified field, which in turn can also quantify the temperature profile. The advent of spatial control over the phase and amplitude of light waves can lead to major advances in fields from imaging and information technology to biomedical optics.

O 71.4 Wed 18:00 Poster D

Programming large-area geometrical phase metasurfaces with the plasmonic phase-change material In_3SbTe_2 — •Florian Bontke[1], Lukas Conrads[1], Andreas Ulm[2], Matthias Wuttig[1], Robert Schmitt[2], and Thomas Taubner[1] — [1]I. Institute of Physics (IA), RWTH Aachen — [2]Fraunhofer IPT

Phase-change materials (PCMs) have been established as a versatile basis for non-volatile switchable metasurfaces based on a change in the refractive index [1]. Here we employ the novel switchable infrared plasmonic PCM In_3SbTe_2 (IST) whose optical properties can be switched between dielectric (amorphous) and metallic (crystalline) in the full infrared range. While IST has been employed for direct writing of metallic nanoantennas [2] and emissivity shaping metasurfaces [3], its potential for large-area geometric phase metasurfaces remains unexplored. Using circularly polarized light, the geometric Pancharatnam-Berry phase of rotated antennas can be used to obtain full 2π phase control. In this work, a fast commercial laser writing setup is utilized to realize large-area beam steering and other functional metasurfaces for the mid-infrared. Due to the fast writing speed, producing millimeter-scale metasurfaces with over 10^6 individual antennas is feasible. The possibility of creating large-area metasurfaces via direct laser writing without cumbersome fabrication techniques paves the way towards mass production and rapid prototyping of reconfigurable meta-optics in the infrared.

[1] Wuttig et al. *Nat. Photon.* **11**, 465 (2017) [2] Hessler et al. *Nat. Com.* **12**, 924 (2021) [3] Conrads et al. *Adv. Opt. Mat.* **11**, 8 (2023)

O 71.5 Wed 18:00 Poster D

Bloch surface waves on multilayer structures made from tantalum pentoxide and silicon dioxide — •Amélie Wagner and Stefan Linden — Physikalisches Institut, Universität Bonn, Nussallee 12, 53115 Bonn, Germany

Bloch surface waves (BSWs) are electromagnetic surface waves that are confined to the interface between a periodic dielectric multilayer structure and a homogeneous dielectric medium. Guiding of BSWs with low propagation losses can be achieved by depositing thin dielectric ridges on top of the multilayer film. BSW waveguides are therefore an attractive alternative to dielectrically loaded surface plasmon polariton waveguides, whose propagation length is limited by Ohmic damping. Here we report on the design and fabrication of BSW waveguides. The multilayer structure consists of alternating tantalum pentoxide (Ta2O5) and silicon dioxide (SiO2) layers deposited by electron beam evaporation. The required thicknesses of the layers are determined beforehand with the help of transfer matrix calculations. In order to test the optical quality of the fabricated samples, we perform total internal reflection measurements in Kretschmann configuration. In the next step, we will deposit dielectric ridges on top of the multilayers by electron beam lithography and evaluate the propagation length by leakage radiation microscopy.

O 71.6 Wed 18:00 Poster D

Scalable 3D Printing of Micro-Optical Elements for Optical Fibres — •Mika Mc Keever and Stefan Linden — Physikalisches Institut, Universität Bonn, Nussallee 12, 53115 Bonn, Germany

3D printing via direct laser writing, utilising two-photon polymerisation has revolutionised the fabrication of micro-optics for optical fibre applications. Previous advancements include the successful printing of optical devices, such as collimating lenses, directly onto the ends of optical fibre. However, the existing technique presents challenges, particularly the manual printing process for each individual fibre, resulting in time-intensive production and difficulties in aligning optical elements with the fibre core. This work addresses these issues by introducing a technique for printing hanging optical elements on a glass substrate, which can be attached to the fibre end. This is achieved by inserting the fibre into a mask that is printed on top. This method enables the simultaneous printing of multiple optical elements, accelerating production, while maintaining the same printing parameters as direct fibre printing. Moreover, the use of the mask facilitates alignment of the optical elements with the fibre core. By adopting this approach, optical elements with dimensions of up to 300 micrometres can be printed.

O 71.7 Wed 18:00 Poster D

Non-equilibrium electrons generated from the extended two-temperature model to drive chemical dynamics at surfaces — •Henry T. Snowden and Reinhard J. Maurer — University of Warwick, Coventry, UK

A mechanistic understanding of ultrafast light-matter interactions with surfaces and nanoparticles is invaluable for the study of ultrafast dynamics at surfaces. The two-temperature model (2TM) represents the most common approach to model light-matter interaction. It assumes that electrons remain in a Fermi-Dirac distribution, which is heated by a laser source term. This assumption is invalid immediately after a laser pulse, where a short-lived population of non-equilibrium electrons is generated. Here we will explore the extended two-temperature model (e2TM), proposed by Carpene [Phys Rev B 74, 24301 (2006)], including further extensions from Uehlein et al. [Nanomaterials 12, 1655 (2022)]. We will show that the e2TM captures information consistent with simulations based on the Boltzmann transport equation while maintaining much of the simplicity and computational efficiency of the 2TM. We incorporate many parameters directly calculated from first principles, such as the density-of-states (DOS) and properties derived thereof. We present a systematic assessment of the dependence of the e2TM on the type of metal and surface termination and directly relate our results to time-resolved spectroscopy results. Finally, we will examine the necessary properties of a material required to generate long-lived non-equilibrium electrons and suggest possible material candidates.

O 71.8 Wed 18:00 Poster D

Energy and momentum distribution of surface plasmon-induced hot carriers — •Christopher Weiss[1], Tobias Eul[2], Eva Prinz[1], Benjamin Stadtmüller[1], and Martin Aeschlimann[1] — [1]Department of Physics and Research Center OPTIMAS, University of Kaiserslautern-Landau, Germany — [2]Institute of Experimental and Applied Physics, University of Kiel, Germany

Are the spectroscopic properties of plasmon- and photon-induced carriers fundamentally different? This question is crucial for the advancement of plasmonic energy conversion. Electrons excited at bulk plasmon resonance show a distinct distribution in energy and momentum space [1], while the electron distribution of surface plasmon-induced carriers is still under debate [2].

To address this issue for surface plasmons, we aim to separate the energy and momentum distributions of surface plasmon polariton (SPP)-induced hot electrons from those of photoexcited ones. Exciting a circular ring-slit geometry with radially and azimuthally polarised light, we can clearly separate the plasmon and photon-induced hot carriers in space and time. Our time-resolved photoemission electron microscope (PEEM) is then able to capture their signatures in real time. In this contribution, we show that there are spectroscopic differences between photoemitted electrons and those generated by plasmoemission in the centre of the structure.

[1] Reutzel et al., Phys. Rev. Lett. 123 (2019), 017404

[2] Hartelt et al., ACS Nano 15, 12 (2021), 19559–19569

O 71.9 Wed 18:00 Poster D

Ionization-assisted coherent optical two-dimensional nanoscopy — •Philipp Kessler, Luisa Brenneis, Victor Lisinetskii, Matthias Hensen, and Tobias Brixner — Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg

Optical two-dimensional (2D) spectroscopy has proven to be a powerful tool for the investigation of electronic couplings and ultrafast energy transport phenomena. The combination of this technique with photoemission electron microscopy (PEEM), i.e., coherent "2D nanoscopy", enables the investigation of surface system dynamics with high spatio-temporal resolution [1]. However, photoemission may require multiple interactions with individual excitation pulses, causing overlapping signal contributions. The use of an additional ionization pulse circumvents this problem by projecting the population of the system's states into the detection channel via photoemission, as shown in the gas phase [2]. Here, we present modifications of our existing PEEM setup [3] to realize this scheme. This includes second-harmonic generation on a separated beam path to produce the ionization pulse and optimization of the pulse-shaper generated four-pulse excitation sequence, which is accurately reconstructed by spectral interferometry. We further show a new beam stabilization system which enables stable incoupling into the freestanding PEEM by independently stabilizing both beam paths.

[1] M. Aeschlimann et al., Science 333, 1723 (2011).

[2] Uhl et al., Optica 8, 1316 (2021).

[3] Huber et al., Rev. Sci. Instrum. 90, 113103 (2019).

O 71.10 Wed 18:00 Poster D

s-SNOM calibration using multiple tapping harmonics for complex permittivity reconstruction — •Dario Siebenkotten[1], Bernd Kästner[1], Arne Hoehl[1], Manuel Marschall[1], and Shuhei Amakawa[2] — [1]Physikalisch-Technische Bundesanstalt, Abbestr. 2-12 10587 Berlin — [2]Graduate School of Advanced Science and Engineering, Hiroshima University, Higashihiroshima, Japan

Infrared scattering-type scanning near-field optical microscopy (s-SNOM) has found wide-spread success in the nanoscale investigation of material properties such as the free-carrier density and crystal structure. The deep sub-wavelength resolution of s-SNOM is achieved by the interaction between the sharp apex of a metallized AFM tip under illumination and the sample surface below it. This complex system poses a difficult inverse problem for the recovery of sample properties from measurements. Recently, a calibration method has been proposed that describes this interaction by means of calibration parameters, instead of fitting the physical characteristics of a heavily idealized tip model [1]. However, the proposed calibration model was designed for a stationary tip, but the use of periodic tip height modulation is essential for s-SNOM measurements. Here, we propose an extension to the model that includes the tip modulation by making use of multiple tapping harmonics. We validate our proposed extension on doped silicon microstructures by reconstructing the permittivity over a broad spectral range and extracting the free-electron density and damping.

[1] Guo et al. Appl. Phys. Lett. 118, 041103 (2021)

O 71.11 Wed 18:00 Poster D

Nano-imaging reveals the stacking dependent dispersion of hybrid polaritons in a Trilayer Graphene and Hexagonal Boron Nitride Heterostructure — •Christian Dittmar, Lina Jäckering, Konstantin Wirth, and Thomas Taubner — I. Institute of Physics, RWTH Aachen University

Two-dimensional (2D) van der Waals (vdW) materials can be combined to heterostructures [1]. Hexagonal Boron Nitride (hBN) and Trilayer Graphene (TLG) are two appealing 2D vdW materials. hBN is a natural hyperbolic material and enables low-loss and volume-confined hyperbolic phonon polaritons (HPhPs) [2]. TLG is gate-tuneable and hosts surface plasmon polaritons. TLG occurs in either a Bernal (ABA) or rhombohedral (ABC) stacking order [3]. Uniting isotopically pure hBN and TLG in a heterostructure enables the formation of hybridized hyperbolic plasmon-phonon polaritons (HP^3). These (HP^3) can be launched and imaged by a scattering-type scanning near-field optical microscope. HP^3 combine the tunability of graphene and the low losses of the HPhPs in hBN. For the first time, we image stacking dependent propagating polaritons in a hBN-TLGN heterostructure. The stacking dependency manifests in the different propagation wavelength originating from different dispersions. The results lay the foundation for distinguishing different stacking orders of TLG encapsulated in hBN and for identifying domain boundaries in TLG. We also emphasize the possibility to image buried structures below an hBN layer. [1] Basov et al. *Science* **354**, 6309 (2016), [2] Caldwell et al. *Nature Reviews Materials* **4**, 552 (2019), [3] Chen et al. *Nature* **487**, 77 (2012)

O 71.12 Wed 18:00 Poster D

Orientation-dependent interpretation of Janus particle scattering spectra — •Felix Hermann Patzschke, Arthur Markus Anton, and Frank Cichos — Leipzig University, Peter Debye Institute for Soft Matter Physics, Linnéstr. 5, 04103 Leipzig

Plasmonic Janus particles consist of dielectric core particles with a thin metallic cap on one side and are widely used in active matter research. The plasmonic cap enhances optical scattering and absorption, allowing for self-propulsion through temperature gradients as well as efficient trapping and tracking. The asymmetry of such a particle gives rise to surface plasmon modes whose excitation is sensitive to the angle at which the particle is illuminated. Even though the angle of illumination strongly influences the particle's scattering response, the optical properties of such metallic caps have hardly been investigated.

We probe the light scattering of individual micrometre-sized, spherical, Au-coated Janus particles by means of Selective Illumination Multiplexed Fourier Plane Spectroscopy. This novel method allows us to explore microparticles' scattering characteristics resolved for wavelength, angle of illumination and scattering angle. In addition, we supplement our experimental results with finite-element simulations and correlate spectral markers to orientation-dependent surface plasmon modes.

This additional information on the correlation of angular and spectral information could pave the way for new methods of orientation detection. They also shed new light on the interaction of such spherically capped particles with light inducing forces and torques.

O 71.13 Wed 18:00 Poster D

Nanoscale Plasmonic Su-Schrieffer-Heeger Chains — •Benedikt Schurr[1,3], Luisa Brenneis[2], Felix G. Kaps[3,5], Matthias Hensen[2], Philipp Grimm[1,3], Philipp Kessler[2], Tobias Helbig[4], Tobias Hofmann[4], Thorsten Feichtner[1,3], Monika Emmerling[1], Susanne C. Kehr[3,5], Tobias Brixner[2], Ronny Thomale[3,4], Lukas M. Eng[3,5], and Bert Hecht[1,3] — [1]NanoOptics & Biophotonics Group, Experimental Physics 5, University of Wuerzburg — [2]Institut fuer Physikalische und Theoretische Chemie, University of Wuerzburg — [3]Wuerzburg-Dresden Cluster of Excellence ct.qmat — [4]Institute for Theoretical Physics and Astrophysics, 1-4: University of Wuerzburg, Am Hubland, 97074 Wuerzburg, Germany — [5]Institut fuer Angewandte Physik, Technische Universitaet Dresden, 01062 Dresden, Germany

The Su-Schrieffer-Heeger (SSH) model describes one-dimensional (1D) periodic chains of coupled resonators with alternating coupling strengths. It features topologically protected and localized edge states. We fabricate and characterize plasmonic particle SSH-chains with alternating gaps in the nanometer range by focused He-ion beam milling starting from single-crystalline gold micro-platelets. Finite-difference time-domain (FDTD) simulations show the occurrence of edge modes for such geometries whose eigenfrequencies can be controlled by the particle length and the gap size. Using scattering scanning near-field optical microscopy (sSNOM) as well as photoemission electron microscopy (PEEM) of different chain configurations we obtain highly resolved near-field maps indicating edge-state formation.

O 71.14 Wed 18:00 Poster D

Contrast Mechanism Investigation of Single Core-shell Nanoparticles in s-SNOM Using Finite Element Simulations — •Dinghe Dai, Dario Siebenkotten, Richard Ciesielski, and Bernd Kästner — Physikalisch-Technische Bundesanstalt, Abbestr. 2-12, 10587 Berlin

Scattering-type scanning near-field optical microscopy (s-SNOM) is a widely used technique for nanoscale characterization of surface structures, surface polaritons, and biomolecular systems. Recently, core-shell nanoparticles have gained increasing interest in the fields of medical and material sciences. To investigate the properties of single core-shell nanoparticles using s-SNOM, we simulate the near-field contrast characterized by different geometrical and optical properties with the finite element method using the commercial software JCMSuite. We observe different resonances, which shift according to the geometrical properties. Field distribution plots indicate that these resonances may orignate from the interaction between the nanoparticle and the tip as well as between the nanoparticle and the substrate. The simulation results demonstrate the fundamental ability to determine geometrical and optical properties of individual nanoparticles using s-SNOM.

O 71.15 Wed 18:00 Poster D

Improved electron spectrometer for PINEM experiments in an SEM — •Isabella Probst[1], Roy Shiloh[2], Johannes Illmer[1], Franz Schmidt-Kaler[1], and Peter Hommelhoff[1] — [1]Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU), Erlangen, Deutschland — [2]Hebrew University, Jerusalem, Israel

Photon-induced near-field microscopy (PINEM) was demonstrated as a method for imaging electromagnetic fields with femtosecond time resolution. It has developed into intriguing electron light coupling experiments. When electrons interact with an optical nearfield, they can absorb or emit an integer number of photons, modulating the electron spectrum. Most PINEM experiments have been performed in transmission electron microscopes (TEMs). Scanning electron microscopes (SEMs) provide larger sample chambers, are less cost intensive and offer a different range of electron energies. In our SEM-PINEM setup, the electron energy is measured with a custom-built spectrometer [1]. To improve the spectral resolution of our spectrometer, we designed a magnetic lens-based telescope. The performance of the lenses for different positions, coil currents and electron energies was simulated. This improved system will allow investigating a plethora of materials and effects, previously unreachable in an SEM, with improved clarity.

[1] Shiloh, Chlouba, Hommelhoff, PRL 128, 235301 (2022)

O 71.16 Wed 18:00 Poster D

Scattering of controlled plasmonic particles configurations on the mirror — •Aleksei Overchenko and Frank Cichos — Leipzig University, Peter Debye Institute for Soft Matter Physics, Linnéstr. 5, 04103 Leipzig

Gold particles have found wide application in sensorics due to their field enhancement propensity that is used for example in Surface-Enhanced Raman Spectroscopy (SERS), Photothermal Therapy and Plasmonic Photovoltaics.

Field enhancement structures can be realised by brining two spheres into proximity, putting a sphere on a mirror or fabricating a bowtie antenna. Surface functionalization often requires complicated modification methods such as lithography and chemical binding, e.g., of DNA origami.

Here, we deliberately manipulate induvial as well as multiple gold particles in front of a gold mirror by local light induced temperature fields generating thermo-osmotic flow fields. We characterize the resulting single, dimeric and higher order clusters, their plasmonic coupling and the coupling of the particles to a thin gold film by scattering spectroscopy. The system is highly reconfigurable as no binding is required and analytes may be bound to the gold film to enable spatially resolved SERS or other field enhancement phenomena.

O 71.17 Wed 18:00 Poster D

Investigation of lithiated carbon as active plasmonic material system — •Valentin Maile, Mario Hentschel, and Harald Giessen — 4th Physics Institute, University of Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany

Active plasmonic structures are a key component in the recent advances in optical technologies due to their ability to confine and manipulate light on the nano-scale and thus aid the miniaturization of optical devices. A key component for future devices is switchabilty and tunability of the optical resonances. Recently, the electrically switchable metal-to-insulator transition of the organic conjugated polymer PEDOT: PSS was used to realize switchable plasmonic devices at up to 30Hz modulation speed. Here, we introduce a different concept based on lithium-intercalated carbon, a material system known from battery research. The reversible lithium intercalation in the carbon lattice causes an increase of charge carrier density and thus a change in the optical properties of the material, its color changing from black to golden, rendering it an interesting material system for switchable plasmonic nanostructures. In this work, lithiated forms of carbon and their change in optical properties are explored as a possible switchable material system for plasmonic nanostructures. This ansatz expands the toolkit of active plasmonic structures for metasufaces and nanooptics.

O 71.18 Wed 18:00 Poster D

Decoding the Emission from Gold Nanoparticles — •Jan Kutschera, Wouter Koopman, Felix Stete, and Matias Bargheer — University of Potsdam

Surface-enhanced Raman spectroscopy (SERS) has been a valuable tool in biochemical sensing, single-molecule detection, and photocatalysis for several decades. The molecular fingerprints in SERS spectra are always accompanied by a broad background originating from the plasmonic nanoparticles themselves.

In particular, highly clustered nanoparticles, as used in many applications, can produce strong backgrounds. Due to its dependence on the electron distribution of the plasmonic material, understanding this nanoparticle response is crucial for comprehending the mechanisms behind plasmon-enhanced phenomena, such as photochemistry, photovoltaics, and magnetism. While the discussion regarding the underlying processes of this nanoparticle response is ongoing, it is most often attributed to photoluminescence (PL) or electronic Raman scattering (ERS) and thus to intraband processes.

Especially for gold nanoparticles, the contribution of interband transitions must be considered. Utilizing photoluminescence excitation (PLE) spectroscopy, we show the strong dependence on interband excitation, while the emission spectrum continues to be determined by plasmon-enhanced intraband processes. Furthermore, the presented results support the theory of interband-enabled, Auger-like excitations in the conduction band of gold, followed by intraband emissions in the visible spectrum.

O 71.19 Wed 18:00 Poster D

Automation workflow for ML training on infrared spectra prediction — •Giulio Benedini[1,2], Matti Hellstrom[1], and Luuk Visscher[2] — [1]Software for Chemistry and Materials B.V., De Boelelaan 1083, 1081HV Amsterdam, The Netherlands — [2]Department of Theoretical Chemistry, Vrije Universiteit Amsterdam, De Boelelaan 1083, 1081 HV Amsterdam, The Netherlands

Infrared spectroscopy provides information on atomic structure while being cheap and non-invasive technique. Its wide application is hindered by its spectra interpretation. Computational simulations could solve this problem but ab initio approaches have very high computational costs. The use of machine learning interatomic potentials (MLIP) can solve this problem.[1] This work is about workflows to train and assess MLIPs tuned for IR spectra predictions. The FieldSchNet[2] MLIP was used for fitting while workflows were made with PLAMS[3]. The workflow has been developed to favor a re-usable approach. Hyperparameters of FieldSchNet have been optimized with the help of an automated workflow. The training resulted in mean absolute errors for prediction of frequencies of $10cm^{-1}$ and for intensities 7 km/mol on a dataset composed of ethanol and acetaldehyde. Currently active learning scheme needs to be now extend further to include predictions of IR spectra. [1] Chem. Rev. 2021, 121, 16, 10142*10186 [2] Chem. Sci., 2021, 12, 11473 [3] AMS 2023.1, SCM, Theoretical Chemistry, Vrije Universiteit, Amsterdam, The Netherlands, http://www.scm.com.

O 71.20 Wed 18:00 Poster D

Plasmonically induced RAFT polymerization on metal nanoparticles — Pascal Riebler, •Sergio Kogikoski Junior, Matthias Hartlieb, and Ilko Bald — Institute of Chemistry, University of Potsdam, Germany

Reversible addition-fragmentation chain transfer (RAFT) polymerization is one of the most versatile methods to synthesize complex polymeric architectures. This work used plasmonic Ag and Au plasmonic nanoparticles as activation agents instead of standard chemical procedures to generate the radical that will initiate the polymerization. The reactions were done in a usual photochemical setup in solution. Different irradiation wavelengths were used to investigate the activation of the chain transfer agent and the plasmonic nanoparticles. Spectroscopic and microscopic methods extensively characterized the obtained products. SERS results showed that the chain transfer agent used to modify the nanoparticles was detached from the surface to accommodate the newly synthesized polymer. Such a process of detachment generated a surface where monomers could polymerize until the surface coverage was complete, and the charge transfer from the NP to the chain transfer agent was hindered because of the distance. Our systematic results will pave the way for using plasmonic nanoparticles as reliable sources of RAFT-synthesized polymers.

O 71.21 Wed 18:00 Poster D

Hot-electron mediated chirality transfer in single nanoparticles — Seunghoon Lee[1,2], •Chenghao Fan[1], Artur Movsesyan[3], Johannes Bürger[1], Fedja J. Wendisch[1], Leonardo de S. Menezes[1], Stefan A. Maier[1,4], Haoran Ren[4], Tim Liedl[1], Lucas V. Besteiro[5], Alexander O Govorov[3], and Emiliano Cortés[1] — [1]Faculty of Physics, LMU München, 80539 Munich, Germany — [2]Department of Chemistry, Dong-A University, Busan 49315, South Korea — [3]Department of Physics and Astronomy, Ohio University, Ohio 45701, United States — [4]School of Physics and Astronomy, Monash University, Victoria 3800, Australia — [5]CINBIO, University of Vigo, 36310 Vigo, Spain

Recently, chiral plasmonic nanostructures have drawn tremendous interest due to their unique chiroptical properties. Here, we thoroughly investigate the plasmon-assisted growth of chiral nanoparticles from achiral Au nanocubes (AuNCs) via circularly polarized light (CPL) without involvement of any chiral molecules. We identify the structural chirality of our synthesized chiral plasmonic nanostructures by using circular differential scattering (CDS) spectroscopy correlated with scanning electron microscopy imaging at both the single-particle and ensemble levels. Theoretical simulations, including hot-electron surface maps, reveal that the plasmon-induced chirality transfer is mediated by the asymmetric distribution of hot electrons on achiral AuNCs under CPL excitation. Results presented here reveal fundamental aspects of chiral light-matter interaction, influencing the future design and optimization of chiral sensors and chiral catalysis, among others.

O 71.22 Wed 18:00 Poster D

Polarization sensitive plasmonic induced chemical reaction observed by chiral SERS — •Shashank Gahlaut[1], Ki Tae Nam[2], Alexander Govorov[3], and Ilko Bald[1] — [1]Institute of Chemistry, University of Potsdam, Potsdam-14476 Germany — [2]Department of Materials Science and Engineering, Seoul National University, Seoul 08826, S. Korea — [3]Institute of Fundamental and Frontier Sciences, University of Electronic Science and Technology of China, Chengdu 610054, China Department of Physics and Astronomy, Ohio University, Athens, Ohio 45701, United States

Plasmonic induced chemical reactions on nanoparticles has attracted the attention of community of material science. Metallic nanoparticles possess the potential of confining the light into sub-wavelength region. The surface plasmon excitation enables the enhancement of local electric field in the close vicinity of particles. The decay of surface plasmons results into the formation of hot charge carriers and localized heat. Solar to chemical energy conversion has become the most attractive application in photocatalysis by plasmonic NPs. Here, we have observed polarization sensitive photochemistry on chemically synthesized chiral plasmonic nanocubes (gold helicoids, AuNC). Dehalogenation of 8-Bromoadenine (8-BrAde), a radiosensitizer, by metallic hot electrons has been targeted as a model chemical reaction. Left-handed AuNC show higher rate of reaction with LCP light as compared to opposite combination of L-AuNC and RCP light. Photochemical g-factor was found to be very large as compare to the optical g-factor. This g factor increases with the increase in laser power.

O 72: Poster: Scanning Probe Microscopy: Light Matter Interaction at Atomic Scales

Time: Wednesday 18:00–20:00 Location: Poster D

O 72.1 Wed 18:00 Poster D

Luminescence of molecules with STM – from electron to photon excitations — Benjamin Doppagne[1], Shuiyan Cao[1], Song Jiang[1], Katharina Kaiser[1], Luis Parra Lopez[1], Anna Rosławska[1], Kirill Vasilev[1], Tomas Neuman[1], Andrei G. Borisov[2], Javier Aizpurua[3], Michelangelo Roméo[1], Alex Boeglin[1], Frederic Cherioux[4], Eric Le Moal[2], Hervé Bulou[1], Fabrice Scheurer[1], and •Guillaume Schull[1] — [1]Université de Strasbourg, CNRS, IPCMS, UMR 7504, F-67000 Strasbourg, France. — [2]Institut des Sciences Moléculaires d'Orsay (ISMO), UMR 8214, CNRS, Université Paris-Saclay, 91405 Orsay Cedex, France. — [3]Center for Materials Physics (CSIC-UPV/EHU) and DIPC, Donostia-San Sebastián, Spain — [4]Université Bourgogne Franche-Comté, FEMTO-ST, UFC, CNRS, Besançon, France

STM-induced luminescence (STML) and tip-enhanced photoluminescence (TEPL) enable fluorescence imaging of molecules with sub-nanometer spatial resolution. While the former approach relies on tunneling electrons as an excitation source, the latter involves exciting the molecule with light. In this summary, I will highlight recent results from our team and compare the respective benefits of each method.

O 72.2 Wed 18:00 Poster D

Interactions between plasmons and excitons in single molecules — •Miguel Varea[1,2], David Mateos[1,2], Óscar Jover[1,2], Koen Lauwaet[1], Alberto Martín-Jimenez[1], and Roberto Otero[1,2] — [1]IMDEA Nanoscience, Madrid, Spain — [2]Dep. de Física de la Materia Condensada and Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid

The interaction of localized plasmons and excitons in quantum emitters results in plexcitons, hybrid light-matter states, offering unique prospects for controlling single photon states. Scanning Tunneling Luminescence (STM-L) proves crucial in characterizing exciton-plasmon interactions, providing control over cavity dimensions and knowledge of molecular geometry. However, the incomplete comprehension of electron-tunneling luminescence processes hinders its full exploitation. Specifically, the optical behavior of metal-interfacing molecules lacks explanation when no insulation separates the molecule from the substrate. This study addresses this gap by studying changes in plasmonic modes within the cavity induced by organic molecules adsorbed on the metal surface. Despite a substantial drop in luminescence intensity due to the molecules, our analysis primarily links this decline to differences in the electronic structures of the molecules and the metal surface. By isolating the strictly optical effects of molecule presence, luminescence spectra become comparable, though not entirely identical. This contribution underscores the molecule-specific and aggregation-dependent nature of these modifications, paving the way for using STM-L to explore plasmon-exciton coupling at the nanoscale.

O 72.3 Wed 18:00 Poster D

Luminescence on the atomic scale in a Terahertz Scanning Tunneling Microscope — •Johannes Schust, Kurt Lichtenberg, Felix Huber, Susanne Baumann, and Sebastian Loth — Institute for Functional Matter and Quantum Technologies, University of Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany

The interplay between light and matter has a key role in solid-state research, and it constitutes a fundamental element of numerous contemporary technologies. Therefore, exploring the photonic properties of novel quantum systems is of high significance, while pushing the frontiers of both temporal and spatial resolution remains a persistent challenge. Here, we implement electroluminescence sensing into an ultra-fast Terahertz scanning tunneling microscope (STM), i.e. extend the system's capability to couple light directly out of the junction. Our exceptional vacuum chamber design features a thin slot that hosts the whole measuring unit in a flow-cryostat, while quartz windows on both sides give optical access and allow for the strait-forward adjustment of all further components at ambient conditions, precluding any complicated in-vacuum optical adjustments. Versatile optical concepts can be easily implemented and modified to the experimental needs in that way. With our unique setup, we harness the remarkable time resolution obtained by single Terahertz voltage pulses for time-resolved electroluminescence experiments. In particular, we will unravel field-induced electroluminescence with femtosecond time resolution, while simultaneously addressing samples with the atomic resolution provided by scanning probe microscopy.

O 72.4 Wed 18:00 Poster D

Time-Resolved Measurements with THz-STM on Semiconducting $MoTe_2$ — •Vibhuti Rai, Junyoung Sim, Carlos Williams, Sergey Trishin, Nils Bogdanoff, Tom Seifert, Tobias Kampfrath, Christian Lotze, and Katharina J. Franke — Freie Universität Berlin, Department of Physics, Arnimallee 14, 14195 Berlin, Germany

To access time scales of elementary excitations, pump-probe schemes using ultrashort laser pulses are the method of choice. Combining these with scanning tunnelling microscopy (STM) would not only give the required time resolution but also provide spatial resolution on the sub nanometer scale [1]. In this work, we use a home-built THz setup combined with an STM working at 5K in ultra-high vacuum. We study the charge carrier dynamics of the semiconducting $MoTe_2$ bulk sample [2]. An infrared pump pulse is used to excite the sample across its direct band gap of 1eV. To probe the dynamics, we use the THz field as a transient bias that modulates the tunnelling junction. By delaying the IR pulse with respect to the THz pulse, we explore the dynamics of the excited states and also use them further to spatially resolve and study surface defects.

O 72.5 Wed 18:00 Poster D

Coupling Single-Cycle Terahertz Fields into a STM: Characterization and Time-Resolved Measurements — •Junyoung Sim, Vibhuti Rai, Carlos Williams, Sergey Trishin, Nils Bogdanoff, Tom Seifert, Tobias Kampfrath, Christian Lotze, and Katharina Franke — Freie Universtiät Berlin, Department of Physics, Arnimallee 14, 14195 Berlin, Germany

Scanning tunneling microscopy (STM) is an ideal tool to study the structure and electronic properties of atomic-scale nanostructures on surfaces. Excitation-relaxation dynamics in these systems occur typically on timescales of picoseconds or even faster, which is beyond the electronic bandwidth of STM. To explore this ultrafast regime, Cocker et al. coupled ultrashort terahertz (THz) pulses into a STM junction and implemented a pump-probe detection scheme [1].

In this work, we follow a similar approach by setting up a THz source and coupling it to a 5K- STM. Here, we provide a preliminary characterization of our set up with a few benchmarking experiments. We measure the near-field THz pulse that modulates the photocurrent excited by a preceding optical pulse between STM tip and Ag(111) surface. We also probe field emission resonance by the transient voltage from the THz pulse. We then perform infrared-pump THz-probe measurement to investigate the dynamics of excitation processes across the bandgap on the semiconducting transition metal dichalcogenide MoTe2 [2].

[1] Cocker et al. Nature Photonics 15, 558-569 (2021)

[2] Sankar et al. Chem. Mater., 29, 2, 699-707 (2017)

O 72.6 Wed 18:00 Poster D

Azobenzene derivatives in the vicinity of Cobalt islands on a gold surface — •Bensu Günay, Christophe Nacci, and Leonhard Grill — Chemistry department, University of Graz, Graz, Austria

The purpose of this study is to investigate whether plasmonic effects can affect the isomerization behavior of molecular switches in the vicinity of cobalt islands on a metallic surface. We have chosen azobenzene derivatives, a prototypical molecular switch, as functional units and an Au(111) surface as substrate, which was studied by low-temperature scanning tunneling microscopy. The combination of these molecules with the gold surface is well known [1] and therefore represents a suitable model system. While in previous studies, switching of the molecules was typically induced by voltage pulses [1], we have induced isomerization here by light. Importantly, Cobalt islands were grown on the surface with the purpose of localized plasmonic effects around these islands. The surface, which was illuminated for different durations at various wavelengths, was then studied in view of different switching rates in the vicinity of the Cobalt islands as compared to remote locations.

[1] M. Alemani et al., JACS 128, 14446 (2006)

O 72.7 Wed 18:00 Poster D

Characterization of C_{60} derivatives by Atomic Force Microscopy — •Paul Mosser[1], Antoine Hinaut[1], Thilo Glatzel[1], Shi-xia Liu[2], Silvio Decurtins[2], and Ernst Meyer[1] — [1]Department of Physics, University of Basel, Klingelbergstrasse 82, CH 4056 Basel, Switzerland — [2]Department of Chemistry and Biochemistry, University of Bern, Freiestrasse 3, CH 3012 Bern, Switzerland

Pristine C_{60} as a rigid spherical molecule is a 3D strong electron acceptor. The absorption spectrum of fullerene changes greatly as it approaches a metal surface, and a significant dependence on the distance between the molecule and the substrate is expected. For this purpose, functional groups with the binding affinity of Au substrates can be covalently attached to the fullerene core with any regioisomeric pattern and used to tune the coupling of the compounds with the local plasmons. In this work, N-pyridyl-3,4-fulleropyrrolidine (C_{60}-Py) is studied as it is supposed to form Au-N bonds on a gold surface. C_{60}-Py were evaporated on an Au(111) surface and characterized using nc-AFM and KPFM at room temperature and under UHV conditions. The high-resolution AFM images of C_{60}-Py islands show a periodicity difference compared to pristine C_{60} islands showing that the pyridyl-pyrrolidine group has an influence on the self assembly on Au(111). KPFM measurements show a contrast in CPD between Au, the pristine C_{60} islands and the C_{60}-Py islands of 200mV.

O 72.8 Wed 18:00 Poster D

Understanding photocurrents in plasmonic tunnel junctions driven by continuous-wave optical fields — •Henrik Wiedenhaupt[1], Chenfang Lin[1,2], Adnan Hammud[1], Martin Wolf[1], Takashi Kumagai[1,3], and Melanie Müller[1] — [1]Fritz-Haber Institute of the Max-Planck Society, Berlin, Germany — [2]Hunan University, Changsha, China — [3]Institute for Molecular Science, Okazaki, Japan

Understanding the mechanism by which optical radiation is converted into direct electrical (DC) current through plasmonic nanogaps is important for the future design of optical rectennas. One question concerns the transition from quantum (photon-driven) to classical (field-driven) optical rectification in continuous-wave (cw) driven tunnel junctions. The transition from field- to photon-driven rectification can be characterized by (i) the relation of the tunneling time to the light period (adiabaticity criterion), and (ii) by the relation of the photon energy to the induced optical voltage (quantum detection criterion). The transition between these regimes depends on the size of the tunneling gap, its current-voltage nonlinearity, and the frequency and intensity of the light field. Here, we investigate photocurrents through a cw-driven plasmonic junction of a scanning tunneling microscope at optical frequencies. We aim to identify the transition from field- to photon-driven tunneling by studying the dependence of photocurrent-voltage curves on the STM gap size and the frequency and intensity of the driving cw-laser field.

O 72.9 Wed 18:00 Poster D

Excitation of single 4f metal-organic molecules with light — •Adrian Ebert[1], Lukas Gerhard[1], Lia Mayer[1], Shuran Liu[1], Marjan Krstic[2], Barbora Brachnakova[3], Senthil Kumar Kuppusamy[3], Julia Feye[4], Peter W. Roesky[4], and Wulf Wulfhekel[1] — [1]Institute for Quantum Materials and Technology, KIT — [2]Institute for Theoretical Solid State Physics, KIT — [3]Institute of Nanotechnology, KIT — [4]Institute for Organic Chemistry, KIT

We study light emitting 4f metal-organic complexes in a low temperature, ultra-high vacuum scanning tunneling microscope (STM). While the system has been successfully used to couple light out emitted from single molecules, it's also suitable to couple light in using the same optical path. The addition of a pulsed light source for coupling light into the STM junction will greatly extend the possibilities to study the exciton dynamics.

4f metal-organic complexes have an immense advantage in comparison to purely organic chromophores or chromophores based on d-electrons, because the 4f states are highly localized hampering non-radiative recombination leading to long lifetimes of the excited states. They combine the extremely sharp emission lines including spin-orbit coupling and hyperfine coupling with functionalization by the ligands to enhance the coupling to the photon field. In future opto-electronic quantum devices the complexes can act as single photon emitters and their up- and down-conversion can be used to couple photons of different wavelength.

O 72.10 Wed 18:00 Poster D

Plasmonic probe preparation for tip-enhanced Raman spectroscopy and photoluminescence on single molecules — •Petr Kahan, Amandeep Sagwal, Rodrigo Ferreira, and Martin Švec — Institute of Physics, Czech Academy of Sciences; Cukrovarnická 10/112, CZ16200 Praha 6, Czech Republic

Various approaches to preparation of plasmonic tips for tip-enhanced Raman scattering (TERS) and photoluminescence (PL) are known [1,2] and were applied to spectroscopy on single molecules. Such preparation procedures are rather complex and time-consuming, hence not adequate for routine applications where more tips need to be prepared and tested efficiently at low input costs. Here we explore a new methodology of the TERS/PL tips fabrication, based on etching [3] silver and gold wires, and subsequent ion sputtering [4], aiming to obtain high purity surface of the resulting tips, which is required for good plasmonic activity and avoiding any spurious Raman scattering from the tip contamination. We optimize the procedure based on screening of the tip quality using the scanning electron microscopy, being able to improve reproducibility and prospects of a successful application of such tips in demanding experiments measuring single-molecule luminescence and Raman spectroscopy with scanning probe techniques.

[1] Li, M. et al. Journal of Raman Spectroscopy 47, 808-812 (2016). [2] Yang, B. et al. JPCC 122, 16950-16955 (2018). [3] Walker, P. & Tarn, W. H. CRC handbook of metal etchants. (CRC press, 1990). [4] Hoffrogge, P., Kopf, H. & Reichelt, R. J Appl Phys 90, 5322-5327 (2001).

O 72.11 Wed 18:00 Poster D

Theory of photo and electroluminescence in a scanning tunneling microscope — •Tomáš Neuman[1], Sofia Canola[1], Anna Roslawska[2], Katharina Kaiser[3], Alex Boeglin[3], and Guillaume Schull[3] — [1]Institute of Physics, Czech Academy of Sciences, Cukrovarnická 10, 16200 Prague, Czech Republic — [2]Max-Planck Institute for Solid-State Research, DE-70569 Stuttgart, Germany. — [3]Université de Strasbourg, CNRS, IPCMS, UMR 7504, F-67000 Strasbourg, France

I will present a theoretical perspective on a microscopy technique that combines the atomic-scale resolution of a scanning-tunneling microscope (STM) with optics. I will explain how this method, taking advantage of phenomena such as the optical Stark shift or plasmonic Purcell effect, can reveal excited-state properties of nanoscale samples with unprecedented spatial resolution. Besides introducing the method and the mechanism of STM-induced luminescence (STML) applied to dye molecules, I will show theoretical considerations concerning some of its applications and extensions including the study of vibronic features in the STML spectra and imaging, phototautomerization of a single molecule, and a theoretical view on correlated photon emission from electrically driven organic molecules in an STM.

O 72.12 Wed 18:00 Poster D

Theoretical Study of Electronic and Optical Properties in Modified Graphene Nanoribbons — •Jian Cheng Wong[1], Jiang Song[2], Sofia Canola[1], Alex Boeglin[2], Guillaume Schull[2], and Tomáš Neuman[1] — [1]Institute of Physics, Czech Academy of Sciences, Cukrovarnická 10, 16200 Prague, Czech Republic — [2]Université de Strasbourg, CNRS, IPCMS, UMR 7504, F-67000 Strasbourg, France

The electronic and optical properties of graphene nanoribbons (GNRs) depend on geometric configurations such as width and edge structure. The spin properties of these GNRs have been widely studied both theoretically and by scanning tunneling microscopy experiments. However, studies exploring their optical properties remain scarce. Scanning tunneling microscopy-induced luminescence (STML) provides sub-nanometer-scale spatial resolution to study these properties using light emitted from the sample. Pioneering STML studies [1] on modified GNRs unravel localized excitations that exhibit interesting optical properties. Despite this recent experimental progress, comprehensive theoretical description of these optical excitations in structures of varying size and geometry remains to be theoretically explored. Using first-principles electronic structure methods, we elucidate the electronic and optical properties of these localized excitations.

[1] Song et al., Science, 379(6636), 1049-1054 (2023).

O 72.13 Wed 18:00 Poster D

Probing Deformations of Heptahelicene Molecules via

TERS — •Amandeep Sagwal[1,2], Rodrigo Cezar De Campos Ferreira[1], Jiří Doležal[1,3], and Martin Švec[1,4] — [1]Institute of Physics, Czech Academy of Sciences; Czech Republic — [2]Faculty of Mathematics and Physics, Charles University; Czech Republic — [3]Institute of Physics, École Polytechnique Fédérale de Lausanne; Switzerland — [4]Institute of Organic Chemistry and Biochemistry, Czech Academy of Sciences; Czech Republic

The Raman scattering from single molecules can be substantially enhanced by placing them within the nanocavity between a sharp plasmonic tip and sample using the Tip-Enhanced Raman Spectroscopy (TERS) technique [1][2]. TERS in ultrahigh vacuum and cryogenic conditions can also reveal the relations between adsorption geometry, electronic states, and vibrational modes of single molecules [3]. Here we study the behaviour of heptahelicene (BA7H) molecules on Ag (111) showing two different conformers i.e. so-called bright and dark [4]. TERS measurements are conducted using a laser with wavelength 633 nm, along with the lifting of molecules from the substrate. We observe significant energy shifts in the Raman peaks corresponding to the geometrical changes of the molecules and attempt to assign them to the normal vibrational modes of the molecules calculated by TD-DFT. [1] Lee, J., Crampton, K.T., Tallarida, N. et al. Nature 568, 7882 (2019). [2] Jaculbia, R. B. et al. Nat. Nanotechnol. 15, 105110 (2020). [3] R. C. de Campos Ferreira, et al. arXiv:2310.12546, (2023) [4] O. Stetsovych, J. Am. Chem. Soc., 140, 3, 940946 (2018)

O 73: Overview Talk Karina Morgenstern

Time: Thursday 9:30–10:15 Location: HE 101

Invited Talk O 73.1 Thu 9:30 HE 101

Ice structures and dynamics on surfaces investigated on the local scale — •Karina Morgenstern — Physical Chemistry I, Ruhr-Universität Bochum, Germany

The importance of ice in Earth's atmosphere is crucial for its climate and hydrological cycle. For instance, sea salt aerosols play an important role in atmospheric chemistry. Though such general principles of atmospheric chemistry are well established on a macroscopic level, we have only a poor molecular-scale understanding of the processes involved. This limits our ability to model quantitatively processes such as how atmospheric ice influences clouds and climate. Investigating model systems by scanning tunnelling microscopy promises to fill this lack of knowledge, for instance, how water ice nucleates on different surfaces and in the presence of polar impurities. This talk will give a general introduction to the challenges and promises of using surface science tools to investigate the structure and dynamics of ice nucleation. It will be shown how water nucleates and grows on metal surfaces starting from individual molecules and how the presence of organic multi-polar molecules and cations alters this growth. By investigating the attachment of individual water molecules to these particles by low-temperature scanning tunnelling microscopy, we gain microscopic insight into the interaction of water with charged particles. Our STM study gives an unprecedented view of water ice interacting with polar species. Moreover, we present an example, of how the interaction influences the reactivity of photo-induced dissociation of chlorobenzene, a typical process induced in clouds by the sun's action.

O 74: Focus Session: $SrTiO_3$: A Versatile Material from Bulk Quantum Paraelectric to 2D Superconductor I (joint session TT/KFM/MA/O)

Strontium titanate ($SrTiO_3$) is a paradigmatic material that plays an important role in various fields of solid-state physics, surface science and catalysis: The pure bulk phase is a wide-band-gap semiconductor that upon cooling becomes a textbook quantum paraelectric. When slightly doped, $SrTiO_3$ turns into a Fermi-liquid-type metal that becomes superconducting at extremely low charge carrier density. $SrTiO_3$-based surfaces and interfaces host un-conventional electronic states such as quasi-two-dimensional electron liquid, magnetism and superconductivity. Despite intensive studies over the past decades, $SrTiO_3$ continues to reveal surprising new phenomena that challenge the established views on this material. To this end achieving light-induced nonequilibrium states and the recent preparation of a 2D oxide based on $SrTiO_3$ opens new playgrounds for research. This Focus Session will present exciting developments in the study of electronic states that are based on the peculiar properties of $SrTiO_3$.

Please note that this Focus Session comprises four parts: Posters are presented within the TT poster session TT58 (Wed 15:00-18:00, poster area E). Invited talks are compiled in the session TT62 (Thursday, 9:30 to 12:45, H0104), Contributed talks will be presented in sessions TT72 (Thursday 15:00-18:00, H0104) and TT83 (Fri 9:30-12:30, H0104).

Organizers: Rossitza Pentcheva, University of Duisburg-Essen, Marc Scheffler, University of Stuttgart

Time: Thursday 9:30–12:45 Location: H 0104

Invited Talk O 74.1 Thu 9:30 H 0104

Ferroelectricity and Superconductivity in $SrTiO_3$ — •Susanne Stemmer — University of California, Santa Barbara, USA

Polar superconductors have attracted significant interest for their potential to host unconventional superconductivity. One candidate is doped strontium titanate ($SrTiO_3$), which can undergo successive ferroelectric and superconducting transitions. Recent experimental observations of a factor of two enhancement of the superconducting transition temperature in ferroelectric samples and the fact that both ferroelectricity and superconductivity vanish around the same carrier density, hint at common physical interactions relevant for both phenomena. We will discuss our understanding of ferroelectricity in strained $SrTiO_3$ films, and experiments aimed at elucidating the connection between superconductivity and ferroelectricity.

Although the ferroelectric transition of strained, undoped $SrTiO_3$ is usually described as a classic displacive transition, we show that it has pronounced order-disorder characteristics. Increasing the carrier concentration causes polar nanodomains to break up into smaller clusters. (Local) polar order appears to be essential to the superconducting state. For example, in strained $SrTiO_3$ films, suppression of superconductivity is correlated to the destruction of the (global) ferroelectric state, either by overdoping, by decreasing the film thickness or by alloying large amounts of a rare earth ion. We discuss how the length scale of polar order emerges as an important parameter in controlling the superconductivity of $SrTiO_3$.

Invited Talk O 74.2 Thu 10:00 H 0104

Dilute superconductivity in doped strontium titanate — •Kamran Behnia — LPEM-ESPCI, Paris, France

Dilute superconductivity survives in bulk strontium titanate when the Fermi temperature falls well below the Debye temperature. The onset of the superconducting dome is dopant dependent. The threshold density fpr superconductivity is much lower for mobile electrons introduced by removing oxygen atoms compared to those brought by substituting Ti with Nb. Our study of quantum oscillations reveals a difference in the band dispersion between the dilute metals made by these doping routes and our band calculations demonstrate that the rigid band approximation does not hold when mobile electrons are introduced by oxygen vacancies. We identify the band sculpted by these vacancies as the exclusive locus of superconducting instability in the ultradilute limit.

Invited Talk O 74.3 Thu 10:30 H 0104

Polarons and Excitons in quantum-paraelectric SrTiO$_3$ — •Cesare Franchini — University of Vienna & Bologna

SrTiO$_3$ stands as one of the most extensively investigated materials, captivating attention due to its distinctive electronic properties emerging from its quantum paraelectric nature. Positioned on the cusp of various collective phases, this material holds significant potential for exploitation in electronic and optical applications. In this presentation, we delve into the biphonon collective behaviors and quasiparticle properties of SrTiO$_3$ in both bulk and reduced dimensions, leveraging a combination of single-particle and many-body methods supported by machine learning techniques. Our exploration commences with an examination of temperature-dependent quantum and anharmonic effects employing a synergy of machine-learned potentials and the stochastic self-consistent harmonic approximation [1,2]. Shifting focus, we investigate the electron-phonon-driven formation of polarons, scrutinizing the interplay between spatially localized small polarons and dispersive large polarons in both bulk SrTiO$_3$ [3,4] and on the bulk-terminated SrTiO$_3$(001) surface [5,6]. In conclusion, our study delves into the optical and excitonic properties, with particular emphasis on the emergence of strongly bound excitonic peaks in the monolayer limit [7,8].

[1] Adv. Quantum Technol. 6 (2023) 2200131
[2] Phys. Rev. Mater. 7 (2023) L030801
[3] Phys. Rev. B 91 (2015) 085204
[4] npj Computational Materials 125 (2022)
[5] Phys. Rev. Mater. 3, 034407 (2019); Phys. Rev. B 103 (2021) L241406
[6] Phys. Rev. Mater. 7 (2023) 064602
[7] Phys. Rev. Mater. 5 (2021) 074601
[8] arXiv:2303.14830

15 min. break

Invited Talk O 74.4 Thu 11:15 H 0104

Controlling ferroelectrics with light — •Andrea Cavalleri — Max Planck Institute for the Structure and Dynamics of Matter, Hamburg — Department of Physics, University of Oxford

I will discuss how irradiation of ferroelectrics with intense, far and mid-infrared pulses, which are made resonant with certain phonon modes, can be used to manipulate the ferroelectric polarization. Two cases have been identified so far. On the one side, irradiation of a low temperature ferroelectric phase (e.g. in LiNbO$_3$) can achieve switching of the polarization. In incipient ferroelectric phases (e.g. in SrTiO$_3$), can lead to the formation of a long range ordered phase with stronger ferroelectricity than the paraelectric ground state. The microscopic physics of these phenomena are only in part clear, and I will discuss progress in this area.

Invited Talk O 74.5 Thu 11:45 H 0104

Terahertz electric field driven dynamical multiferroicity in SrTiO$_3$ — •Stefano Bonetti — Ca' Foscari University of Venice, Venice, Italy

In recent years, the ultrafast dynamical control and creation of novel ordered states of matter not accessible in thermodynamic equilibrium is receiving much attention. Among those, the theoretical concept of dynamical multiferroicity has been introduced to describe the emergence of magnetization by means of a time-dependent electric polarization in non-ferromagnetic materials. However, the experimental verification of this effect is still lacking. Here, we provide evidence of room temperature magnetization in the archetypal paraelectric perovskite SrTiO$_3$ due to this mechanism. To achieve it, we resonantly drive the infrared-active soft phonon mode with intense circularly polarized terahertz electric field, and detect a large magneto-optical Kerr effect. A simple model, which includes two coupled nonlinear oscillators whose forces and couplings are derived with ab-initio calculations using self-consistent phonon theory at a finite temperature, reproduces qualitatively our experimental observations on the temporal and frequency domains. A quantitatively correct magnitude of the effect is obtained when one also considers the phonon analogue of the reciprocal of the Einstein - de Haas effect, also called the Barnett effect, where the total angular momentum from the phonon order is transferred to the electronic one. Our findings show a new path for designing ultrafast magnetic switches by means of coherent control of lattice vibrations with light.

O 74.6 Thu 12:15 H 0104

Emergence of a quantum coherent state at the border of ferroelectricity in SrTiO$_3$ — •Matthew Coak[1,2], Charles Haines[2], Cheng Liu[2], Stephen Rowley[2,3], Gilbert Lonzarich[2], and Siddharth Saxena[2] — [1]School of Physics and Astronomy, University of Birmingham, Birmingham, UK — [2]Cavendish Laboratory, University of Cambridge, Cambridge, UK — [3]Centro Brasileiro de Pesquisas Fisicas, Rio de Janeiro, Brazil

SrTiO$_3$ exists on the border of ferroelectricity in the vicinity of a quantum critical point (QCP). It is this proximity to a quantum critical point and the fluctuations associated with it which are responsible for SrTiO$_3$'s strikingly non-classical dielectric susceptibility.

I will discuss our results utilising ultra-high precision measurements of the the dielectric susceptibility to demonstrate an unconventional quantum paraelectric state exhibiting ‘order by disorder’ - a fluctuation-induced enhancement of electric polarization up to a coherence temperature T^*. We show that in the vicinity of T^* this phenomenon can be understood quantitatively in terms of the hybridization of the critical electric polarization field and the volume strain field of the lattice.

We argue that this coherent optical-acoustic phonon state emerges from the QCP and is critical to our understanding of the mechanisms behind the quantum criticality and the phenomena resulting from it. At still lower temperatures, well below T^*, we observe a breakdown of this unconventional form of quantum paraelectricity and the emergence of a new instanton liquid phase.

O 74.7 Thu 12:30 H 0104

Dynamics of the critical phonon modes in quantum paraelectric SrTiO$_3$ — •Shiyu Deng[1,2], Charles S. Haines[1,3], Matthew J. Coak[1,4], Alexandre Ivanov[2], Andrea Piovano[2], Andrew R. Wildes[2], and Siddharth S. Saxena[1] — [1]Cavendish Laboratory, University of Cambridge — [2]Institut Laue-Langevin — [3]University of East Anglia — [4]University of Birmingham

The proximity of SrTiO$_3$ to a ferroelectric quantum critical point (FE QCP) has become a promising new branch of the study of quantum critical phenomena. New forms of quantum order have been reported in SrTiO3 different from the quantum paraelectric state via dielectric measurements.

We report our recently performed triple-axis inelastic neutron scattering experiments on single-crystal SrTiO$_3$ at the temperature and pressure region of interest. These were the first direct measurements deep into the enigmatic 'quantum polar-acoustic state' in the vicinity of the FE QCP. Measurements are taken at and around $q = 0$ in multiple directions in reciprocal space to explore the transverse acoustic and soft optical phonon modes and their hybridization. In addition, we explore how pressure affects the underlying phonon modes in SrTiO$_3$. Our observations address directly the coupling of the soft optical mode with the acoustic phonons, and its response to external pressure. We believe this could help us understand the importance of anharmonic lattice dynamics and quantum fluctuations in SrTiO$_3$.

O 75: Focus Session: Wetting on Adaptive Substrates I (joint session CPP/DY/O)

The focus session aims to discuss recent developments in the wetting dynamics of adaptive, deformable, and switchable surfaces.

Time: Thursday 9:30–11:00 Location: H 0107

Invited Talk O 75.1 Thu 9:30 H 0107

Extraordinarily slippery liquid-repellent surfaces using self-assembled monolayers — •Robin Ras — Aalto University, Espoo, Finland

Water-repellent surfaces have the attractive property of staying dry, and find applications in self-cleaning, anti-icing, anti-fogging and much more. Liquid-repellent surfaces, especially smooth solid surfaces with covalently grafted flexible brushes or alkyl monolayers, are the focus of an expanding research area.[1] Surface-tethered flexible species are highly mobile at room temperature, giving solid surfaces a unique liquid-like quality and unprecedented dynamical repellency.

We challenge two common assumptions on liquid-repellency.[2] It is generally assumed that water-repellent surfaces requires hydrophobicity. We demonstrate a hydrophilic surface with the unusual combination of low sliding angle and low contact angle. Surface heterogeneity is generally acknowledged as the major cause of increased contact angle hysteresis and friction of droplets. Here we challenge this long-standing premise for chemical heterogeneity at the molecular length scale.

Furthermore, we demonstrate world's most slippery surface, by combining self-assembled monolayers and surface structuring. Finally, by a suitable surface texture, we can trap a thin air layer for months, opening new strategies for underwater applications.[3]

[1] Nature Chemistry Reviews (2023) https://doi.org/10.1038/s41570-022-00455-w[2] Nature Chemistry (2023) https://doi.org/10.1038/s41557-023-01346-3 [3] Nature Materials (2023) https://doi.org/10.1038/s41563-023-01670-6

O 75.2 Thu 10:00 H 0107

Volatile binary mixtures on polymer brushes — •Jan Diekmann und Uwe Thiele — Institut für Theoretische Physik, Universität Münster, 48149 Münster, Germany

We present a mesoscopic thin-film model in gradient dynamics form for binary liquid mixtures on brush-covered substrates incorporating volatility in a narrow gap. Thereby, we expand models established in [1, 4-6] by incorporating two substances present in each of three bulk phases - liquid, brush and gas. We discuss the different contributions to the free energy, thereby employing Flory-Huggins theory of mixing for the condensed phases and assuming ideal gases for the vapor phase. Interface energies are modeled as linear interpolations of known limiting cases. The resulting six-field model is then analyzed with numerical time simulations showing results with a focus on lateral concentration gradients, notably at the contact line.

[1] S. Hartmann, C. Diddens, M. Jalaal, and U. Thiele. JFM 960, 2023. doi: 10.1017/jfm.2023.176. [2] S. Hartmann, J. Diekmann, D. Greve, and U. Thiele. 2023. doi: 10.48550/ARXIV.2311.07307. [3] S. Schubotz, Q. A. Besford, S. Nazari, P. Uhlmann, E. Bittrich, J.-U. Sommer, and G. K. Auernhammer. Langmuir, 39, 2023. doi: 10.1021/acs.langmuir.2c03009. [4] L. A. Smook, G. C. R. van Eck, and S. de Beer. Macromolecules, 53, 2020. doi: 10.1021/acs.macromol.0c02228. [5] U. Thiele and S. Hartmann. EPJ-ST 229, 2020. doi:10.1140/epjst/e2020-900231-2. [6] Özlem Kap, S. Hartmann, H. Hoek, S. de Beer, I. Siretanu, U. Thiele, and F. Mugele. JFM 158, 2023. doi: 10.1063/5.0146779.

O 75.3 Thu 10:15 H 0107

Wetting Phenomena in Hierarchically Porous Silicon: How Experiments and 2D Fluid-Dynamic Simulations Complement Each Other — •Stella Gries[1,2], Stefan Schulz[1], Marc Thelen[1,2], Silja Flenner[3], Imke Greving[3], and Patrick Huber[1,2] — [1]Institute for Materials and X-ray Physics, Hamburg University of Technology, Hamburg, Germany — [2]Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany — [3]Institute of Materials Physics, Helmholtz Zentrum Hereon, Geesthacht, Germany

Nature is an expert in designing highly efficient, multi-functional (hybride-)materials such as hierarchically capillary systems in respiratory systems or plants. These systems achieve large internal surfaces while allowing an optimized mass transport. This is used in plants to perform capillarity- induced motions, transport substances to reaction sites and remove educts from chemical processes. We are aiming to mimic such systems with artificially produced hierarchically porous silicon. The bimodal, hierarchical structure leads to a different imbibition behavior than porous systems with a monomodal pore-size distribution. Therefore, we used 2D finite element fluid dynamic simulations to achieve deeper insights into single-pore events and competing Laplace pressures in pore sections with distinct pore sizes. The simulations are related to the experimental results from dilatometry, mass-uptake and synchrotron-based, in-situ X-ray radiography imbibition experiments. This allows a complete description of the transport phenomena and will help us to tailor the material for applications in capillarity-driven pumps or energy harvesting from natural processes.

O 75.4 Thu 10:30 H 0107

Percolation in Networks of Liquid Diodes — Camilla Sammartino, Yair Shokef, and •Bat-El Pinchasik — Tel Aviv University, School of Mechanical Engineering, Israel

Liquid diodes are surface structures that facilitate the spontaneous flow of liquids in a specific direction. In nature, they are used to increase water collection and uptake, reproduction, and feeding. However, pump-free large networks with directional properties are exceptional and are typically limited up to a few centimeters. Here, we simulate, design, and 3D print networks consisting of hundreds of liquid diodes. We provide structural and wettability guidelines for directional transport of liquids through these networks and introduce percolation theory in order to identify the threshold between a connected network, which allows fluid to reach specific points, and a disconnected network. By constructing well-defined networks with uni- and bidirectional pathways, we experimentally demonstrate the applicability of models describing isotropically directed percolation. We accurately predict the network permeability and the liquid final state. These guidelines are highly promising for the development of structures for spontaneous, yet predictable, directional liquid transport. In addition, they comprise an initial realization of complex liquid circuits, analogoues to electric circuits.

O 75.5 Thu 10:45 H 0107

Wetting underneath droplets on an oily surface — •Shiva Moradimehr and Kirsten Harth — Fachbereich Technik, TH Brandenburg, Brandenburg a. d. Havel

When a liquid drop impacts on a smooth surface, a thin layer of air evolves between the drop and the surface. The drop deforms under the influence of the ambient air that needs to be squeezed out before the drop can touch the substrate. The actual air layer profile depends on the impact velocity, ambient gas, drop liquid as well as the deformability of the substrate. For small impact Weber number, the air film ruptures before drop rebound. The wetting front connecting the drop and layer liquids propagates at velocities of few meters per second.

We study the wetting front velocity for droplets impacting on hard substrates covered by thin oil layers using high-speed interferometry at oblique incidence. The impact velocity, viz. the thickness of the entrained air layer, and the oil layer properties are varied. Both a model for contact spreading on a thin film or an film-rupture based model are initial candidates to describe the wetting front propagation.

O 76: Focus Session: 2D Transition Metal Carbides, Nitrides and Carbonitrides II (joint session DS/MM/O)

Surface functionalization & defects and Water & gas Interaction

Time: Thursday 9:30–12:15 Location: A 053

Invited Talk O 76.1 Thu 9:30 A 053

Computational insights into the surface functionalization and defects in MXenes — •Hannu-Pekka Komsa — Microelectronics Research Unit, University of Oulu, Finland

Two-dimensional (2D) transition metal carbide and nitride MXenes offer rich chemistry with extraordinary properties. The surface of MXenes is terminated by -O, -OH, and -F groups during the synthesis. However, there is limited understanding on how the surface composition depends on the synthesis conditions and on the type of MXene (transition metal, carbon vs. nitrogen, and the number of atomic layers).

We developed a multi-scale computational scheme to simulate the distribution and the thermodynamically favorable composition of the functional groups on the MXene surfaces. We considered the most popular MXene systems such as Ti$_2$C, Ti$_3$C$_2$, Ti$_2$N, Ti$_4$N$_3$, Nb$_2$C, and Nb$_4$C$_3$. The surface accommodates mixtures of functional groups for all considered MXenes with similar distributions regardless of the type of metal, carbon or nitrogen species and number of atomic layers. These findings are shown to be important for an accurate prediction of properties and stability of these materials. Next, we investigated the propensity of metal vacancy formation under synthesis conditions. The results suggest that vacancy formation is crucial step in initiating MXene oxidation. Finally, to support experimental characterization, we simulated the Raman spectra as a function of functional group distribution, temperature, and disorder, and the materials' response to electron irradiation in electron microscope.

O 76.2 Thu 10:00 A 053

In situ monitoring of surface termination of Ti3C2Tx MXene with Raman spectroscopy — •Julian Plaickner[1,2], Tristan Petit[2], Peer Bärmann[2], Thorsten Schultz[2,3], Norbert Koch[2,3], and Norbert Esser[1,4] — [1]Technische Universität Berlin, Institut für Festkörperphysik, Hardenbergstrasse 36, 10623 Berlin — [2]Hardenbergstrasse 36 — [3]Institut für Physik & IRIS Adlershof, Humboldt Universität zu Berlin, 12489 Berlin — [4]Leibniz Institut für Analytische Wissenschaften ISAS eV, Schwarzschildstrasse 8, 12489 Berlin

In the last years, MXenes have attracted attention due to an interesting combination of physical properties. A better knowledge of the surface chemistry of MXenes is critical for the implementation in applications. A promising investigation approach is annealing-induced desorption of surface terminations, because it might make the surface very active for further functionalizations. In this work, the effect of annealing on the Ti3C2Tx MXene surface chemistry in UHV has been investigated with Raman spectroscopy in ultra-high vacuum. Changes in the Raman spectra are correlated with the desorption of the major part of the F surface termination at 650 °C. Most of the observed spectral features show a significant broadening already before annealing. We attribute this to the intrinsic disordered nature of the MXenes due to a mixed surface termination and a broad background associated to electronic Raman scattering. With this study we identified of the vibrational signatures associated with F-termination and provided new insights into the interpretation of the vibrational spectra of Ti3C2Tx MXenes.

O 76.3 Thu 10:15 A 053

Hydrogenation as a mean to remove halogen functionalization from of Ti3C2Tz thin films — •Silvano Lizzit[1], Florian Brette[2,3], Hanna Pazniak[4], Monika Shied[1], Paolo Lacovig[1], Florent Boucher[3], Vincent Mauchamp[2], and Rosanna Larciprete[5] — [1]Elettra-Sincrotrone Trieste, Trieste (I) — [2]Uni. Poitiers, ISAE-ENSMA, CNRS, PPRIME, Poitiers (F) — [3]CNRS-IMN, Nantes (F) — [4]Uni. Grenoble Alpes, CNRS, Grenoble INP, Grenoble (F) — [5]CNR- ISC, Roma (I)

The nature and the density of the chemical groups terminating the MXenes surface determine their electronic and chemical properties. Therefore, the manipulation of the surface termination allows to change the way these compounds interact with the surrounding environment. In this study we explored the possibility of using the functionalization with H atoms as a mean to modify the surface termination of T3C2Tz thin films. To this aim we used photoelectron spectroscopy with synchrotron radiation to investigate the surface reactions induced by the exposure to atomic hydrogen. Simulation of the valence band spectra by DFT calculations combined with the analysis of the core level spectra allowed us to elucidate the changes in the chemical bonding determined by the interaction with H atoms. It turned out that, in addition to the formation of C-H and -O-H bonds, sample hydrogenation removed the halogen terminating atoms, decreasing the F and Cl concentrations to less than 20% of the initial values. After removing the hydrogenated phases at 400 K, the dehydrogenated surface exhibited a chemical reactivity higher than that of the pristine sample.

O 76.4 Thu 10:30 A 053

Chemical characterization of defects in Ti3C2Tx MXenes by soft X-ray spectroscopy — •Arsène Chemin[1], Zoé Dessoliers[1], Robert W. Lord[2], Yury Gogotsi[2], and Tristan Petit[1] — [1]Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Berlin, Germany — [2]A.J. Drexel Nanomaterials Institute, Drexel University, Philadelphia, Pennsylvania, USA

MXenes offer a wide spectrum of properties with numerous fields of application. Yet, these properties can be strongly affected by defects, and a detailed understanding of their nature and structure is essential. Despite many observations of such defects, grasping their chemical natures is challenging. In this work, high-resolution X-ray Photoelectron Spectroscopy (HR-XPS) and, at the Ti L- and C,O K-edges, soft X-ray Absorption Spectroscopy (XAS), performed at the BESSY II synchrotron in Berlin, are used to investigate the chemical nature of defects in Ti3C2Tx MXenes.

15 min. break

Invited Talk O 76.5 Thu 11:00 A 053

MXenes as materials for carbon capture, storage, and usage technologies: Computational insights & predictions — •Francesc Viñes — Departament de Ciència de Materials i Química Física & Institut de Química Teòrica i Computacional (IQTCUB), Universitat de Barcelona, c/ Martí i Franquès 1-11, 08028 Barcelona, Spain

Environmentally-wise, pristine MXenes have been proposed as suited materials for carbon capture and storage (CCS) technologies, as predicted by computational density functional theory (DFT) simulations on suited models.1 Such MXenes display a high affinity towards carbon dioxide (CO2), strongly binding it, and activating it by charge transfer, gaining a bent CO2 with elongated bonds. The CCS capabilities highlighted through kinetic phase diagrams (KPD) were confirmed by experiments,2 showing a high CO2 uptake due to the MXene high surface area, specially high when MXene is made of light transition Ti or V metals. Furthermore, given the CO2 activation pristine MXenes can be used in carbon capture and use (CCU) technologies, acting as heterogeneous catalysts. A recent DFT study shows MXenes use as heterogeneous catalysts for the reverse water gas shift reaction (RWGS), where a KPD analysis reveals a swing mode RWGS operation, where CO2 is first converted into CO oxidizing the MXene surface, later regenerated by hydrogen treatment releasing water.

1 Morales-García et al. J. Mater. Chem. A 6 (2018) 3381. 2 Persson et al. Adv. Mater. 31 (2019) 1805472. 3 Morales-Salvador et al. ACS Catal. 11 (2021) 11248.

O 76.6 Thu 11:30 A 053

Effect of Surface Terminations on the Water Intercalation into Ti3C2Tz MXene Thin Films — Aditya Sharma[1], Thierry Ouisse[1], Ulf Wiedwald[2], Andrei Chumakov[3], Fabrice Wilhelm[4], and •Hanna Pazniak[1] — [1]Université Grenoble Alpes, CNRS, Grenoble INP, LMGP, Grenoble, France — [2]University of Duisburg-Essen and Center for Nanointegration Duisburg-Essen, Duisburg, Germany — [3]Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany — [4]European Synchrotron Radiation Facility, Grenoble, France

MXenes are 2D materials whose surface is terminated by functional groups that drastically affect the properties of MXenes, including their affinity for water. In this study, we synthesized Ti3C2 MXenes with 1) mixed (=O, -F, and -OH) and 2) halogen (-Cl) terminations and prepared thin films by drop casting. X-ray absorption near edge structure shows different local chemical environment of Ti depending on the terminations, which is also confirmed by density functional theory. Next, we expose the MXene thin films to various humidity levels and studied the water intercalation by wide angle X-ray scattering. We observed that the interplanar spacing of mixed-terminated MXenes varied from 11.74 Å to 16.97 Å with increasing relative humidity from 0% to 100%, respectively. In the Cl-terminated MXenes, however, the interplanar spacing remains much smaller and only changes from 11.0 Å to 12.8 Å as function of relative humidity. In both cases, the process of water intercalation is reversible.

Funded by ANR-23-CE09-0031-01 project.

O 76.7 Thu 11:45 A 053

Isotopic Analysis of Intercalated Protons in Ti$_3$C$_2$T$_x$ MXene for Electrochemical Energy Storage using in-situ FTIR Spectroscopy — •Andreas Weisser[1], Mailis Lounasvuori[1], Namrata Sharma[1], Kyle Matthews[2], Teng Zhang[2], Yury Gogotsi[2], and Tristan Petit[1] — [1]Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, 12489 Berlin, Germany — [2]A. J. Drexel Nanomaterials Institute and Department of Materials Science and Engineering, Drexel University, Philadelphia, PA 19104, USA

MXenes, a family of 2-dimensional transition metal carbides, are a promising candidate for use in energy storage applications due to their high capacitance. Here, we monitor the confined water and protons in Ti$_3$C$_2$T$_x$ MXene electrodes during cyclic voltammetry using operando FTIR spectroscopy. Isotopic exchange with deuterium oxide is used to allow for a better understanding of the proton dynamics inside the MXene interlayers. In-situ FTIR measurements are being performed with a dedicated electrochemical cell able to measure the different vibrational modes of the confined species. The measurements are conducted in attenuated total reflectance mode (ATR).

The focus of this presentation will be on analyzing the change of the vibrational modes of water and deuterium during electrochemical cycling versus the applied potential to the working electrode. This behaviour will be discussed with regard to the intercalation of protons and the reordering of the intercalated water or deuterium. The results will be placed in the context of MXene in the role of an electrode for pseudocapacitor applications.

O 76.8 Thu 12:00 A 053

Electronic Structure of V2CTx MXene in aqueous solutions studied using in situ Scanning Transmission X-Ray Microscopy. — •Namrata Sharma[1], Kyle Matthews[2], Andreas Weisser[1], Mailis Lounasvuori[1], Markus Weigand[1], Yury Gogotsi[2], and Tristan Petit[1] — [1]Helmholtz-Zentrum Berlin fur Materialien und Energie GmbH, 12489 Berlin, Germany — [2]A. J. Drexel Nanomaterials Institute and Department of Materials Science and Engineering Drexel University Philadelphia, 19104 PA, USA

MXenes, a class of 2D transition metal carbides and nitrides have attracted much attention in many applications, thanks to their layered structure, hydrophilicity, and surface terminations. Recently, a new synthesis protocol has significantly improved the quality and shelf life of V2CTx MXene. This study focuses on the behaviour of V2CTx MXene in aqueous solutions studied using In-situ Scanning Transmission X-Ray Microscopy (STXM) with dedicated electrochemical flow cell at the synchrotron BESSY II. STXM provides element-specific nanomaterial electronic structure characterization in liquid at ~50nm spatial resolution. Here we characterize the electronic structure of pristine and aged V2CTx MXene in air, water, ZnSO4, and H2SO4, using X-ray Absorption Spectroscopy (XAS) at V L-edge and O K edge. The stability of individual V2CTx MXene flakes is monitored through high resolution chemical imaging over several hours in aqueous electrolyte. In this work we stress upon on the relevance of chemical imaging allowed by STXM for insights into oxidation and hydrolysis of MXene in aqueous environment at the nanoscale.

O 77: Gerhard Ertl Young Investigator Award Competition

Time: Thursday 10:30–13:00 Location: HE 101

O 77.1 Thu 10:30 HE 101

Atomic-scale insights into frictional energy dissipation mechanisms during single-molecule manipulation — •Lukas Hörmann[1], Alfred J. Weymouth[2], and Reinhard J. Maurer[1] — [1]University of Warwick, Coventry, UK — [2]University of Regensburg, Regensburg, Germany

Friction causes significant energy loss in any moving mechanical device. As the miniaturisation of devices reaches the quantum limit, so do dynamical dissipation processes. Fundamentally quantum mechanical mechanisms govern friction at the nanoscale. We account for all relevant quantum mechanical effects, such as charge transfer, or van der Waals interactions, by employing density functional theory, and machine learning and use the probe particle model to simulate energy dissipation. Using this approach, we investigate dynamic friction at the natural limit of a singular atom moving a single chemical bond. This enables us to explore how the local bonding environment of the underlying sample surface affects the energy dissipation that a probe particle experiences. Focussing on the example of a CO-functionalised lateral force microscope that measures frictional energy dissipation above various organic adlayers on Cu(111), we find strong correlations between the local bonding environment and the energy dissipation. Our findings capture the qualitative trends found in experiment. Finally, we present a mechanistic interpretation of our findings that provides insights into the underlying physics of atomic/lateral force microscopy measurements.

O 77.2 Thu 11:00 HE 101

Analyzing structures of biomolecules at single molecule level by direct imaging on surface — •Kelvin Anggara — MPI for Solid State Research, Stuttgart

Diverse structures of biomolecules in living systems pose challenges for present analytical methods in analyzing their individual molecular structures. Ensemble-averaged measurement on molecules with extensive structural variation leads to loss of structural information for individual molecules. We herein bypass this problem by imaging and analyzing single biomolecules deposited intact on surfaces. Biomolecules were transferred to the gas-phase by electrospray, mass-selected, and soft-landed at a cold single-crystal metal surface, using the Electrospray Ion Beam Deposition (ESIBD) technique. The deposited molecules were subsequently imaged one-at-a-time by Scanning Tunneling Microscopy (STM) and interpreted by DFT calculations to reveal their individual structures on surface. We successfully applied our approach to glycans (1) as well as lipids and proteins densely decorated with glycans (2), whose analyses remain intractable by current analytical methods. Critical to our success is the choice of surfaces to deactivate diffusion and dissociation of biomolecules on surface, as well as the systematic molecular manipulation by the STM tip to unfold the intact biomolecule on surface. The ESIBD+STM+DFT approach opens a new avenue to access individual structures of any biomolecules that can be electrosprayed and deposited onto a surface. 1. Nature 582, 375-378 (2020). 2. Science 382, 219-223 (2023).

O 77.3 Thu 11:30 HE 101

Controlling the Yu-Shiba-Rusinov states of radical molecules on the superconductor Pb(111) — •Chao Li, Jung-Ching Liu, Outhmane Chahib, Thilo Glatzel, Remy Pawlak, and Ernst Meyer — Department of Physcis, University of Basel, Klingelbergstrasse 82, 4056 Basel, Switzerland

Radical molecules are considered to be a promising candidate for the future quantum computer. Here, we studied a radical molecule of 4,5,9,10-tetrabromo-1,3,6,8-tetraazapyrene (TBTAP) on a superconductor Pb(111). Low-temperature scanning tunneling microscope images reveal its rectangle shape on Pb(111). Moreover, scanning tunneling spectra show a Yu-Shiba-Rusinov (YSR) state inside the superconductor gaps on the molecules which was ascribed to one electron transferred from its environment. The evolution of YSR energy with tip-to-molecule distance shows a quantum phase transition between the singlet state and doublet state. Moreover, different constructions of the dimer and trimmer show typical coupling features among the molecules.

O 77.4 Thu 12:00 HE 101

Investigating many-body phenomena through molecular nanostructures — •Shantanu Mishra — IBM Research Europe - Zurich, 8803 Rüschlikon, Switzerland

This two-part contribution shows that individual organic molecules and molecular nanostructures on surfaces serve as an outstanding material platform to access many-body physics. In the first part, the tip-induced synthesis of an elusive organic molecule, namely indeno[1,2-a]fluorene, will be demonstrated on thin insulating NaCl films on (111) coinage metal surfaces [1]. The molecule exhibits ground state bistability, wherein it can be stabilized either in a (high-spin) open-shell state, or a (low-spin) closed-shell state, each of which shows characteristic experimental bond length alternation and frontier orbital densities that agrees with theory. Switching between open- and closed-shell states of an individual molecule is observed by changing its adsorption site on NaCl. In the second part, the on-surface synthesis of organic S = 1 antiferromagnetic quantum spin chains on Au(111) surface will be shown [2]. A systematic study of length-dependent magnetic excitations in both open-ended and cyclic spin chains reveal gapped spin excitations in the bulk, with the gap saturating for sufficiently long spin chains, and fractional S = 1/2 edge states at the chain termini, which manifest as Kondo resonances. It will be shown that these spectral features are direct evidence of emergent symmetry-protected topological order in the spin chains, ratifying Haldane's conjecture for integer-spin antiferromagnetic chains. [1] S. Mishra et al. arXiv:2303.04483 (2023). [2] S. Mishra et al. Nature 598, 287 (2021).

O 77.5 Thu 12:30 HE 101

Topology and chirality driven chemical reactions in multifold chiral semimetals — •Qun Yang and Claudia Felser — Max Planck Institute for Chemical Physics of Solids, Dresden 01187, Germany

The interplay between chirality and topology nurtures many exotic electronic properties. For instance, topological chiral semimetals display multifold band crossings with large Chern numbers in the bulk state and superlong topologically robust surface states without resistance and resistant to surface perturbations at the surface, which allows the entire crystal or surface to serve as catalytic active sites. These materials have the potential to revolutionize the design principle of high-performance catalysts for electrochemical energy conversion. In this talk, I will present our recent progress in developing highly efficient water-splitting electrocatalyst based on chiral multifold semimetals through theoretical and experimental studies. We found that these materials break the performance record of normal noble metal electrocatalysts. Through the theoretical band structure calculations incorporating the topological approach, we reveal the interaction mechanism between topological states and catalytic reactions. We found that the monopole-like orbital angular momentum texture driven by the chirality in the materials also offers great potential for exploring the asymmetric adsorption of chiral molecules, paving the way for understanding the asymmetric chemical reactions based on inorganic chiral materials.

O 78: Focus Session: Ultrafast Processes in Organic Semiconductors and Perovskites I (joint session O/CPP)

The fundamental processes determining the fate of excitons or charge carriers in organic semiconductors or perovskites are happening on an ultrafast timescale, i.e. 100 fs to 10 ps. This, for example, includes singlet-fission, excimer and polaron formation or hot carrier relaxation. Thus, for the development of efficient opto-electronic devices a deep understanding of the processes on this time scale is of utmost importance. Most importantly, theoretical and experimental methods must be combined to successfully decipher the studied phenomena as the many degrees of freedom in these soft matter materials often defy a straightforward explanation from the data. This Focus Session brings together theoretical and experimental scientists fostering an exchange of ideas on the various computational and experimental methods of the ultrafast sciences to the field of organic semiconductors and perovskites.

Organizers: Sebastian Hammer (U Montreal), Petra Tegeder (U Heidelberg)

Time: Thursday 10:30–13:00 Location: MA 004

Topical Talk O 78.1 Thu 10:30 MA 004

Transport and trapping in molecular materials at the picosecond time scale — •Marina Gerhard — Department of Physics and Material Sciences Center, Philipps-Universität Marburg, Germany

The Excitonic character of photoexcited states and the soft lattice of organic semiconductors pose guidelines for the design of devices, which are substantially different from those of their inorganic counterparts. To exploit the benefits of organic semiconductors, it is essential to comprehend the fate of photogenerated excitons at early times after their generation. In this context, molecular materials with a well-defined structure serve as ideal model systems to study processes such as exciton transport, excimer formation, singlet fission and energetic funneling. This contribution focuses on two recent studies, in which we explore the exciton dynamics in molecular systems using time and spatially resolved photoluminescence spectroscopy. For tetracene, the transport of excitons is driven by the interplay between singlet fission and triplet fusion. We demonstrate that this peculiar transport mechanism shows a pronounced temperature dependence and that it is severely hampered by the presence of trap states. The second study investigates heterostructures comprised of anthradithiophene with small concentrations of pentacene, which can efficiently mediate singlet fission through a favorable energy level arrangement of both compounds, demonstrating a system in which exciton funneling opens the possibility to spatially and spectrally separate light harvesting and singlet fission moieties.

O 78.2 Thu 11:00 MA 004

Fluctuations and exciton dynamics in molecular semiconductors — •Alexander Neef[1], Samuel Beaulieu[2], Sebastian Hammer[3], Shuo Dong[4], Tommaso Pincelli[1,5], Julian Maklar[1], R.Patrick Xian[6], Anke Krueger[7], Martin Wolf[1], Laurenz Rettig[1], Jens Pflaum[8] und Ralph Ernstorfer[1,5] — [1]Fritz-Haber-Institut — [2]Centre Lasers Intenses et Applications (CELIA) — [3]McGill University — [4]Chinese Academy of Sciences — [5]Technical University Berlin — [6]University of Toronto — [7]University of Stuttgart — [8]University of Wuerzburg

Time- and angle-resolved photoemission spectroscopy (trARPES) is a cutting-edge technique to study the electronic structure of materials out-of-equilibrium. Advances in instrumentation now allow taking multidimensional photoemission data sets of delocalized electrons in conventional semiconductors and the reconstruction of spatially confined molecular orbitals. I will present our recent results on trARPES studies of the fluctuation-dominated electronic structure of molecular semiconductors. Furthermore, I will talk about how trARPES made it possible to decipher a complex multiexcitonic process: singlet exciton fission [1]. Apart from the answers gained, a pressing question emerged: how can singlet exciton fission go uphill in energy in certain systems? Our work highlights the need for theories that are able to capture the behavior of excited states in a fluctuating landscape.

[1] Neef et al., Nature 616 (2023) https://www.nature.com/articles/s41586-023-05814-1

O 78.3 Thu 11:15 MA 004

Efficient Intramolecular Singlet Fission in Weakly-Interacting Heterodimers — •Oskar Kefer[1], Lukas Ahrens[2], Jie Han[3], Jan Freudenberg[2], Frank Rominger[2], Andreas Dreuw[3], Uwe H. F. Bunz[2], and Tiago Buckup[1] — [1]PCI, Universität Heidelberg, Heidelberg — [2]OCI, Universität Heidelberg, Heidelberg — [3]IWR, Universität Heidelberg, Heidelberg

Intramolecular singlet fission (iSF) converts one excited singlet (S_1S_0) into two triplets when two chromophores are covalently bound by a bridge. The bridge also determines the strength of intramolecular interaction. Usually, strong interaction leads to fast formation of the intermediate correlated triplet-pair ($^1[T_1T_1]$), but also hinders its dissociation. A Spiro-linker enforces a perpendicular π-system-arrangement around the central spiro-carbon, leading to long triplet-lifetimes that favor separation into $T_1 + T_1$ without compromising $^1[T_1T_1]$-formation efficiency.

We expand on this approach and demonstrate a new class of heterodimers that exploit the favorable properties of spiro-conjugation with enhanced iSF dynamics. Quantum-chemical calculations (DFT) and transient absorption spectroscopy link accelerated iSF-kinetics to changes in the S_1S_0- and $^1[T_1T_1]$-energy landscape. The limited interaction during the lifetime of $^1[T_1T_1]$ creates favorable conditions for triplet-separation, which are formed with a yield of up to 174%. This new approach furnishes efficient heterogeneous iSF materials with reduced intramolecular interaction due to spatial fixation between chromophores and large, tunable absorption bandwidths.

Topical Talk O 78.4 Thu 11:30 MA 004

Ultrafast charge-transfer dynamics in organic and hybrid interfaces from first principles — •Caterina Cocchi — Institut für Physik und CeNaD, Carl von Ossietzky Universität Oldenburg

Charge-transfer dynamics at interfaces in the sub-ps timescale is ruled by complex interplays of electronic and vibrational degrees of freedom. First-principle methods based on real-time time-dependent density functional theory in conjunction with Ehrenfest dynamics offer a practical tool to access and rationalize these phenomena in realistic systems. Considering different organic and hybrid interfaces [1-3], I will discuss the mechanisms ruling charge-transfer dynamics triggered by a resonant pulse. I will illustrate how the intrinsic characteristics of the involved compounds and their mutual interactions affect qualitatively and quantitatively the dynamics of charge carriers in the earliest stages of light-driven coherent excitations. I will highlight methodological aspects that are necessary to reproduce the physics involved [4-6] and point out open issues to be addressed by future developments.

[1] M. Jacobs et al., Adv. Phys. X 5, 1749883 (2020). [2] M. Jacobs et al., ACS Appl. Nano Mater. 5, 5187 (2022). [3] M. Jacobs et al., J. Phys. Chem. A 127, 8794 (2023). [4] J. Krumland et al., J. Chem. Phys. 153, 054106 (2020). [5] J. Krumland et al., Phys. Rev. B 106, 144304 (2022). [6] M. Jacobs et al., arXiv:2311.01776

O 78.5 Thu 12:00 MA 004

Charge separation in a porphyrin-based metal organic framework incorporating C_{60} fullerenes — •Martin Richter[1], Xiaojing Liu[2], Pavel Kolesnichenko[1], Christof Wöll[2], and Petra Tegeder[1] — [1]Physikalisch-Chemisches Institut, Universität Heidelberg, Im Neuenheimer Feld 253/229, 69120 Heidelberg, Germany — [2]Institut für Funktionelle Grenzflächen, Karlsruher Institut für Technologie, Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany

Porphyrin-based materials are attracting great interest due to their wide range of applications. They are often embedded into metal organic frameworks (MOF) to manipulate and investigate the relations between function and structure. Together with an electron acceptor charge separation can be achieved being beneficial for uses e.g. in solar cells or photocatalysis. Experiments on ZnTPP (Zinc 5,15-bis-(3,4,5-trimethoxyphenyl)-10,20-bis-(4-carboxyphenyl)) in a surface anchored MOF (SURMOF) loaded with fullerene C_{60} inside the pores have shown that photoexcitation increases the conductivity by up to two orders of magnitude.[1] Here, we investigate excited states dynamics after optical excitation with transient absorption spectroscopy on the femto- to nanosecond scale. Charge separated states can be identified, which have a lifetime of a few hundred picoseconds. The charge separation occurs after excitation of both the Soret band and the Q band.

1. X. Liu, Angew. Chem. Int. Ed. 2019, 58, 9590.

O 78.6 Thu 12:15 MA 004

Ultrafast charge separation and band structure dynamics in metal-phthalocyanine/WSe$_2$ heterostructures — •Gregor Zinke, Sebastian Hedwig, Benito Arnoldi, Martin Anstett, Martin Aeschlimann, and Benjamin Stadtmüller — University of Kaiserslautern-Landau, Germany

2D-Van-der-Waals materials and their heterostructures are a highly promising class of materials due to their low dimensional nature and diverse electronic properties, especially regarding their charge and spin carrier dynamics. In addition to chemical tunability, the spin-dependent electronic properties of such 2D heterostructures can also be altered on ultrafast timescales by the formation of transient charge-separated states. In this contribution, we focus on optically induced interlayer charge and spin transfer processes in heterostructures consisting of molecule films on the surface of a bulk WSe$_2$ crystal. Using spin-, time- and angle-resolved photoemission with XUV radiation, we can simultaneously monitor the excited carriers as well as the corresponding hole dynamics. For molecule films made of the metal phthalocyanines (MPcs) CuPc and FePc, we discuss the temporal evolution of optically excited spin and charge carriers in these MPc /WSe$_2$ heterostructures. We will illustrate the influence of the distinct orbital character of the central metal atom and structural order of the MPc molecules on the ultrafast electron and hole dynamics at the MPc/WSe$_2$ interface and discuss their impact on the transient changes in the interfacial energy level alignment.

O 78.7 Thu 12:30 MA 004

Disentangling the multiorbital contributions of excitons by photoemission exciton tomography — •G. S. Matthijs Jansen[1], Wiebke Bennecke[1], Andreas Windischbacher[2], Ralf Hemm[3], David Schmitt[1], Jan Philipp Bange[1], Christian Kern[2], Daniel Steil[1], Sabine Steil[1], Marcel Reutzel[1], Martin Aeschlimann[3], Peter Puschnig[2], Benjamin Stadtmüller[3,4], and Stefan Mathias[1] — [1]I. Physikalisches Institut, Georg-August-Universität Göttingen — [2]Institute of Physics, University of Graz — [3]Department of Physics and Research Center OPTIMAS, University of Kaiserslautern — [4]Institute of Physics, Johannes Gutenberg-University Mainz

The opto-electronic response of organic semiconductors is well-known to be dominated by excitons, i.e. quasiparticles that consist of bound electron-hole pairs. As excitons are realizations of a correlated many-particle wave function, experimental and theoretical methods must probe and characterize the the full electron-hole wavefunction, rather than just the single-particle orbitals. Recently, we have shown that time-resolved photoemission momentum microscopy can probe the entangled wavefunction and thereby unravel the exciton's multiorbital electron and hole contributions [1]. We demonstrate this for the prototypical organic semiconductor buckminsterfullerene (C_{60}) and achieve unprecedented access to key properties of the exciton state including localization, charge-transfer character, and ultrafast exciton formation and relaxation dynamics.

[1] W. Bennecke et al., arXiv preprint arXiv:2303.13904 (2023)

O 78.8 Thu 12:45 MA 004

Formation of charge-transfer excitons across the hybrid PTCDA/WSe$_2$ interface — •Wiebke Bennecke[1], David Schmitt[1], Jan Philipp Bange[1], Ignacio Gonzalez Oliva[2], Anna Seiler[1], Lukas Renn[1], Mattis Langendorf[1], Daniel Steil[1], Sabine Steil[1], R. Thomas Weitz[1], Peter Puschnig[3], Claudia Draxl[2], Marcel Reutzel[1], G. S. Matthijs Jansen[1], and Stefan Mathias[1] — [1]I. Physikalisches Institut, Georg-August-Universität Göttingen — [2]Institut für Physik, Humboldt-Universität zu Berlin — [3]Institute of Physics, University of Graz

Heterostructures consisting of van-der-Waals materials and organic molecules are a promising material platform due to their potential to combine the flexibility of organic chemistry with the remarkable optoelectronic properties of 2D materials. Here, we use femtosecond photoemission momentum microscopy to study ultrafast exciton dynamics in monolayer PTCDA adsorbed on WSe$_2$. By employing the concepts of photoemission orbital tomography, which has recently been shown to be capable of capturing key excitonic properties [1, 2], we are able to unambiguously identify the different excitonic states formed after optical excitation of WSe$_2$. We find that a localized long-lived charge-transfer exciton is formed on a picosecond timescale, where the hole resides in the WSe$_2$ and the electron is being transferred to the lowest unoccupied molecular orbital of PTCDA.

[1] C. Kern et al., Phys. Rev. B **108**, 085132 (2023)
[2] W. Bennecke et al., arXiv preprint arXiv:2303.13904 (2023)

O 79: 2D Materials V: Growth, Structure and Substrate Interaction

Time: Thursday 10:30–13:00 Location: MA 005

O 79.1 Thu 10:30 MA 005

Exfoliation of large-area monolayers using template stripped gold — •Laxman Nagireddy[1,2,3], Neil R. Wilson[1], Maria Christine Richter[2,3], Karol Hricovini[2,3], Matthew D. Watson[4], Cephise Cacho[4], Nicholas D. M. Hine[1], and Samuel J. Magorrian[1] — [1]University of Warwick, Coventry, UK — [2]CY Cergy Paris Université, CEA, LIDYL,Gif-sur-Yvette, France — [3]Université Paris-Saclay, CEA, LIDYL, Gif-sur-Yvette, France — [4]Diamond Light Source, Harwell Science and Innovation Campus, Didcot, UK

The diverse family of two-dimensional materials exhibits a myriad of applications owing to their unique electronic, optical, and chemical properties. While mechanical exfoliation has been a pivotal method for obtaining high-quality two-dimensional materials, existing techniques are constrained by limitations in yield, lateral size, and contamination. Although numerous studies have explored the use of gold for exfoliating large-area monolayers[1], these approaches often prove unsuitable for surface-sensitive techniques such as photoemission spectroscopy.

In this study, we present a novel methodology for preparing an atomically flat gold surface using a template stripped gold method [2], subsequently employed for the exfoliation of various two-dimensional materials. Successful exfoliation was achieved for WSe2, CrSBr, and FePS3. The obtained monolayers were characterized using angle-resolved photoemission spectroscopy (ARPES).

References:

[1] M Velicky, et al., ACSnano 12.10 10463-10472 (2018).

[2] N Vogel, et al., Nanoscale 4.13 3820-3832 (2012).

O 79.2 Thu 10:45 MA 005

Janus SeMoS monolayers grown by chemical vapor deposition on Au(111) — •Julian Picker[1], Mahdi Ghorbani-Asl[2], Maximilian Schaal[3], Silvan Kretschmer[2], Felix Otto[3], Marco Gruenewald[3], Christof Neumann[1], Antony George[1], Torsten Fritz[3], Arkady Krasheninnikov[2], and Andrey Turchanin[1] — [1]Institute of Physical Chemistry, Friedrich Schiller University Jena — [2]Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf — [3]Institute of Solid State Physics, Friedrich Schiller University Jena

Janus transition metal dichalcogenide monolayers (TMD MLs) are asymmetric two-dimensional (2D) crystals consisting of two different chalcogens on their opposite faces. Their unique structure results in the in-built electric dipole and leads to new physical phenomena compared to conventional TMD MLs such as MoS_2 or $MoSe_2$. Here we report an epitaxial growth of Janus SeMoS MLs on Au(111) by chemical vapor deposition. In the growth process, sulfur atoms intercalate between the $MoSe_2$ ML and Au(111) resulting in the exchange of the bottom selenium layer with sulfur [1]. We studied the structure of the formed Janus SeMoS MLs by scanning tunneling microscopy (STM) and low-energy electron diffraction (LEED). The electronic structure was characterized by angle-resolved photoelectron spectroscopy (ARPES). Density functional theory (DFT) calculations demonstrate a good agreement with the experimental findings.

[1] Z. Gan *et al.*, Adv. Mater. **34**, 2205226 (2022).

O 79.3 Thu 11:00 MA 005

Growth of Titanium Ditelluride on Au(111) — •Andreas Raabgrund, Hannah Loh, Alexander Wegerich, Tilman Kisslinger, Lutz Hammer, and M. Alexander Schneider — Universität Erlangen-Nürnberg, 91058 Erlangen, Germany

Aiming at the MBE growth of transition metal ditelluride (MTe_2) films, especially in the monolayer limit, the formation of and interaction with the substrate interface is of fundamental interest. The growth of a MTe_2 film can be achieved either by the tellurization of the desired metal substrate [1] or by the deposition of Te [2] or any other chalcogenide [3] and a desired transition metal on a substrate of suitable choice.

In this contribution we follow the latter approach and investigate tellurium and titanium on Au(111) by LEED-IV, DFT, and STM. Starting from submonolayer coverages of Ti and Te, a first stable, ordered $(3\sqrt{3} \times 3\sqrt{3})R30^\circ$ superstructure is found. With increasing amount of Ti and a proper annealing step a $(5 \times \sqrt{3})_{\mathrm{rect}}$ structure develops. For higher amounts of Te and Ti, a closed hexagonal film with a lateral lattice parameter of 4/3 times that of Au(111) is found by LEED and STM. STM topography and spectroscopy measurements indicate the growth of a $TiTe_2$ layer on a Te containing interface to the Au(111) substrate.

[1] T. Kißlinger et al., Phys. Rev. B **108**, 205412 (2023)

[2] K. Lasek et al., ACS Nano **14**, 8473 (2020)

[3] S. G. Sørensen et al., ACS Nano **8**, 6788 (2014)

O 79.4 Thu 11:15 MA 005

Influence of the substrate on pore creation in single-layer MoS_2 using highly charged ion irradiation — •Yossarian Liebsch[1], Leon Daniel[1], Lucia Skopinski[1], Carolin Frank[1], Umair Javed[2], Jani Kotakoski[2], and Marika Schleberger[2] — [1]University Duisburg-Essen, Duisburg, Germany — [2]Universität Wien,Vienna, Austria

Ion irradiation serves as a versatile tool for modifying 2D materials and surfaces, enabling the creation of defects, pores, and incisions. However, characterizing the nanometer-sized structural changes induced by ion irradiation has proven challenging, often necessitating high-resolution TEM. Notably, direct characterization of such defects has primarily focused on suspended 2D materials, neglecting the substrate's influence on defect formation during irradiation. To address this issue, we irradiated single-layer MoS_2 on a Si/SiO_2 substrate using highly charged ions (HCIs) at 20 keV and 180 keV and different charge states. Subsequently, the material was transferred to a TEM-grid and analyzed using scanning transmission electron microscopy (STEM). Well-defined, round pores were observed for all kinetic and potential energies, indicating a rather small influence of sputtered substrate material. In contrast to suspended MoS_2, significant differences in both pore size and creation efficiency were evident. These observations hold particular significance for 2D material applications that utilize ion irradiation as a mean to create pores and catalytically active sites, as they allow for a precise control of pore density and defective area by choosing appropriate irradiation parameters.

O 79.5 Thu 11:30 MA 005

Phase engineering in tantalum sulfide monolayers on Au(111) — Daniela Dombrowski[1,2], Abdus Samad[3], Kai Mehlich[1], Udo Schwingenschlögl[3], •Thais Chagas[1], and Carsten Busse[1,2] — [1]Department Physik, Universität Siegen, Germany — [2]Institut für Materialphysik, Westfälische Wilhelms-Universität Münster, Germany — [3]Physical Science and Engineering Division, King Abdullah University of Science and Technology (KAUST), Saudi Arabia

We prepare monolayers of tantalum sulfide on Au(111) by evaporation of Ta in a reactive background of H_2S. Under sulfur-rich conditions, monolayers of 2H-TaS_2 develop, whereas under sulfur-poor conditions TaS forms, a structure that can be derived from 2H-TaS_2 by removal of the bottom S layer. We analyze the alignment of the layers with respect to the substrate and the relation with the domains in the Au(111) herringbone reconstruction using scanning tunneling microscopy. With the help of density functional theory calculations, we can determine the registry of the two phases with the substrate. We develop a growth process that allows the preparation of uniquely oriented 2H-TaS_2 on Au(111). 2H-TaS_2 and TaS have a remarkably similar in-plane lattice structure and we observe the formation of lateral 2H-TaS_2-TaS heterostructures with atomically well-defined and defect-free boundaries. We observe mirror twin boundaries within 2H-TaS_2 along the S- and Ta-edge.

O 79.6 Thu 11:45 MA 005

Isolating the optical response of a MoS_2 monolayer under extreme screening of a metal — •Tao Yang[1], Stephan Sleziona[1], Erik Pollmann[1], Eckart Hasselbrink[2], Peter Kratzer[1], Marika Schleberger[1], R. Kramer Campen[1], and Yujin Tong[1] — [1]Fakultät für Physik, Universität of Duisburg-Essen, Germany — [2]Fakultät für Chemie, Universität of Duisburg-Essen, Germany

Transition metal dichalcogenides monolayers, as two-dimensional (2D) direct bandgap semiconductors, hold promise for advanced optoelectronic and photocatalytic devices. Interaction with three-dimensional (3D) metals, like Au, profoundly affects their optical properties, posing challenges in characterizing the monolayer's optical responses within the semiconductor-metal junction. In this study, using precise polarization-controlled final-state sum frequency generation (FS-SFG), we successfully isolated the optical responses of a MoS_2 monolayer from a MoS_2/Au junction. The resulting SFG spectra exhibit a linear lineshape without A or B exciton features, attributed to strong dielectric screening and substrate induced doping. The linear lineshape illustrates the expected constant density of states at the band edge of the 2D semiconductor, typically obscured by excitonic interactions in weak-screening conditions. Extrapolation yields the onset of a direct quasiparticle bandgap of about 1.65 ± 0.20 eV, indicating significant bandgap renormalization. This study enriches understanding of the optical responses of a 2D semiconductor in extreme screening conditions and provides a critical reference for 2D semiconductor-based photocatalytic applications.

O 79.7 Thu 12:00 MA 005

Direct Beam-Induced Chemical Etching of 2D Materials in Transmission Electron Microscope — •Mitisha Jain, Silvan Kretschmer, and Arkady Krasheninnikov — Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf, Bautzner Landstraße 400, 01328 Dresden, Germany

The interaction of energetic electrons with the specimen during imaging in a transmission electron microscope (TEM) can give rise to the formation of defects or even complete destruction of the sample. This is particularly relevant to atomically thin two-dimensional (2D) materials. Depending on electron energy and material type, different mechanisms such as knock-on (ballistic) damage, inelastic interactions including ionization and excitations, as well as beam-mediated chemical etching can govern defect production. Using first-principles calculations combined with the McKinley-Feshbach formalism, we investigate the latter channel of damage creation in two representative 2D materials, MoS_2 and h-BN with adsorbed single adatoms (H, C, N, O, etc.), which can originate from molecules always present in the TEM column. We assess the ballistic displacement threshold energies T for the host atoms in 2D materials when adatoms are present and demonstrate that T can be reduced, as chemical bonds are locally weakened due to the formation of new bonds with the adatom. We conclude that while adatoms should play a role in damage creation in MoS_2, formation of defects in h-BN sheets at electron energies well below the knock-on threshold cannot be explained by the presence of adatoms, that is by the direct electron beam-induced chemical etching.

O 79.8 Thu 12:15 MA 005

Optoelectronic Interactions of WS_2-ZnO for Scalable LEDs Based on Two-Dimensional Materials — •Osamah Kharsah[1], Leon Daniel[1], Denys Vidish[2], Dedi Sutarma[1], Jonah von Kuczkowski[1], Stephan Sleziona[1], Ulrich Hagemann[3], Kevin Musselman[2], and Marika Schleberger[1] — [1]Universität Duisburg-Essen, Fakultät für Physik and CENIDE, Duisburg, Germany — [2]University of Waterloo, Mechanical and Mechatronics Engineering, Waterloo,Canada — [3]Interdisciplinary Center for Analytics on the Nanoscale (ICAN) and CENIDE, Duisburg, Germany

Two-dimensional tungsten disulfide (WS_2) is a promising material for scalable large-area light-emitting diodes (LED) due to its direct bandgap, stability, and high photoluminescence (PL). Teamed with zinc oxide (ZnO), an electron-transport n-type semiconductor, they form the basis for an n-i-p LED architecture. This study explores the interactions between WS_2 and both single-crystalline ZnO and spatial atomic layer deposition (SALD)-grown ZnO. Emphasizing the optoelectronic interaction of this heterostructure, various characterization techniques such as PL and Raman spectroscopy, Atomic Force Microscopy, Kelvin Probe Force Microscopy, and X-ray Photoelectron Spectroscopy are employed. The results reveal a stronger optoelectronic interaction in WS_2-SALD ZnO, attributed to higher strain. Furthermore, we observe energy-level alignment at the interface in both cases, while noting the presence of OH groups in SALD ZnO but their absence in single-crystalline ZnO.

O 79.9 Thu 12:30 MA 005

Atomically thin transition metal halide $NiBr_2$ grown on Au(111) — •Daniel Rothhardt[1,2,3], Zuned Ahmend[1,2], Christopher Penschke[4], Hao Liu[1,2], Regina Hoffmann-Vogel[3], Peter Saalfrank[4], Hans Josef Hug[1,2], and Amina Kimouche[3] — [1]Swiss Federal Laboratories for Materials Science and Technology, CH-8600 Dübendorf, Switzerland — [2]Department of Physics, University of Basel, CH-4056 Basel, Switzerland — [3]Institute of Physics and Astronomy, University of Potsdam, 14476 Potsdam-Golm, Germany — [4]Institute of Chemistry, University of Potsdam, 14476 Potsdam-Golm, Germany

In recent years, two-dimensional (2D) van der Waals materials have emerged as a new platform for studying low-dimensional quantum phenomena. Due to their limited symmetry, they display electrical, optical, and magnetic properties that are different compared to their 3D counterparts. As novel 2D materials, transition metal dihalides (TMHs) emerge as promising platform for stable ferro- or antiferromagnetic ordering in two dimensions. Epitaxial single-layer TMHs require accurate control of the growth parameters and an improved understanding of the interactions between TMHs and the surface. In this work, we employ frequency-modulated scanning force microscopy in the non-contact mode, combined with Kelvin probe microscopy and scanning tunneling microscopy, to examine various phases and layer thicknesses of $NiBr_2$ on Au(111) revealing a complicated growth mode in the low coverage regime.

O 79.10 Thu 12:45 MA 005

Atomically precise vacancy lattices in epitaxially grown $FeBr_2$ and $CoBr_2$ on Au(111) — Feifei Xiang[1], Neeta Bisht[2], Binbin Da[1], Christian Neiss[2], Andreas Görling[2], and •Sabine Maier[1] — [1]Department of Physics, Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany — [2]Department of Chemistry and Pharmacy, Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany

The generation of extensive 2D periodic patterns of point defects in 2D materials, such as vacancy lattices, has been a challenging task until now. We report on the growth and structure of epitaxially grown 2D transition metal dihalides on Au(111) featuring periodically assembled halogen vacancies that result in alternating coordination of the transition metal ion and can function as antidot lattices.[1] Using low-temperature STM/ncAFM and LEED, we identified the structural properties of intrinsically patterned $FeBr_2$ and $CoBr_2$ monolayers grown epitaxially on Au(111). Density-functional theory indicates that Br-vacancies are favored due to low formation energies, and the formation of a vacancy lattice substantially reduces the lattice mismatch with the underlying Au(111). We demonstrate that interfacial strain engineering presents a versatile strategy for controlled patterning in 2D with atomic precision over several hundred nanometers to solve a longstanding challenge of growing atomically precise antidot lattices.
[1] F. Xiang, et al. arXiv preprint arXiv:2305.06489, 2023.

O 80: Scanning Probe Microscopy: Light Matter Interaction at Atomic Scales II

Time: Thursday 10:30–12:30 Location: MA 041

O 80.1 Thu 10:30 MA 041

All-optical subcycle microscopy with atomic resolution — •Valentin Bergbauer, Thomas Siday, Johannes Hayes, Felix Schiegl, Fabian Sandner, Peter Menden, Martin Zizlsperger, Svenja Nerreter, Sonja Lingl, Jascha Repp, Jan Wilhelm, Markus A. Huber, Yaroslav A. Gerasimenko, and Rupert Huber — Department of Physics and Regensburg Center for Ultrafast Nanoscopy (RUN), University of Regensburg, 93040 Regensburg, Germany

Near-field microscopy has prompted a revolution in nano-videography of quantum systems on subcycle timescales and with a spatial resolution down to ∼10 nm. Yet, the nanoscale geometry of the tip apex has prevented access to atomic resolution. We present a fundamentally new approach which brings all-optical microscopy to the atomic scale while retaining subcycle temporal resolution for the first time, by exploiting extreme nonlinearities within tip-confined evanescent light fields. We demonstrate the capabilities of this Near-field Optical Tunnelling Emission (NOTE) microscope by imaging nanometre-sized packing defects on the surface of gold, alongside tracing the subcycle quantum flow of electrons between the scanning tip and a semiconducting van der Waals trilayer in real-time. NOTE microscopy is not only compatible with insulating samples, where no rectified currents can flow but also enables us to combine all-optical subcycle spectroscopy with atomic resolution. In doing so, NOTE provides direct access to atomic scale quantum light-matter interaction and dynamics on their natural length and timescales.

O 80.2 Thu 10:45 MA 041

Excitation mechanism for STM-induced luminescence — •Song Jiang[1], Tomáš Neuman[2], Rémi Bretel[2], Alex Boeglin[1], Fabrice Scheurer[1], Eric Le Moal[2], and Guillaume Schull[1] — [1]Université de Strasbourg, CNRS, IPCMS, UMR 7504, F-67000 Strasbourg, France — [2]Institut des Sciences Moléculaires d'Orsay (ISMO), UMR 8214, CNRS, Université Paris-Saclay, 91405 Orsay Cedex, France

Fluorescence of neutral and charged molecules has been reported with subnanometer resolution in scanning tunneling microscope induced-luminescence (STML) experiments over the last years. Despite these significant developments, the detailed mechanisms of STML of individual molecules remained to be fully understood. Here, we proposed a "universal" model relying on successive carrier tunneling events that is based on a systematic experimental STML study of quinacridone (QA) molecules adsorbed on four-monolayer (4 ML) NaCl/Ag(111). Based on a comparison between the spatial, bias voltage, and tunneling current dependences of the charged and neutral emission, and on conductance data, a many-body description of the system is set. It reveals that four different charged states of QA (QA^-, QA^0, QA^+, and QA^{2+}), involving three different spin multiplicities, can be populated within a single voltage sweep. This excitation mechanism is backed up by data obtained for QA deposited on NaCl/Au(111) where the ground state of the molecule is a doubly positively charged singlet state (S^{2+}). This model bears a universal character that can be applied to STML experiments dealing with charged and neutral molecules.

O 80.3 Thu 11:00 MA 041

Structure and optical characterisation of Dibenzoterrylene on Au(111) and C_{60} — •Andreas Reutter[1,2], Yannis Hilgers[1,2], Franziska Hirt[2,3], Stefan Kück[2,3], Markus Etzkorn[1,2], and Uta Schlickum[1,2] — [1]Institut für Angewandte Physik, TU Braunschweig — [2]Laboratory for Emerging Nanometrology, TU Braunschweig — [3]Physikalisch-Technische Bundesanstalt, Braunschweig

Polycyclic aromatic hydrocarbons are promising candidates for molecule based single photon sources since they offer high brightness and narrow spectral emission lines. One interesting candidate is dibenzoterrylene (DBT, $C_{38}H_{20}$) which has recently been investigated in anthracene nanocrystals. However, anthracene tends to sublime at room temperature and the fabrication process has a low reproducibility. We have grown DBT on Au(111) and in C_{60} buffer layers in vacuum with organic molecular beam epitaxy. Using confocal laser scanning microscopy, we find photon emission from DBT clusters when embedded in a C_{60} Matrix. In addition, we have investigated the structure with low-temperature STM. For sub-monolayer coverage, DBT molecules self-assembly on the well-known Herringbone reconstruction on the Au(111) surface. Surprisingly, the DBT molecules prefer the fcc over the hcp areas of the herringbones in contrast to other molecules. We plan to fabricate DBT-C_{60} sandwich samples with individual DBT molecules implemented in the interlayer and to study their optical properties regarding line width and single photon characteristic of this system.

O 80.4 Thu 11:15 MA 041

Directional emission of plasmonic light by atomic-scale defects — •David Mateos[1,2], Óscar Jover[2], Miguel Varea[1,2], Koen Lauwaet[1], Daniel Granados[1], Rodolfo Miranda[1,2], Antonio Isaac Fernández Domínguez[3], Alberto Martín Jiménez[1], and Roberto Otero[1,2] — [1]IMDEA Nanoscience, Madrid, Spain — [2]Departamento de Física de la Materia Condensada and Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid, Madrid, Spain — [3]Departamento de Física Teórica de la Materia Condensada and Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid, Madrid, Spain

Manipulating the directionality of subwavelength light sources is demanded in broad applications, including sensing, photonics, optoelectronics, and energy harvesting. In this respect, nanoscale objects can control the angular distribution of scattered light below the diffraction limit. It has been demonstrated that properly engineering the size, shape, and arrangement of nanostructures in the few-nanometer scale affords control of the emission properties. Still, the effect of atomistic structures remains unexplored. In this work, we show that atomic-scale objects can also modify the directionality of plasmonic emission. The radiative density of optical states of the nanocavity formed by a sharp metallic tip at tunneling distance to a metallic surface is locally modified by a monoatomic height step. Comparison with electromagnetic calculations demonstrates that the observed changes arise from light emission tilting of the picocavity plasmons. Thus, atomic-like structures influence the light emission properties of nanoscale objects.

O 80.5 Thu 11:30 MA 041

Electroluminescence of VOPc in different adsorption configurations — Vibhuti Rai[1,2], •Lukas Gerhard[2], Christof Holzer[3], Carsten Rockstuhl[3,4], and Wulf Wulfhekel[2] — [1]Fachbereich Physik, Freie Universität Berlin, 14195 Berlin, Germany — [2]Institute for Quantum Materials and Technologies, Karlsruhe Institute of Technology (KIT), 76131 Karlsruhe, Germany — [3]Institute of Theoretical Solid State Physics, Karlsruhe Institute of Technology (KIT), 76131 Karlsruhe, Germany — [4]Institute for Nanotechnology, Karlsruhe Institute of Technology (KIT), 76131 Karlsruhe, Germany

In the last few years, scanning tunneling microscopy (STM) induced luminescence has mostly focused on planar molecules absorbed in a flat configuration. The main component of the electric field in the STM junction, however, is oriented perpendicular to the substrate. Here we show electroluminescence of Vanadyl-phthalocyanine (VOPc) deposited on NaCl on Au(111) which exhibits a strong dipole component perpendicular to the surface and different adsorption configurations [1]. As the structural change of VOPc is related to the spin structure [2], this offers a possibility to explain the observed differences in the light emission pattern in terms of charge state, lifetime and efficiency.

[1] S. Debnath et al, Angewandte Chemie 2022, 61 (25). [2] K. Kaiser et al, ACS Nano 2019, 13 (6).

O 80.6 Thu 11:45 MA 041

Electron and phonon driven mobility of the ammonia clusters adsorbed on a copper surface — •Prashant Srivastava[1], Daniel Miller[2], and Karina Morgenstern[1] — [1]Chair of physical chemistry I, Ruhr University of Bochum, Germany — [2]Department of Chemistry, Hofstra University, United States2Department of Chemistry, Hofstra University, United States

Understanding molecular kinetics induced by interfacial energy transfer at metal-molecule interfaces is important in molecular electronics and battery research. We explore the ultrashort laser pulse-induced mobility of ammonia clusters adsorbed on a copper surface at a single-molecule level by a combination of low-temperature scanning tunneling microscopy with an adjoined femtosecond laser and density functional theory. The diffusion of single molecules and desorption of molecules from clusters above an area of 1.35 nm2 is observed at laser fluences below 0.390 mJ/cm2. At higher laser fluences, single molecules desorb. In this talk, we present fluence-dependent changes in the ammonia coverage to understand the excitation mechanism. We discuss how the ammonia molecules and clusters are excited by electrons and phonons in a fluence range from 0.117 mJ/cm2 to 0.625 mJ/cm2. The study contributes to a control of molecular kinetics via interfacial energy transfer.

O 80.7 Thu 12:00 MA 041

TDDFT study of the orthosilicic acid molecule under electrostatic field and laser irradiation — •Tommaso Morresi — ECT* - FBK, Trento, Italy

The Tomographic Atom Probe (TAP) is a rapidly developing technique in the field of condensed matter analysis. Recently, a combination of TAP with intense THz pulses to control ion emission has been proposed [1]. This mechanism enables high spatial resolution and chemical sensitivity. In this configuration, the TAP analysis could be extended to bio-samples as an alternative to 4D microscopy. This investigation is the ultimate goal of the EIC Pathfinder project MIMOSA. Within this collaborative framework with a strong experimental endeavour, the theoretical understanding and computational modelling of the atom evaporation mechanism is of paramount importance. In this work, we study the interaction of THz-lasers with the orthosilicic acid molecule, which is the main ingredient of the matrix embedding the bio-material in the experimental setup. By using time-dependent density functional theory for electrons in combination with molecular dynamics for ions, we analyse the dynamics of laser-assisted field evaporation of OH ions from the molecule. While the scheme is similar to the one proposed in Ref. [2], the novelty of this work is mainly represented by the frequency of the laser in TDDFT simulations. We show the critical fields allowing the evaporation of OH ions in the THz regime and also the effect of additional external electrostatic fields on top of the time-dependent ones. [1] - A. Vella et al., Sci. Adv. 7, 7 (2021) [2] - E.P. Silaeva et al., PRB 92, 155401 (2015)

O 80.8 Thu 12:15 MA 041

Selectively Addressing Plasmonic Modes and Excitonic States in a Nanocavity Hosting a Quantum Emitter — •Alberto Martin-Jimenez[1], Oscar Jover[2], Koen Lauwaet[1], Daniel Granados[1], Rodolfo Miranda[1,2], and Roberto Otero[1,2] — [1]IMDEA Nanoscience, Madrid, Spain — [2]Dep. De Física de la Materia Condensada & IFIMAC, Universidad Autónoma de Madrid, Madrid, Spain

Controlling the interaction between the excitonic states of a quantum emitter and the plasmonic modes of a nanocavity is key for the development of quantum information processing devices. In this contribution we demonstrate that the tunnel electroluminescence of electrically insulated C60 nanocrystals enclosed in the plasmonic nanocavity at the junction of a scanning tunneling microscope can be switched from a broad emission spectrum, revealing the plasmonic modes of the cavity, to a narrow band emission, displaying only the excitonic states of the C60 molecules by changing the bias voltage applied to the junction. Interestingly, excitonic emission dominates the spectra in the high-voltage region in which the simultaneously acquired inelastic rate is low, demonstrating that the excitons cannot be created by an inelastic tunnel process. These results point toward new possible mechanisms for tunnel electroluminescence of quantum emitters and offer new avenues to develop electrically tunable nanoscale light sources.

O 81: Nanostructures at Surfaces I

Time: Thursday 10:30–12:45 Location: MA 042

O 81.1 Thu 10:30 MA 042

One-dimensional electronic structure of phosphorene chains — •Maxim Krivenkov[1], Maryam Sajedi[1], Dmitry Marchenko[1], Evangelos Golias[2], Oliver Rader[1], and Maxim Krivenkov[1] — [1]Helmholtz-Zentrum Berlin für Materialien und Energie, Elektronenspeicherring BESSY II, Albert-Einstein-Str. 15, 12489 Berlin, Germany — [2]MAX IV Laboratory, Lund University, Fotongatan 2, 22484, Lund, Sweden

Phosphorene, a 2D allotrope of phosphorus, is very appealing to electronic technology due to its semiconducting properties with narrow band gap. Further reduction of dimensionality could give rise to exotic properties of its electronic structure. Using angle-resolved photoemission we studied P atomic wires self-assembled on Ag(111) substrate. Dispersion of the P band measured along and perpendicular to the wires, reveals pronounced electronic confinement in a 1D band, dispersionless in perpendicular direction to the wire. Our density functional calculations, apart from precise reproduction of the 1D band for the P/Ag(111), predict a 1D to 2D metallic transition in the electronic structure upon increasing density of the nanowire array.

O 81.2 Thu 10:45 MA 042

Perturbations in quantum corrals — •Fabian Stilp, Marco Weiss, and Franz J. Giessibl — Institute of Experimental and Applied Physics, University of Regensburg, 93040 Regensburg, Germany

The surface state of Cu (111), a quasi-2-dimensional electron gas, is trapped to a small surface area of about 15 x 15 nm^2 by placing 48 CO-molecules in a circular shape on the surface via atomic manipulation. By doing so, one creates a quantum corral with discrete electronic states forming an artificial atom. This structure can be described reasonably well by an infinitely high circular potential well leading to corral states with Bessel-type radial functions and an angular momentum normal to the surface. To investigate the influence of single atom or molecule perturbations on quantum corrals we bring Fe atoms and CO molecules inside the corral and measure the response of the corral states.

Thanks to the large corral diameter, one can study the structure of the wave functions within that artificial atom by AFM and STM showing an angular dependence of the corral states after placing the perturbations inside the corral. This change of the wave functions leads to an energy shift of a few meV. By investigating the change of the corral states due to the perturbation, one can draw conclusion about the interaction between this artificial atom and a natural atom. Here we expand the interpretation of the adatom acting repulsively on the corral states as stated by Stilp et al. [1].

[1] F. Stilp, A. Bereczuk, J. Berwanger, N. Mundigl, K. Richter, F.J. Giessibl, Science **372**, 1196-1200 (2021).

O 81.3 Thu 11:00 MA 042

Intermediate metal-insulator phases of individual VO$_2$ nanocrystals for multilevel memory — Peter Kepič, Michal Horák, Jiří Kabát, Filip Ligmajer, Andrea Konečná, and •Vlastimil Křápek — Brno University of Technology, Czechia

Vanadium dioxide (VO$_2$) is a strongly correlated material that exhibits metal-insulator transition (MIT) around 340 K [1]. A broad conductivity hysteresis of VO$_2$ is vital for its applications as memory or memristor devices. In our contribution, we study the hysteresis of temperature-induced MIT in individual high-density VO$_2$ nanocrystals using analytical electron microscopy. We utilize low-loss and core-loss electron energy loss spectroscopy (EELS) combined with in-situ heating to analyze the metal-insulator transition [2] in more than 40 nanocrystals. We retrieve the parameters of the hysteresis loop and demonstrate their dependence on the size of the nanocrystals. Interestingly, some nanocrystals exhibit switching from the insulating to the metallic phase by parts, with the intermediate phases allowing to design multilevel memory. We also show by correlating the image contrast with EELS that signatures of MIT are observable simply and efficiently using annular dark-field imaging.

[1] P. Kepič *et al.*, ACS Photonics **8**, 1048 (2021).

[2] J. Krpenský, M. Horák *et al.*, arXiv 2309.11980.

O 81.4 Thu 11:15 MA 042

Nanoflower-like VSx@NC as a promising anode for stable potassium-ion storage — •Vincent Hartmann, Yulian Dong, Huaping Zhao, and Yong Lei — Fachgebiet Angewandte Nanophysik, Institut für Physik & IMN MacroNano, Technische Universität Ilmenau, Germany

Potassium-ion batteries (PIBs) are promising candidates for large-scale energy storage due to their low cost and abundant potassium content in the earth's crust. However, owing to the large radius of K+ ions, the electrode material structure is easily damaged during the potassiation/depotassiation process, hindering their further applications. VSx has become a new PIB anode material due to its low cost, high theoretical capacity, and unique structure, which can provide sites for potassium storage. Here, constructing a flower-like VSx covered by nitrogen-doped carbon as an anode electrode for PIBs can effectively enhance electrical and mechanical properties and provide abundant active sites for pseudocapacitive behavior to achieve fast kinetics. As a result, it exhibits excellent cycling stability with a special capacity of 249.3 mAh g-1 at 100 mA g-1 after 1000 cycles with an initial discharge capacity of 293.8 mAh g-1. The strategy in this work provides inspiration for the rational design of advanced nanostructured electrode materials to develop PIB anodes with long cycle life.

O 81.5 Thu 11:30 MA 042

Investigation of highly efficient black titania nanotube photocatalyst by soft-X-ray spectroscopy — •Hesham Ali Fahmy Abdalla Hamad — City of Scientific Research and Technological Applications (SRTA-City), Alexandria, Egypt

The recent discovery of black TiO2 nanoparticles with enhancing solar absorption, especially in the visible and near-infrared region will trigger an explosion of interest in the application of TiO2 in a diverse set of solar energy systems; but black TiO2 nanoparticles really remain a mystery. Here we elucidate more properties and try to understand the inner workings of black TiO2 nanotubes with hydrogenated disorders in a surface layer surrounding a crystalline core. In this work, TiO2 nanotubes (TNT) and its black hydrogenated TiO2 nanotubes (BTNT) were synthesized and investigated by characterization techniques. The merit of this project is to investigate the prepared synchrotron-based X-ray absorption fine structures (XAFS). It probed the unoccupied and occupied molecular orbitals of densities of states for O 2p and Ti 3d hybrid orbital characteristics, respectively. The reduction behavior and electronic and crystalline structure of a series of TNT and BTNT as photocatalysts will be investigated by using synchrotron-based XAFS. These techniques provide novel opportunities for tackling the structure and the dynamics of chemical and physical systems in solution.

O 81.6 Thu 11:45 MA 042

Diffusion studies on Pb islands and on the wetting layer in Pb/Si(111)-(7x7) — •Paul Philip Schmidt, Felix Hartmann, Lea Faber, and Regina Hoffmann-Vogel — University of Potsdam, Institute of Physics and Astronomy, Germany

Metallic structures on semiconductors offer a wide range of technical applications. This presentation will focus on the diffusion behavior of Pb/Si(111)-(7x7). It is known that Pb initially forms a wetting layer on Si, from which islands then form. Previous research has shown that this system exhibits explosive island growth and abnormally fast diffusion [1][2]. In our study, we focus on the wetting layer. After fabricating the 7x7 reconstruction on Si(111) under ultra-high vacuum conditions, we evaporate between 2 and 7 monolayers Pb at substrate temperatures between 120 and 300K. The diffusion was studied using non-contact cantilever scanning force microcopy (NC-SFM) and simultaneous Kelvin probe force microscopy (KPFM). While the topographical data of the NC-SFM essentially confirmed known data on the growth of the islands, the KPFM showed changes in the local contact potential difference (LCPD). In one experiment, a local imbalance was additionally generated on an island by manipulating a Pb island using the SFM-tip. Despite the low time resolution of an SFM measurement, it was possible in this way to determine the time scales of the mass transport. [1] M. Hupalo et. al. Phys. Rev. B, 23 (2007) [2] K. L. Man et al. Phys. Rev. Lett., 101 (2008)

Topical Talk O 81.7 Thu 12:00 MA 042

Trapping single atoms of noble gases in nanocages: from fundamental studies to applications — •Jorge Anibal Boscoboinik — Center for Functional Nanomaterials, Brookhaven National Laboratory, Upton, NY, USA

Confinement effects can result in exciting properties in the chemistry and physics of small molecules. In this talk, we will explore the trapping of noble gases in the confined space inside silicate nanocages and at the interface between these cages and their metal support. Understanding the fundamental aspects of these confinement effects can lead to designing new materials for specific applications. Based on the lessons from the fundamental studies for noble gas trapping in 2D-model systems, we are developing new nanoarchitectures that are potentially scalable and can be used for various applications related to producing and detecting noble gases.

O 81.8 Thu 12:30 MA 042

Automating measurements on the nanoscale: Artificial Intelligence versus classical analysis of SPM data — •Tim J. Seifert[1], Ziba Akbarian[1,2], Birka Lalkens[2], Ingo Busch[3], Harald Bosse[3], and Uta Schlickum[1,2] — [1]Institut für Angewandte Physik, Technische Universität Braunschweig — [2]Laboratory for Emerging Nanometrology LENA, Braunschweig — [3]Physikalisch-Technische Bundesanstalt, Braunschweig

The continuous trend in research and technology towards structures on the nanometer scale drives the growing interest in imaging mechanisms using Scanning Probe Microscopes (SPM). As the experimental methods continue to evolve, the increasing output of data requires fast, reliable and accurate analysis methods avoiding the need for an experienced user. The automatization of image analysis procedures for SPM mainly consists of well-established routines using classical methods requiring laborious manual work. Artificial Intelligence (AI) based analysis techniques have recently attracted great interest hoping to provide true autonomous imaging and analysis procedures. While the accuracy of classical methods is often limited by noise, AI can overcome these challenges, albeit with the additional need for high amounts of labeled training data. Here we present a framework to analyze SPM data and extract distance information using AI-based methods trained on synthetic data, as well as classical alternatives, highlighting the benefits of each approach. The procedure is applied to measure a novel DNA-Origami based Single-Molecule length reference providing a fast, cheap and accurate way to calibrate SPMs on the nanoscale.

O 82: Electronic Structure Theory I

Time: Thursday 10:30–12:30 Location: MA 043

O 82.1 Thu 10:30 MA 043

Black-box, accurate, and efficient prediction of band structures with Koopmans functionals — •Edward Linscott[1], Nicola Colonna[1], Junfeng Qiao[2], and Nicola Marzari[1,2] — [1]Paul Scherrer Institut, Villigen, Switzerland — [2]École Polytechnique Fédérale de Lausanne, Lausanne, Switzerland

Over the past fifteen years we have developed Koopmans functionals, a computationally efficient approach for predicting spectral properties in a functional framework.[1] These orbital-density-dependent functionals impose a generalized piecewise linearity condition that ensures that orbital energies match the corresponding electron removal/addition total energy differences (in contrast to semi-local DFT, where a mismatch between the two lies at the heart of the band gap problem and the unreliability of Kohn-Sham band structures more generally). Koopmans functionals prove to be very powerful, yielding band structures and molecular orbital energies with comparable accuracy to self-consistent GW approaches but at greatly reduced computational cost and complexity.[2] This talk will cover the theory of Koopmans functionals and how recent developments – namely, the development of automated workflows via the `koopmans` code[3] and projectability-based Wannierization with manifold remixing[4] – have brought us one step closer to black-box prediction of accurate band structures.
[1] Dabo et al., PRB 82 (2010), Borghi et al., PRB 90 (2014)
[2] Nguyen et al., PRX 8 (2018), Colonna et al., JCTC 15 (2019)
[3] koopmans-functionals.org, Linscott et al., JCTC 19 (2023)
[4] Qiao et al., npj Comput. Mater. 9, 206 & 208 (2023)

O 82.2 Thu 10:45 MA 043

Efficient but accurate approximation to the exact exchange in hybrid density functionals — •Sebastian Kokott[1,2], Volker Blum[3], and Matthias Scheffler[1] — [1]The NOMAD Laboratory at the FHI of the Max Planck Society and IRIS-Adlershof of the Humboldt Universität zu Berlin, Germany — [2]Molecular Simulations from First Principles e.V., Berlin, Germany — [3]Thomas Lord Department of Mechanical Engineering and Materials Science, Duke University, Durham, NC, USA

We introduce a computationally efficient screening for the Coulomb potential of the exact exchange terms in hybrid density functionals. The proposed approach approximates the long-range exact exchange contributions and noticeably speeds up its numerical evaluation. The method is implemented in the all-electron electronic structure code FHI-aims. We test and demonstrate the approach for prototypical semiconductors and organic crystals using the PBE0 functional. The results show that the accuracy is affected only little, but computational costs for dense materials are reduced significantly. Further, we explore how this methods enables large-scale hybrid DFT simulations.

O 82.3 Thu 11:00 MA 043

Surrogate models for the electron density and related scalar fields — Joseph Abbott[1], •Raymond Chong[1], Alan Lewis[2], and Michele Ceriotti[1] — [1]Laboratory of Computational Science and Modeling (COSMO), IMX, École Polytechnique Fédérale de Lausanne, Switzerland — [2]University of York, United Kingdom

The electron density is a central quantity in electronic structure calculations and a fundamental property of molecules and materials, allowing access to in principle any ground state electronic property. Density-functional theory (DFT) is an ab initio method that calculates the electron density of a system by solving self- consistently the Kohn-Sham equations. However, the cubic scaling in system size makes calculations for large systems intractable. As a complementary approach, machine learning (ML) surrogate models are being developed that directly predict the self-consistent electron density at a fraction of the cost of DFT, and with more favourable linear scaling. One framework in particular focuses on learning the coefficients of basis functions fitted to the real-space density using a local equivariant ML model.

An end-to-end pipeline for the training and prediction of the the electron density is presented here. Built on top of a modular and scalable software stack, the 'bypassing' of the Kohn-Sham equations to access the density of larger systems becomes increasingly possible. Furthermore, the framework can be applied to scalar field related to the electron density, opening the door to interesting applications such as the ML-driven imaging of scanning tunneling microscopy (STM).

O 82.4 Thu 11:15 MA 043

Conventional definitions of the absolute energy reference can lead to suboptimal machine learning performance for the electronic density of states — •Wei Bin How, Sanggyu Chong, Federico Grasselli, Kevin Kazuki Huguenin-Dumittan, and Michele Ceriotti — Ecole Polytechnique Fédérale de Lausanne, Lausanne, Switzerland

The electronic density of states (eDOS) provides a simple and clear picture of the distribution of energy states for electrons, granting key insights regarding the behaviour of electrons in the material. Currently, the most popular way to obtain the eDOS is through DFT calculations, but the cubic scaling behaviour of DFT has motivated a push to apply machine learning to obtain the eDOS at a much lower cost. However, when constructing the dataset for the machine learning problem, one has to choose an energy reference for the dataset. Current conventions define the energy reference of the eDOS of bulk systems using either the average Hartree potential in the cell or the Fermi level of the entire system. As the absolute value of these quantities are typically not well defined for bulk systems, eDOSes of different bulks have to be interpreted individually. However, machine learning methods typically treat these energy references as absolute which may hinder prediction performance. In this talk, we explore two different ways to provide an optimal absolute energy reference for machine learning and showcase the significant improvement in performance over conventional definitions.

O 82.5 Thu 11:30 MA 043

Prediction of the Single Particle Electronic Hamiltonian for Periodic Systems — •Hanna Türk, Divya Suman, Jigyasa Nigam, and Michele Ceriotti — EPFL, Lausanne, Switzerland

The electronic structure of a material provides essential information on a materials properties. For most materials, it can directly be computed by ab initio calculations. However, for complex systems such as material grain boundaries and interfaces, which often govern relevant chemical processes, the required large simulation cells exceed the feasiblity of such methods.

Here, we take a first step to develop a neural network based model to predict the effective single particle electronic Hamiltonian for periodic systems from given atomic environments. This apporach has proven successful for molecules[1], and we now adapt it to bulk materials. By expanding the framework to learn the realspace Hamiltonian, which includes all relevant periodic translations, sampling of the entire k-phase becomes possible. Our versatile framework yields an accurate description of the relevant electronic structure, which can be used to obtain band structures and electronic conductivities. In perspective, the prediction on local environments allows training of a model on small cells, and the trained model can then be used to obtain electronic structure information on larger, more complex systems.

[1] J. Nigam, M. J. Willatt, M. Ceriotti, J. Chem. Phys. 2022, 156, 014115, DOI 10.1063/5.0072784.

O 82.6 Thu 11:45 MA 043

Electronic excited states from physically-constrained machine learning — Edoardo Cignoni[1], •Divya Suman[2], Jigyasa Nigam[2], Lorenzo Cupellini[1], Benedetta Mennucci[1], and Michele Ceriotti[2] — [1]Dipartimento di Chimica e Chimica Industriale, Università di Pisa, Pisa, Italy — [2]Laboratory of Computational Science and Modeling (COSMO), IMX, École Polytechnique Fédérale de Lausanne, Switzerland

The integration of machine learning (ML) techniques with quantum mechanical (QM) calculations has opened new avenues in predicting a variety of electronic properties of molecules. Our work investigates a fundamental question, which is of great relevance to drive these developments: should machine learning (ML) be directly employed to predict desired properties or be synergistically combined with physically-grounded operations? To this end we introduce an integrated modelling approach in which we build a symmetry-adapted ML model that targets the properties of interest while learning the minimal-basis, single-particle electronic Hamiltonian as an intermediate. This approach also enables predictions of properties other than the ones used during the training process, like the molecular excited states. The resulting architecture, therefore, inherits the accuracy of

QM calculations, as well as the transferability to larger, more complex molecules as well as a variety of ground and excited states properties, while being orders of magnitude faster compared to the traditional electronic structure methods.

O 82.7 Thu 12:00 MA 043

Algorithmic inversion on sum over poles to embed interacting many-body systems — •Alessandro Carbone[1], Tommaso Chiarotti[1], Massimo Capone[2], and Nicola Marzari[1,3] — [1]École Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland — [2]International School for Advanced Studies (SISSA), via Bonomea 265, 34136 Trieste, Italy — [3]Paul Scherrer Institut, 5232 Villigen, PSI, Switzerland

Quantum embedding methods are powerful techniques to study interacting correlated electrons beyond mean-field theories. An established approach is dynamical mean-field theory, which tackles the problem by mapping the strongly correlated electrons into an Anderson impurity model. Here, we explore a different avenue to solve Dyson-like equations at $T = 0K$ — the algorithmic inversion on sum over poles — providing an embedding formulation on the real axis. We first discuss the analytical derivation of the method and compare it with an exact diagonalization of the Anderson model. We then test this solver on a one-dimensional Hubbard ring in a self-consistent framework, and conclude discussing applications to general many-body Hamiltonians and other quantum embedding strategies.

O 82.8 Thu 12:15 MA 043

Total energies from the Klein and the Luttinger-Ward functionals: analytical benchmarking of conserving many-body approximations — •Giovanna Lani[1] and Nicola Marzari[1,2] — [1]École Polytechnique Fédérale de Lausanne, Lausanne — [2]Laboratory for Materials Simulations, Paul Scherrer Institute, Villigen, Switzerland

We investigate analytically the performance of two many-body energy functionals, derived respectively by Klein and Luttinger and Ward, at different levels of diagrammatic approximations (2B, GW and T-matrix), for the calculation of total energies. We benchmark our results on the extended two-site Hubbard model, which is analytically solvable and for which several exact properties can be calculated. Despite its simplicity, this model is prototypical of the H_2 dissociation, a notoriously difficult problem to tackle for most mean-field based approaches. We show that both functionals exhibit good to excellent variational properties, particularly in the case of the Luttinger-Ward one, which is in close agreement with fully self-consistent calculations, and we elucidate the relation between the accuracy of the results and the different input one-body Green's functions. Provided that such input is wisely chosen, we demonstrate how the Luttinger-Ward functional can be used as a computationally less expensive alternative to fully self-consistent many-body calculations, without sacrificing the precision of the results. In virtue of this accuracy, we argue that this functional can also be used to rank many-body approximations at different regimes of electronic correlation, thus bypassing the need for self-consistency.

O 83: Focus Session: Novel Approaches to Surface Plasmon Generated Charges and Heat for Photocatalysis II (joint session O/CPP)

Time: Thursday 10:30–13:15 Location: MA 141

Topical Talk O 83.1 Thu 10:30 MA 141

Designing Plasmonic Photocatalysts — •Emiliano Cortes — Nanoinstitute Munich, Faculty of Physics, University of Munich (LMU), Munich, Germany

Nanostructured surfaces and colloids with designed optical functionalities - such as plasmonic & photonic nano and metamaterials - allow efficient harvesting of light at the nanoscale. Exploiting light-driven matter excitations in these artificial materials opens up a new dimension in the conversion and management of energy at the nanoscale [1-4]. One example are plasmonic and photonic catalysts, that can indeed confine solar energy into molecular regions [4-11]. In this talk, I will present our recent efforts in order to understand the light-matter interaction in plasmonic nanoparticles and metamaterials for photocatalysis [1-11].

References [1] E. Cortés, Nature 614, 230-232 (2023) [2] E. Cortés, et al. Nature Rev. Chem., 6, 259-274 (2022) [3] E. Cortés, et al. Chem. Rev. 122, 15082-76 (2022) [4] S. Ezendam, et al. ACS Energy Letters, 7, 778-815 (2022) [5] M. Herran, et al. Adv. Funct. Mat., 2203418 (2022) [6] J. Gargiulo, et al. Nature Communications 14, 3813 (2023) [7] L. Nan, et al. Nano Letters 23 (7), 2883-2889 (2023) [8] A. Stefancu, et al. ACS Nano 17 (3), 3119-3127 (2023) [9] L. Hüttenhofer, et al., Adv. Energy Mat., 46, 2102877 (2021) [10] M. Herran, et al. Nature Catalysis, accepted (2023) [11] S. Ezendam, et al. ACS Nano, accepted (2023)

O 83.2 Thu 11:00 MA 141

Plasmon assisted Catalytic Conversion of CO2 with Disordered Cu-Pd Network — •Oliver Wipf[1], Jelena Wohlwend[1], David Kiwic[2], Markus Niederberger[2], Ralph Spolenak[1], and Henning Galinski[1] — [1]Laboratory for Nanometallurgy, ETH Zürich, Switzerland — [2]Laboratory for Multifunctional Materials, ETH Zürich, Switzerland

The catalytic conversion of carbon dioxide is currently at the forefront of research as a way to mitigate the climate crisis by capturing CO2 and turning it into functional chemical components. However, currently this is a high energy consumption process, thus, developing efficient catalysts is critical. Using solar radiation to drive and catalyze chemical reactions could address this challenge. Here, we explore the plasmon assisted catalytic conversion of CO2 with disordered network metamaterials (DNMs). DNMs are an emerging class of metamaterials with tunable quasi-perfect absorption over a broad range of wavelengths. The optical response of such disordered networks can be conceptualized as coupled dipole-like networks, where the disorder of the metallic network traps and localizes surface plasmon (SP) waves. In this work we harness DNMs as a platform to drive the conversion of CO2 by taking advantage of "hot" carriers, i.e. electron-hole (e-h) pairs, resulting from the non-radiative decay of plasmons. The generation of hot carriers is dependent on the chemistry and the local geometry, both tailorable with our fabrication route. Furthermore, we show that through chemical engineering of the DNMs, selectivity of the reaction products can be achieved.

O 83.3 Thu 11:15 MA 141

Advancing Plasmonic Charge Transfer for Photocatalysis: Bottom-Up Strategies in Energy Harvesting and Optoelectronics — •Swagato Sarkar[1] and Tobias A. F. Koenig[1,2] — [1]Leibniz-Institut für Polymerforschung Dresden e.V., Hohe Straße 6, 01069 Dresden — [2]Center for Advancing Electronics Dresden (cfaed), Technische Universität Dresden, Helmholtzstraße 18, 01069 Dresden

Our study comprehensively explores bottom-up techniques for plasmonic charge transfer, focusing on their collective applications in energy harvesting. Extending the concepts of plasmon-photon hybridization to self-assembled plasmonic nanoparticle chains or deposited metallic nanobars introduces a scalable approach for charge injection to adjacent semiconductor thin films, thus facilitating cost-effective photodetection [Sarkar et al., Adv. Func. Mat., 2021, 2011099]. The fusion of soft-lithography with template-assisted self-assembly further enhances large-scale bottom-up strategies, providing a cost-efficient printing of plasmonic metasurfaces for superior charge transfer and amplified the photocatalytic processes [Sarkar et al., Adv. Func. Mat., 2021, 2105054]. Complementing these studies, introducing a novel interconnected metal-semiconductor grating design showcases enhanced current modulation, allowing for efficient photosensor development by leveraging plasmon-mediated hot electrons [Sarkar et al., Adv. Func. Mat.,2023, 2210172]. Thus, our unified approach to bottom-up techniques in plasmonic charge transfer advances the field and promises efficient and scalable solutions for miniaturized optoelectronic devices.

O 83.4 Thu 11:30 MA 141

Electrochemistry at the surface of electrically connected plasmonic resonators — •Paul Mörk[1], Amro Sweedan[2], Muhammad Y. Bashouti[2], Bert Hecht[1], and Thorsten Feichtner[1] —

[1]Nano-Optics & Biophotonics Group, Experimental Physics 5, University of Würzburg, Am Hubland, 97074 Würzburg, Germany — [2]Ilse-Katz Institute for Nanoscale Science & Technology, Ben-Gurion University of Negrev, POB 653, Beer-Sheba Campus, Building 51, 8410501, Israel

Electrochemical reactions can be enhanced by catalytic processes at gold surfaces. Nanostructuring can lead to plasmonic resonances in the visible wavelength which may be used for photon routing, local heating or hot electron generation. This way it is possible to tailor the properties of gold for specific photo-electrochemical processes.

Here we discuss the use of electrically connected double-wire gratings as an effective plasmonic platform [1]. The gratings are fabricated from monocrystalline gold platelets via focused ion beam milling. We observe the electrochemical oxidation and reduction of these gold double-wire gratings at ambient conditions when applying voltages up to 10 V over gaps of 20 nm in width. We further show the dimerization of 4-aminothiophenol under continuous irradiation and the reverse reaction in presence of an applied voltage. Finally, we show a first implementation of the gratings as electrodes within nanofluidic channels for future applications as electrochemical cell.

[1] Sweedan, Amro, et al., arXiv preprint arXiv:2308.01395 (2023).

O 83.5 Thu 11:45 MA 141

Atomic scale plasmonic catalysis — •Yicui Kang and Emiliano Cortés — Nanoinstitute Munich, LMU Muenchen, Germany

In the realm of plasmonic catalytic systems, much attention has been devoted to the plasmon-derived mechanisms1, yet the influence of nanoparticles (NPs) crystal facets has been sparsely investigated. In this work2, we study the plasmon-assisted electrocatalytic CO2RR using three different shapes of plasmonic Au nanocube (NC), rhombic dodecahedron (RD) and octahedron (OC) - with different exposed facets: {100}, {110} and {111}, respectively. Upon plasmon excitation, Au OCs and NCs exhibited nearly a doubling in the FE(CO) compared to non-illuminated response. In contrast, RDs showed nearly the same performance in dark or light conditions. Electromagnetic modeling showed higher electric field (EF) enhancement on OCs and NCs on the edges and corners, which in some degree contributes to the enhancement of CO production3. Large-scale atomistic simulations of the electronic structure revealed higher hot carrier abundance on OCs and NCs compared to RDs. Abundant hot carriers on edges facilitate molecular activation, leading to enhanced selectivity and activity. This observation is further supported by plasmon-assisted HER experiments. Our findings highlight the dominance of low coordinated sites over facets in plasmonic catalytic processes, providing valuable insights for designing more efficient catalysts for energy conversion and carbon neutralization. Ref1. ACS Energy Letters 7.2 (2022): 778-815 Ref2. Crystal Facet Effect in Plasmonic Catalysis. Revision (2023) Ref3. Angew.Chem.Int.Ed.61.44 (2022): e202212640

O 83.6 Thu 12:00 MA 141

Translucent aerogel supports for photocatalysis — •David Kiwic, Fabian Matter, and Markus Niederberger — Laboratory for Multifunctional Materials, Department of Materials, ETH Zürich, Vladimir-Prelog-Weg 5, 8093 Zürich, Switzerland

Efficient light absorption is a key part of designing a photocatalytic process. In catalysts light can be scattered or absorbed before reaching the intended absorption site. Metal oxide aerogel supports minimize these light transport losses and their open porous structure enables sufficient gas transport to the active sites. These photocatalysts can be fabricated by synthesizing selected metal oxide nanoparticles, cogelling them with plasmonic nanoparticles, shaping them into granules via extrusion and subsequent super-critical drying. Pd on ZrO2 is known in the field of conventional thermal catalysis to be highly active for CO2 reduction, a key reaction for the decarbonization of our economy. Additionally, Pd nanoparticles strongly absorb visible light and shall be investigated for their photocatalytic activity. Aerogel granules were produced and catalytically tested in thermal and photothermal catalysis with a custom-built reactor setup that enables simultaneous heating and illumination of the granules in the fixed bed. The space time yield of the main product CO was increased manifold by illumination with two white LEDs, demonstrating the great potential of translucent aerogels as support materials for photocatalysis.

O 83.7 Thu 12:15 MA 141

Plasmonically generated low-energy electrons: the decomposition of DNA nucleobases derivatives study case — •Sergio Kogikoski Junior[1], Janka Kopyra[2], Janusz Rak[3], and Ilko Bald[1] — [1]Institute of Chemistry, University of Potsdam, Germany — [2]Faculty of Sciences, Siedlce University of Natural Sciences and Humanities, Poland — [3]Faculty of Chemistry, University of Gdansk, Poland

One outcome of the excitation of plasmonic nanoparticles is the generation of low-energy charge carriers (E < 0.5 eV). Our group showed that such charge carriers could drive chemical reactions at the interface of Au and Ag nanoparticles using different probe molecules, such as bromoadenine. However, it is still unclear how such molecule activation happens and what the energy of these carriers is. Herein, we used modified DNA nucleobases as probes to understand the role of the support metal and the low-energy electrons generated. The activation mechanisms to drive reactions using low-energy electrons are already well-studied in the gas phase of such molecules, providing a suitable theoretical background. SERS was used to obtain the molecular fingerprint of the system during irradiation. Our results show that irradiated AgNPs can decompose uracil and adenine derivatives, possibly related to the higher amount and energy of the generated hot electrons. Also, adenines are readily decomposed, even though studies in the gas phase show that both molecules decompose. The results show an energy landscape of plasmonically generated hot charges and its relation to probe molecule structure.

O 83.8 Thu 12:30 MA 141

Light-driven hydrogen evolution at copper surfaces: machine-learning accelerated nonadiabatic dynamics light the way — •Alexander Spears, Wojciech Stark, and Reinhard J. Maurer — University of Warwick, Coventry, United Kingdom

Light-induced ultrafast dynamics entail the energy transfer between light, electrons, and phonons at interfaces. Whether or not this energy transfer can be harnessed to selectively drive photocatalytic processes remains an open question. Machine-learning interatomic potentials based on electronic structure theory enable the efficient statistical sampling of chemical dynamics at metal surfaces. Molecular dynamics with electronic friction (MDEF) provide a fully anharmonic mixed quantum-classical description of electron-phonon coupling, whereby electronic friction is usually approximated with a free electron gas model. This method has previously been used to model state-resolved scattering experiments or in the calculation of vibrational lifetimes after femtosecond laser pulses, though its validity has yet to be fully assessed. Herein, we present ML-enabled MDEF simulations of thermal and light-driven hydrogen recombination and desorption probabilities from various copper surface facets. We compare results based on electronic friction based on first-order response theory and the free electron gas model and analyse the ability of different approximations to accurately describe the non-adiabatic energy transfer between the surface and adsorbates. Our approach also allows us to study the role of quantum nuclear effects on the dynamics of light-driven associative desorption.

O 83.9 Thu 12:45 MA 141

Strong Light-Matter Interaction to Control Plasmonic Catalysis — •Christian Schäfer, Jakub Fojt, and Paul Erhart — Department of Physics, Chalmers University of Technology, 412 96 Göteborg, Sweden

Optically excited nanoplasmonic particles are efficient catalysts, for which an intricate interplay of internal carrier generation and heat drives the reactions of molecules in close proximity. Here, we establish a connection between optical strong coupling between resonator structures and the nanoplasmonic plus molecule system to non-intrusively control the optical excitation energy. Based on a theoretical description from first principles (1,2,3), we demonstrate that this non-intrusive control can be used to optimize carrier injection into the molecule and thus improves plasmonic catalysis. We discuss current limitations, future development, and the prospects of strong coupling modified plasmonic catalysis.

[1] C. Schäfer and G. Johansson, PRL 128, 156402 (2022). [2] C. Schäfer, J. Phys. Chem. Lett. 2022, 13, 6905-6911. [3] C. Schäfer, J. Fojt, P. Erhart, in progress (2024).

O 83.10 Thu 13:00 MA 141

Anisotropic core-shell particles in the strong coupling regime — •Mathis Noell and Carsten Henkel — Universität Potsdam, Institut für Physik und Astronomie, Germany

Plasmonic particles coated with resonant organic absorbers (e.g., J-aggregates) provide a versatile platform to observe strong coupling.

Calculations using Mie theory result, however, in the prediction of a "spurious" unobserved absorption peak between the upper and lower polariton [1]. The spurious resonance was previously thought to be caused by a negative value of the shell permittivity [2]. Alternatively, recent literature has shown that power broadening of the exciton transition efficiently suppresses the spurious resonance [3]. We demonstrate, using a modified Tavis-Cummings model, that the assumption of an isotropic shell material leads to the excitation of plasmonic dark modes (PDM) by an external field and, consequently, to an absorption line located between the upper and lower polariton. Anisotropic transition dipoles with a fixed orientation relative to the particle surface can fully suppress the coupling between the PDM and the illumination field.

[1] T. Uwada, R. Toyota, H. Masuhara, and T. Asahi, J. Phys. Chem. C 111 (2007) 1549.

[2] T. J. Antosiewicz, S. P. Apell, and T. Shegai, ACS Photonics 1 (2014) 454.

[3] F. Stete, W. Koopman, G. Kewes, C. Henkel, O. Benson, and M. Bargheer ACS Photonics 10 (2023) 2511.

O 84: Electronic Structure of Surfaces I: Spectroscopy, Surface States

Time: Thursday 10:30–13:00 Location: MA 144

O 84.1 Thu 10:30 MA 144

A new beamline at the ASTRID2 synchrotron for spatially resolved ARPES — •Alfred Jones, Paulina Majchrzak, Zhihao Jiang, Klara Volckaert, Deepnarayan Biswas, Chakradhar Sahoo, Marco Bianchi, Nykola Jones, Søren Hoffmann, Philip Hofmann, Jill Miwa, and Søren Ulstrup — Aarhus University, Denmark

Increased scientific interest in 2D materials, heterostructures, and operational devices that incorporate quantum materials necessitates measurement techniques capable to isolating clear signals from such small and complex systems. Here, we present our new spatially resolved ARPES beamline at the ASTRID2 synchrotron, SGM4, where a <4 μm beam spot is produced using an elliptical capillary optic. We demonstrate the capability of this beamline using nanoARPES measurements on several example systems, such as in-operando ARPES from a 2D device.

O 84.2 Thu 10:45 MA 144

Recent progress in ToF-XPEEM and Momentum Microscopy: Theory and Experiment — •O. Tkach[1,2], Q. Nguyen[3], O. Fedchenko[1], S. Chernov[4], D. Kutnyakhov[4], F. Pressacco[4], J. Dilling[4,5], L. Bruckmeier[4,5], F. Scholz[4], M. Scholz[4], M. Hoesch[4], K. Rossnagel[4,5], H.-J. Elmers[1], and G. Schönhense[1] — [1]Univ., Mainz — [2]SumDU, Ukraine — [3]SLAC Nat. Accel. Lab., USA — [4]DESY, Hamburg — [5]CAU Kiel

Cathode-lens instruments (PEEM, LEEM, momentum microscopes MM) usually have a strong accelerating electric field in front of the sample. Here we present an alternative route towards good performance in k-imaging mode. It is based on *field shaping* in the region close to the sample by planar concentric ring electrodes. Ray-tracing reveals: (i) An accelerating lens in front of the sample reduces spherical aberration and field curvature, enabling large k-fields of view. (ii) A retarding lens can tune the field to zero, enabling studies of nonplanar objects in zero field. (iii) Stronger retarding fields direct all slow electrons back to the surface, reducing space-charge effects in pump-probe experiments. Simulations are confirmed by first experiments at PETRA-III and FLASH.

O 84.3 Thu 11:00 MA 144

Control of the asymmetric band structure in Mn$_2$Au by a ferromagnetic driver layer — •Y. Lytvynenko[1,2], O. Fedchenko[1], S. Chernov[1,3], S. Babenkov[1], D. Vasilyev[1], O. Tkach[1], A. Gloskovskii[3], T. R. F. Peixoto[3], C. Schlueter[3], V. Grigorev[1], M. Filianina[1,4], S. Sobolev[1], A. Kleibert[5], M. Kläui[1], J. Demsar[1], G. Schönhense[1], M. Jourdan[1], and H.-J. Elmers[1] — [1]JGU Mainz, Germany — [2]Institute of Magnetism of the NAS and MES of Ukraine — [3]DESY, Germany — [4]Stockholm University, Sweden — [5]SLS, PSI, Switzerland

In this work, the hard X-ray angle-resolved photoemission spectroscopy (HARPES) reveals the momentum-resolved band structure in an epitaxial Mn$_2$Au(001) film capped by a 2 nm thick ferromagnetic Permalloy (Py) layer. By magnetizing the Py layer, the exceptionally strong exchange bias aligns the Néel vector (NV) in the Mn$_2$Au film leading to changes in the electronic properties. Uncompensated Mn interfacial magnetic moments in Mn$_2$Au are identified as the origin of the exceptional exchange bias using XMCD in combination with PEEM. Using time-of-flight momentum microscopy, we measure the asymmetry of the band structure in Mn$_2$Au resulting from the homogeneous orientation of the NV. Comparison with theory shows that the NV, determined by the magnetic moment of the top Mn layer, is oriented antiparallel to the Py magnetization. Our experimental results demonstrate that HARPES can measure the band structure of epitaxial layers beneath a metallic capping layer. We thus confirm that the ferromagnetic capping layer controls the bulk band structure of the AFM film.

O 84.4 Thu 11:15 MA 144

Surface atomic and electronic structure of multiferroic GeTe(111) — •Martin Heinrich[1,2], Juraj Krempasky[1], Gunther Springholz[3], and Matthias Muntwiler[1,2] — [1]Paul Scherrer Institute, Photon Science Division, 5232 Villigen PSI, Switzerland — [2]Swiss Nanoscience Institute, University of Basel, 4056 Basel, Switzerland — [3]Institute of Semiconductor Physics, Johannes Kepler University Linz, 4040 Linz, Austria

Multiferroic materials combine multiple coupled ferroic orders in the same system which could be used to bridge the gap between traditional electronics and spintronics. GeTe is a IV-VI semiconductor with multiple existing applications in optoelectronics and thermoelectrics. It recently gained renewed attention due to the discovery of a large Rashba spin splitting in both bulk and surface states that couples to the ferroelectric polarization. A detailed study of its surface atomic and electronic structure is therefore of high relevance towards a deeper, atomic scale understanding of the relevant interactions.

In this contribution, we discuss synchrotron based photoelectron diffraction (XPD, PhD) data combined with multiple scattering calculations, which indicate a contracted surface layer structure of GeTe(111) compared to its bulk structure. Additionally, scanning tunneling microscopy (STM) measurements reveal triangular island growth and a hexagonal atomic surface structure. dI/dV spectroscopy probes the local density of states, which we compare to spatially averaged photoelectron spectra. We identify the Rashba split surface states and resolve their spatial distribution.

O 84.5 Thu 11:30 MA 144

Spin Texture in Honeycomb Monolayers AgTe/Ag(111) and CuTe/Cu(111) — •Begmuhammet Geldiyev[1], Maximilian Ünzelmann[1], Philipp Kagerer[1], Jakub Schusser[1], Hendrik Bentmann[2], and Friedrich Reinert[1] — [1]Experimentelle Physik 7 and Würzburg-Dresden Cluster of Excellence ct.qmat, Universität Würzburg — [2]Center for Quantum Spintronics, Department of Physics, NTNU, Norway

In this contribution we will investigate the spin texture of honeycomb monolayers CuTe and AgTe [1] grown on Cu(111) and Ag(111), respectively. In case of both materials, our spin-resolved ARPES measurements suggest the existence of Rashba-like spin texture, which originates from the broken inversion symmetry at the surface. Moreover, we here for the first time observe the Rashba-like spin splitting in CuTe/Cu(111). Prior, it could not be seen in spin-integrated ARPES and we deduce a value of 0.25 ± 0.05 eV Å for the Rashba constant. It is approximately a factor of 4 smaller compared to the case of AgTe, almost equal to the relative factor between Rashba constants of Cu$_2$Bi and Ag$_2$Bi alloys [2]. In addition, we elaborate on the formation of out-of-plane spin polarization which can be induced by the underlying honeycomb lattice structure.

[1] M. Ünzelmann et al., Phys. Rev. Lett. 124, 176401 (2020)
[2] H. Bentmann et al., EPL 87, 37003 (2009)

O 84.6 Thu 11:45 MA 144

Exploring the topologically dark surface of a layered weak

3D topological insulator — •Johannes Hessdörfer[1,2], Maximilian Ünzelmann[1,2], Eduardo Carillo-Aravena[2,3], Armando Consiglio[2,4], Michael Ruck[2,3], Domenico Di Sante[5], and Friedrich Reinert[1,2] — [1]Experimentelle Physik VII, Universität Würzburg, Germany — [2]Würzburg-Dresden Cluster of Excellence ct.qmat, Germany — [3]Anorganische Chemie II, Technische Universität Dresden, Germany — [4]Theoretische Physik I, Universität Würzburg, Germany — [5]University of Bologna, Italy

Weak three-dimensional (3D) topological insulators (TI) can be considered as a stack of 2D TIs separated by insulating spacer layers. In the family of $Bi_{14}Rh_3I_9$ based [1] weak TIs, the electronic properties can be modified by altering the spacer layer through substitution. Here, we investigate the compound $Bi_{12}Rh_3Ag_6I_9$, by means of photoemission experiments and density functional theory band structure calculations. The results indicate the presence of surface states within the projected bulk band gap of the topologically dark surface which might alter the topological properties there. These states can be assigned to originate from the two different types of layer terminations. By analyzing the I 4d peak in the X-ray core level spectra, we show how the surface termination can be controlled, e.g. by the cleaving temperature.

[1]Rasche et al., Nat. Mater, 12, 422-425 (2013)

O 84.7 Thu 12:00 MA 144

Novel electronic structures from anomalous stackings in transition metal dichalcogenides — •Mihir Date[1,2], Alex Louat[1], Niels Schroeter[2], and Matthew D. Watson[1] — [1]Diamond Light Source, Diamond Light Source, Harwell Science and Innovation Campus, Didcot, United Kingdom — [2]Max Planck Institute of Microstructure Physics, Halle (Saale), Germany

Advances in photoemission spectroscopy, especially in terms of spatial resolution, have opened a plethora of possibilities in probing variations in the local electronic structure, which are elusive in traditional angle -resolved photoemission spectroscopy (ARPES) experiments. In our experiments, we have shown that in some ”2H”-bulk transition metal dichalcogenides (TMDCs), MoS2, NbS2 and TaS2, minority regions on the sample show spectral signatures of quantum well states. Furthermore, we observe subtle differences in the spectral weight and band splitting in the hole-like pockets around M- and K-points in anomalously stacked MoS2, compared to the bandstructures of its 2H- and 3R-phases. We speculate such electronic structures are derived from local lattice imperfections, where the periodicity of the 2H stacking is broken along the c-axis. We propose that these stacking faults offer a convenient plane for sample cleaving and therefore, are easily captured in a surface sensitive technique like ARPES. Our work not only presents novel electronic structures of traditional TMDCs, but also highlights the strength and importance of spatially resolved ARPES measurements.

O 84.8 Thu 12:15 MA 144

Utilizing matrix element effects to study the orbital texture of Dirac surface state in $PtTe_2$ — •Muthu P. T. Masilamani[1], Jakub Schusser[1], Mohammed Qahosh[2], Lukasz Plucinski[2], Begmuhammet Geldiyev[1], and Friedrich Reinert[1] — [1]Experimentelle Physik VII and Würzburg-Dresden Cluster of Excellence ct.qmat, Universität Würzburg, Würzburg, Germany — [2]Peter Grünberg Institut (PGI-6), Forschungszentrum Jülich GmbH, Jülich, Germany

In this work, we study the matrix element effect of the ARPES intensity to deduce k-space initial state orbital texture featuring different symmetries in type-II Dirac semimetal $PtTe_2$. Our spin- and angle-resolved photoemission data were augmented by the one-step model of the photoemission within the spin-polarized relativistic Korringa-Kohn-Rostoker (SPR-KKR) Green’s function method of the Munich band structure software package. In order to extract information about the different contributions to the resulting spectral weight and spin-polarization, the matrix element used in our one-step model of photoemission calculations includes all experimental parameters such as photon energy, light polarization and geometry configurations. Via such control over the experimental parameters, in order to investigate the orbital wavefunction above and below the Dirac point, we performed polarization-dependent ARPES calculations where we varied the angle of the crystal mirror plane with respect to the experimental mirror plane.

O 84.9 Thu 12:30 MA 144

Observation of Chiral Surface State in Superconductor NbGe — •Mengyu Yao[1], Martin Gutierrez-Amigo[2,3], Subhajit Roychowdhury[1], and Claudia Felser[1] — [1]Max Planck Institute for Chemical Physics of Solids, Dresden, Germany — [2]Departamento de Física, Facultad de Ciencia y Tecnología, Universidad del País Vasco (UPV/EHU), Apartado 644, 48080 Bilbao, Spain — [3]Centro de Física de Materiales (CSIC-UPV/EHU), Manuel de Lardizabal pasealekua 5, 20018 Donostia/San Sebastián, Spain

The interplay between topology and superconductivity in quantum materials harbors rich physics ripe for discovery. In this work, we investigate the topological properties and superconductivity in the nonsymmorphic chiral superconductor NbGe2 using high-resolution angle-resolved photoemis- sion spectroscopy (ARPES), transport measurements, and ab initio calculations. Our ARPES data reveals exotic chiral surface states on the (100) surface, stemming from the inherent chiral crystal structure. Supporting calculations indicate NbGe2 likely hosts elusive Weyl fermions in the bulk electronic structure. Furthermore, we uncover signatures of van Hove singularities that could engen- der enhanced many-body interactions. Additionally, transport measurements demonstrate NbGe2 exhibits superconductivity below 2 K. Taken together, our comprehensive results provide the first concrete evidence that NbGe2 is a promising platform for investigating the interplay between non- trivial band topology, possible Weyl fermions, van Hove singularities, and superconductivity in chiral quantum materials.

O 84.10 Thu 12:45 MA 144

Mode selectivity in electron mediated vibrational relaxation of adsorbed hydrogen on metal surfaces. — •Nils Hertl[1], Connor L. Box[1], and Reinhard J Maurer[1,2] — [1]Department of Chemistry, University of Warwick, Gibbet Hill Road, CV4 7AL, Coventry, United Kingdom — [2]Department of Physics, University of Warwick, Gibbet Hill Road, CV4 7AL, Coventry, United Kingdom

Vibrational relaxation induced by electron-hole pair (ehp) excitations is a common phenomenon in gas-surface experiments on metallic surfaces. Specific adsorbate vibrational degrees of freedom can be more strongly coupled to ehps at metal surfaces than others, which opens an opportunity for mode-selective energy transfer processes that can activate chemical dynamics. In order to promote mode-selective chemical reactions at solid interfaces a detailed understanding of the coupling of the individual vibrations of adsorbates with the electrons and phonons of the metal substrate is essential. To shed light on the mechanisms involved in energy transfer processes present in gas-surface systems, we study adsorbed hydrogen on single crystalline surfaces. Herein, we calculate the lifetimes of vibrational modes of hydrogen adsorbed on the (100) and (110) surfaces of Mo and W via first-principles first-order perturbation theory based on Density Functional Theory (DFT). Our results show a strong mode dependency on the electron-driven relaxation rates of the vibrations. For the vibrations with a Fano-lineshape, our predicted lifetimes are in good agreement with experiments, indicating that the relaxation of those vibrations is dominated by ehp excitations.

O 85: Heterogeneous Catalysis I

Time: Thursday 10:30–12:45 Location: TC 006

O 85.1 Thu 10:30 TC 006

Gazing into Reactors: Adaptive Experimental Design with Concentration Profiles — •Maryke Kouyate, Frederic Felsen, Christian Kunkel, Christoph Scheurer, and Karsten Reuter — Fritz-Haber-Institut der MPG, Berlin

Effective kinetic models of heterogeneous catalytic processes are an indispensable tool for reactor design, optimization and control. Employing simple functional forms like power laws, the adjustable parameters of such models are traditionally fitted to kinetic data measured along local line scans, i.e. by systematically changing individual reaction parameters like a reactant concentration or temperature. Besides the uncertainty of which line scans will optimally determine the model's parameters, this is generally a laborious endeavor involving numerous separate kinetic measurements.

Here we explore the use of profile reactors for an efficient parametrization of effective kinetic models. Developed originally to provide *operando* information along the axis of tubular reactors, a measured concentration profile is effectively nothing but a more complex line scan that provides the kinetic information for all reaction conditions probed along the tube. We propose an adaptive design algorithm that harnesses this wealth of data and provides guidance as to which initial reaction conditions for the next measurement will yield a line scan with maximally complementary information. As demonstrated with simulated data from a plug-flow reactor model, this surpasses conventional adaptive design approaches with "black box" reactors in terms of parameter convergence and required experiments.

O 85.2 Thu 10:45 TC 006

Machine-Learning Assisted Realistic Description of Catalytic Active Centers on the Ternary M1 (Mo,V)O$_x$(100) Surface — •Kyeonghyeon Nam, Yonghyuk Lee, Liudmyla Masliuk, Thomas Lunkenbein, Annette Trunschke, Christoph Scheurer, and Karsten Reuter — Fritz-Haber-Institut der MPG, Berlin

The surface structure and composition of complex heterogeneous catalysts can differ noticeably from assumed, idealistic cuts derived from the bulk material's structure. As a crucial first step toward realistic models of the active surface, we explore the evolution of local atomic-scale structural motifs presented by the catalyst under conditioning and operating conditions. Our focus is on the industrial M1 catalysts used for the selective oxidation of light alkanes. The large primitive cell of the M1 catalyst poses a challenge for a detailed study of all surface terminations using predictive-quality first-principles calculations. To address this challenge, we deconstruct the primitive cell into 'pillar structures' of surface motifs with varying oxygen content. Machine-learned Gaussian approximation potentials, trained against this structural library [1], are used to identify *operando* stable (100) surfaces of ternary M1 catalysts via *ab initio* thermodynamics and comparison to experimental data from electron microscopy. Subsequent electronic structure calculations provide a detailed picture for various (hk0) surfaces, shedding light on the impact of surface stabilization on catalytic centers.

[1] L. Masliuk *et al.*, J. Phys. Chem. C **121**, 24093 (2017).

O 85.3 Thu 11:00 TC 006

Infra-red spectra prediction of functionalized copper nanoparticles using machine learning force fields — •Nitik Bhatia, Ondrej Krejci, and Patrick Rinke — Department of Applied Physics, Aalto University, FI-00076 AALTO, Finland

Vibrational spectroscopy serves as a powerful tool for in vivo chemical reaction analysis, allowing the identification of distinctive peaks as markers for specific chemical groups. Despite yielding valuable insight, the qualitative and quantitative analysis of IR spectra is aggravated by the presence of other, neighboring species. In this work we investigate the vibrational spectra of several small molecules adsorbed on copper nanoparticles, commonly used in heterogeneous catalysis. We compute the infrared spectra using ab initio molecular dynamics (AIMD) [1]. We achieve excellent agreement with experimental results, but at a high computational cost. We therefore train neural network force fields like FieldSchNet [2] on our AIMD data to capture pertinent spectral properties more efficiently ($\sim$2500 times faster). FieldSchNet achieves remarkable accuracy for simple molecules ($<$20 cm^{-1} shift in peak positions). However, functionalized copper nanoparticles introduce a more complex chemical environment and the predicted peak positions are only accurate to $\sim$100 cm^{-1}. To improve the predictive accuracy, we are enhancing our dataset quality and exploring alternative machine learning strategies for fine-tuning the models.

[1] M.-P. Gaigeot, M. Sprik, J. Phys. Chem. B **107**, 10344-10358 (2003). [2] M. Gastegger, K. T. Schütt, K.-R. Müller, Chem. Sci. **12**, 11473-11483 (2021).

O 85.4 Thu 11:15 TC 006

Design of Palladium-Based Alloys for the Catalytic Hydrogenation of Concentrated Acetylene via Mechanochemical Synthesis and Artificial Intelligence — •Lucas Foppa[1], Jacopo de Bellis[2], Klara S. Kley[2], Jonathan Mauss[2], Rohini Khobragade[2], Ferdi Schüth[2], and Matthias Scheffler[2] — [1]The NOMAD Laboratory at the FHI of the MPG and IRIS-Adlershof of the HU Berlin, Germany — [2]MPI Für Kohlenforschung, Germany

The discovery of new materials for catalysis is challenged by the intricate interplay of underlying processes governing the performance. Here, we combine consistent experimental and theoretical data [1] and apply symbolic regression (SR) in order to identify nonlinear relationships between the measured performance and key physicochemical parameters. These parameters are correlated with the most relevant underlying processes governing the reactivity. We apply this approach to the selective hydrogenation of concentrated acetylene on palladium-based alloys synthesized by ball milling.[2] The SR models highlight the crucial interaction of carbon with the catalyst and describe the evolution of the catalyst selectivity with time on stream. Guided by the SR models, new bimetallic and trimetallic alloys are synthesized and their catalytic performance is tested.

[1] R. Miyazaki *et al.*, DOI:10.26434/chemrxiv-2023-x (2023).
[2] K. S. Kley *et al.*, Catal. Sci. Technol. 13, 119 (2023).

O 85.5 Thu 11:30 TC 006

Fischer-Tropsch Kinetics on Cobalt Single Crystals Monitored by Operando STM — •Sebastian Kläger, Katharina M. Golder, and Joost Wintterlin — Ludwig-Maximilians-Universität München, Munich, Germany

The mechanism of the Fischer-Tropsch (FT) synthesis, the reaction of hydrogen and carbon monoxide to give synthetic fuels, is arguably one of the most complex in heterogeneous catalysis. The reaction is highly sensitive to reaction conditions and catalyst pretreatment, and apparent activation energies show poor reproducibility and considerable variations. We present a study in which this issue has been approached on two paths: Single crystal cobalt surfaces have been used as model catalysts on which the FT kinetics could be measured by gas chromatography, and operando scanning tunneling microscopy has been used to image the surfaces under the same conditions. At pressures of 1 bar and temperatures of $\sim$500 K, FT-active, well-controlled systems have been realized that are not disturbed by the complexities affecting the kinetics on supported catalysts. An apparent activation energy has been determined that agrees with a majority of studies on supported catalysts, and also the hydrocarbon selectivity shows similar characteristics. By means of single crystal cobalt surfaces with varying step densities, the kinetic role of atomic steps has been investigated. Our models reproduce the properties of the idealized systems applied in microkinetic simulations, allowing us to test the predictions of recent simulations.

O 85.6 Thu 11:45 TC 006

The relation between substrate, Sm alloy, and surface sensitivity of ceria (111)- and (100)-oriented nano-islands on Ru(0001) and Cu(111). — •Raquel Sanchez-Barquilla, Rudi Tschammer, Lars Buss, Carlos Morales, and Jan Ingo Flege — Applied Physics and Semiconductor Spectroscopy, Brandenburg University of Technology Cottbus-Senftenberg, Germany

Inverse oxide/metal catalysis allows achieving better catalytic performance than its traditional counterpart. For example, in cerium-based inverse catalyst systems, the Ce^{3+} states have been shown to be the active sites for methanol synthesis. This suggests that the activity can be enhanced by promoting those through alloying with trivalent, catalytically active rare-earth metals, as, e.g. Sm. We present low-energy and

X-ray photoemission electron microscopy (LEEM/XPEEM), investigations that show how epitaxially grown (100)- and (111)-oriented CeO_2 islands may be modified and/or alloyed by post-deposited metallic Sm. For the $Ce_{1-x}Sm_xO_{2-\delta}$/Ru(0001) system, the CeO_2 (111)-oriented islands undergo a structural change, concomitant with a partial conversion from Ce^{4+} to Ce^{3+}. Surprisingly, for $Ce_{1-x}Sm_xO_{2-\delta}$/Cu(111) the result is found to be face-dependent since only (100)-oriented CeO_x islands were reduced whereas the (111)-oriented islands remained unaltered. Both systems have been exposed to reducing (H_2) and oxidizing (CO_2) conditions, resulting in higher reduction and in a complete recovery of the Ce^{4+} states, respectively. These unexpected results indicate a complex interaction not only between cerium and the doping element, but also an intricate interplay with the metallic substrate.

O 85.7 Thu 12:00 TC 006

MoS2-based model catalysts for hydrotreatment of biooils: Combined insights from STM & XPS from UHV to operando conditions — •Lars Mohrhusen, Martin Hedevang, and Jeppe V. Lauritsen — Aarhus University, interdisciplinary Nanoscience Center (iNano), Aarhus, Denmark

Hydrotreatments of (fossil) oils using MoS2-based catalysts are well-established processes for heteroatom removal. The process is becoming relevant in the energy transition, e.g. for upgrading of biooils from sustainable feedstocks, f.ex. from biomass pyrolysis, which however appear as complex feedstocks rich in impurities and heteroelements (O, N) and thus remain challenging due to catalyst deactivation.

In contrast to the established use in (oxygen free) hydrodesulphurization (HDS), where in-situ formed S vacancies act as active sites, the sulphide catalyst will be exposed to nitrogenates and oxygenates in the desired hydrodeoxygenation (HDO) and hydrodenitrogenation (HDN) processes. Thus, S atoms may be partially exchanged by N or O, triggering a strong catalyst degradation on the long term.

Herein, we present combined insights from scanning tunnelling microscopy (STM) and x-ray photoelectron spectroscopy (XPS) from UHV to near-ambient pressure (NAP) conditions to mimic hydrotreatments on 2D MoS2 nanoparticles on Au (111) as our model system. Our results show that the MoS2 nanoparticles are surprisingly robust but respond to elevated H2O levels by a small uptake of oxygen leading to an ongoing restructuring of the catalyst. The rate of the oxidation can be steered by the presence of hydrogen in the feed.

O 85.8 Thu 12:15 TC 006

Labile Oxygen-rich Mullite for Ozone Activation to Produce Highly Oxidizing Surface Peroxide — •Linfeng Su[1], Xu Chen[2], Huaping Zhao[1], Zhiyi Lu[2], and Yong Lei[1] — [1]Fachgebiet Angewandte Nanophysik, Institut für Physik & IMN MacroNano, Technische Universität Ilmenau, 98693 Ilmenau, Germany — [2]Key Laboratory of Advanced Fuel Cells and Electrolyzers Technology of Zhejiang Province, Ningbo Institute of Materials Technology and Engineering, CAS, Ningbo, Zhejiang, 315201, PR China

Materials with labile oxygen (Olab) have represented great potential in catalysis owing to their unique electronic structure over conventional adsorbed oxygen and lattice oxygen. In this work, we demonstrate that the Olab can be generated in Mullite-Olab by introducing Al-O4 tetrahedral. The bridging oxygens between Al-O4 and M-O4 (M=Al or Si) are identified as Olab. Mullite-Olab exhibits excellent performance in catalytic ozone activation with a high quasi-first order rate constant (k=0.112 min-1) for degradation of atrazine, while the degradation constant closes to 0 for mullite without Olab. Operando Raman and DFT simulations further reveal that Olab is the catalytic active center, which activates ozone via an unusual surface peroxide pathway to generate surface high-oxidative Olab-O* species. Additionally, the catalytic ozonation based on Mullite-Olab also plays a crucial role in treating actual acrylic fiber wastewater. This work provides an advanced Mullite-Olab catalyst with an unconventional ozonation mechanism, promising for future non-biodegradable wastewater treatment.

O 85.9 Thu 12:30 TC 006

Method development for gas adsorption measurements on proton exchange membrane fuel cell catalyst layers. — •Maria Kolla[1,2], Kläre Christmann[1], Sophia Gierse[1], Ulf Groos[1], and Andreas Bett[1,2] — [1]Fraunhofer ISE, Freiburg, Germany — [2]Albert-Ludwigs-Universität, Freiburg, Germany

This work focuses on developing a method to analyze the porosity and surface area characteristics of materials employed in Proton Exchange Membrane (PEM) fuel cells using gas adsorption technology. A comprehensive understanding of porosity's impact on cell efficiency is essential for designing materials that boost fuel cell performance. Enhanced fuel cell durability, especially in electric vehicles, contributes to transportation advancements. The core of the PEM fuel cell is the catalyst coated membrane (CCM), which consists of a membrane with electrode on each side, called catalyst layers (CL). CL requires substantial porosity to enable gas flow for the occurring reactions. Porosity arises through porous carbon-based powder, but by inserting platinum (Pt) particles as catalyst material and ionomer as binder, porosity is reduced. Hence, the variation in carbon support, Pt particle sizes and amount, amount of ionomer, and finally operation of the CCM, the porosity can vary. To study the impact of the material, and operation variations on the porosity, gas adsorption studies are concluded on a small amount of sample material. Reduction of the material needed for a reliable analysis, is the ultimate objective of this work. The proof of concept will be performed by analyzing different materials and finally comparing the porosity of a fresh and an aged CCM.

O 86: Focus Session: Wetting on Adaptive Substrates II (joint session CPP/DY/O)

The focus session aims to discuss recent developments in the wetting dynamics of adaptive, deformable, and switchable surfaces.

Time: Thursday 11:30–13:00 Location: H 0107

Invited Talk O 86.1 Thu 11:30 H 0107

A multi-scale approach to characterize wetting within a porous medium — •Maja Rücker[1], Ryan T. Armstrong[2], Chenhao Sun[3], Peyman Mostaghimi[2], Steffen Berg[4,5], Paul Luckham[5], Apostolos Georgiadis[4,5], and James E. McClure[6] — [1]Eindhoven University of Technology, Eindhoven, NL — [2]University of New South Wales, Sydney, Australia — [3]China University of Petroleum, Beijing, China — [4]Shell Global Solutions International B.V., Amsterdam, NL — [5]Imperial College London, London, UK — [6]Virginia Tech, Arlington, VA, USA

Considering a porous medium with two fluids in it, the fluid flow and distribution will depend on the wetting characteristic of the system. Recent developments in imaging techniques, such as micro-computed tomography and atomic force microscopy, alongside advances in computational modelling allowed for new concepts linking macroscopic wetting responses to the fundamental microscopic wetting definitions to emerge. We present an approach to upscale wetting parameters building upon energetic and geometric considerations and accounting for the various mechanisms related to wetting, manifesting at those different length scales. Starting from fluid-solid interactions, through the motion of three-phase contact lines to the evolution of fluid configurations in the porous medium, we show experimental observation on a coherent set of fluid/fluid/solid systems and discuss those in relation to the proposed upscaling concept. We will highlight the remaining questions for the characterization of wetting in the context of multiphase flow in porous media and point out future research directions.

O 86.2 Thu 12:00 H 0107

Soft Wetting Transition — •Christopher Henkel[1], Vincent Bertin[2], Jacco H. Snoeijer[2], and Uwe Thiele[1,3] — [1]Institut für Theoretische Physik, Universität Münster, Germany — [2]Physics of Fluids Group, Faculty of Science and Technology, Mesa+ Institute, University of Twente, The Netherlands — [3]Center for Nonlinear Science (CeNoS), Universität Münster, Germany

We investigate the forced receding dynamics of a three-phase contact line on a viscoelastic substrate. Thereby, we use the Landau-Levich (or dip-coating) geometry, where a solid viscoelastic plate is dragged out of a liquid bath. We employ a mesoscopic hydrodynamic model in long-wave approximation, i.e. valid at small contact angle and plate inclination. The elastic response of the substrate follows the Winkler foundation with a Kelvin-Voigt relaxation. In particular, we investigate how the shape and stability of the meniscus change with the plate velocity and the viscoelastic substrate properties. Finally we compare numeric results with asymptotic analytic calculations.

O 86.3 Thu 12:15 H 0107

Demixing around liquid droplets — •Khalil Remini and Ralf Seemann — Experimental Physics, Saarland University, Saarbrücken, Germany

Equilibrium polystyrene droplets are explored sitting on soft solid substrates. The soft solid substrates consist of commercial polydimethylsiloxane (PDMS) elastomer kits with elastic module varying across several orders of magnitude (3-1200 kPa). Inspecting the there-phase contact line of the droplets on a nanoscopic length scale by atomic force microscopy, i.e. on a length scale well below the elasto-capillary length, where capillary forces are higher than the elastic force of the substrate, we find that the ridge formed by the elastic substrate around the droplet is similar to that of a liquid. Material contrasts confirm a liquid ring surrounding the droplets at the three-phase contact line, as a result from stress induced demixing of non-crosslinked PDMS molecules from the PDMS elastomer matrix. This liquid ring extends for softer PDMS elastomers having a larger content of non-crosslinked molecules, but it is present also for the stiffer elastomers. So, the Neumann construction at the three-phase contact line is valid for all tested PDMS elastomers when measuring locally with sufficient resolution.

O 86.4 Thu 12:30 H 0107

Chemical reactions confined in liquid films: dynamics and stability — •Tilman Richter[1], Paolo Malgaretti[1], Thomas M. Koller[2], and Jens Harting[1] — [1]Forschungszentrum Jülich GmbH, Helmholtz-Institut Erlangen-Nürnberg für Erneuerbare Energien (IEK-11), Cauerstr. 1, 91058 Erlangen, Germany — [2]Institute of Advanced Optical Technologies - Thermophysical Properties (AOT-TP), Department of Chemical and Biological Engineering (CBI) and Erlangen Graduate School in Advanced Optical Technologies (SAOT), Friedrich-Alexander-Universit Erlangen-Nuernberg (FAU), Paul-Gordan-Straße 8, 91052 Erlangen, Germany

In catalytic reactions occurring within small liquid droplets or thin liquid films, the yield significantly differs from that in bulk reactions. This variance is primarily due to the fluid interface's substantial surface-to-volume ratio. The fluid interface initiates several phenomena, such as increased surface diffusion, Marangoni flows, and more effective surface interactions, which can boost yield substantially. We develop a theoretical model that demonstrates that the uneven distribution of reactants and products, caused by chemical reactions, can induce Marangoni flows. These flows then alter the spatial distribution of the catalyst. This complex interaction offers novel methods for either inhibiting or promoting the rupture of thin films, as well as for modifying the shape of small droplets.

O 86.5 Thu 12:45 H 0107

Dynamic vesicles on adaptive surfaces — •Lucia Wesenberg, Ben Rasmus Sprötge, Kai-Uwe Hollborn, and Marcus Müller — Institut für theoretische Physik, Georg-August-Universität Göttingen

Vesicles on substrates play a fundamental role in numerous biological transport processes, such as the neurotransmitter release at the synapse, transport vesicles in cells, or the nutrient intake of trees by large vesicles. For all of these processes the adaptive adhesion of the vesicles to a biological substrate is crucial. Furthermore, it is interesting to compare how these adaptive processes differ from wetting of liquid droplets as their shapes seem similar, however, one is governed by bending rigidity and the other by tension.

Here, we study the equilibrium shapes of vesicles as well as their dynamic adaptation to a changing substrate. Our simulations show the significant impact of buoyancy on the vesicle shape, especially in the contact zone. We are able to construct an adsorption diagram summarizing the metastable region of upward buoyant adsorbed vesicles. Further, we study permeable vesicles, adapting dynamically to substrates with a constant adhesion potential with periodically modulated strength or a steplike potential with a given velocity. A step in the potential allows the steering of vesicles, while periodic switching enables controlled vesicle substrate contact. Thus, both cases prove to be an effective control mechanism for biological transport of vesicles.

O 87: Focus Session: Molecular Nanostructures on Surfaces: On-Surface Synthesis and Single-Molecule Manipulation III

Time: Thursday 15:00–18:00 Location: HE 101

Topical Talk O 87.1 Thu 15:00 HE 101

Towards the third dimension: Organic molecular architectures synthesized on solid surfaces by means of solution-based click chemistry — •Michael Dürr — Institut für Angewandte Physik and Zentrum für Materialforschung, Justus-Liebig-Universität Giessen, Germany

In most on-surface reactions, the solid (metal) substrate acts both as a template as well as a catalyst. When we think on expanding the concept of on-surface synthesis into the third dimension, the coupling reactions will not be catalyzed by the substrate any more. Thus, either catalyst-free reaction schemes or new reaction strategies have to be applied. For the growth of 3D structures on silicon surfaces, we have followed two alternative routes: Using a carefully tuned enolether/tetrazine cycloaddition, a second layer of organic molecules was directly coupled under UHV conditions on a prefunctionalized Si(001) surface. As an alternative approach, we combined UHV-based surface functionalization with solution-based click chemistry. In this scheme, ordered multilayers of organic molecules were realized on the prefunctionalized Si(001) surface via a two-step reaction cycle in solution. In order to ensure a controlled layer-by-layer growth, two orthogonal alkyne-azide click reaction schemes based on asymmetric bifunctional molecular building blocks were employed. Each reaction step was monitored by means of XPS demonstrating a high selectivity of the reaction schemes in use. Our approach thus opens the road for the controlled synthesis of organic 3D structures on surfaces.

O 87.2 Thu 15:30 HE 101

Probing dynamic covalent chemistry in a 2D boroxine framework — Paul Leidinger[1], Mirco Panighel[2], Virginia Pérez-Dieste[3], Ignacio Villar-Garcia[3], Pablo Vezzoni[1], Felix Haag[1], Johannes V. Barth[1], Francesco Allegretti[1], Sebastian Günther[1], and •Laerte L. Patera[1,4] — [1]Technical University of Munich, Garching, Germany — [2]CNR-IOM, Trieste, Italy — [3]CELLS-ALBA, Cerdanyola del Valles, Spain — [4]University of Innsbruck, Innsbruck, Austria

The use of dynamic covalent chemistry provides a powerful method for designing covalent organic frameworks, exploiting reversible bond formation to achieve remarkable crystallinity. Here we employed near-ambient pressure X-ray photoelectron spectroscopy to dissect the reversible construction of a two-dimensional boroxine framework on a Au(111) surface. By mapping the pressure-temperature parameter space, we identified the regions where the rates of the condensation and hydrolysis reactions become dominant. This precise identification is critical for enabling thermodynamically controlled growth of highly crystalline frameworks [1]. [1] P. Leidinger, et al., Nanoscale 15, 3, 1068-1075 (2023).

O 87.3 Thu 15:45 HE 101

The Chemical Functionalization of the Biphenylene Network predicted by DFT — •Hendrik Weiske and Ralf Tonner-Zech — Universität Leipzig, Leipzig, Deutschland

2D materials, such as graphene, could introduce a new disruptive technology, replacing some of the currently used materials. The covalent functionalization of graphene is still relatively uncommon and limited to powerful reagents, e.g., free radicals, n-BuLi, or strong nucleophiles.[1] The biphenylene network (BPN) is a recently synthesized 2D material[2] with a promising application range, from photocatalysis to superconductivity. While hydrogenated or fluorinated derivatives of BPN are discussed in the literature already, the molecular functionalization is an unexplored topic in the field. We show that the unique electronic structure of BPN enables functionalization using the stable unsaturated cyclic hydrocarbon cyclooctyne. Cyclooctyne is known to form well-ordered layers on Si(001), which we studied extensively in the past.[3,4] We propose cyclooctyne as a promising molecule to functionalize BPN towards new properties. We extensively studied the adsorption kinetics of cyclooctyne, thermodynamics, chemoselectivity for the reaction, and its effect on the electronic structure.

[1] I. A. Vacchi, A. Bianco, et al., Phys. Sci. Rev., **2017**, *2*. [2] Q. Fan, A. S. Foster, U. Koert, P. Liljeroth, J. M. Gottfried, et al., Science **2021**, *372*, 852-856. [3] F. Pieck, R. Tonner-Zech, Molecules, **2021**, *26*, 6653 [4] L. Pecher, R. Tonner, et al., J. Phys. Chem. C, **2017**, *121*, 26840-26850

O 87.4 Thu 16:00 HE 101

Reactivity of Aromatic Amines on Metal Surfaces — Nan Cao[1,2], Kaifeng Niu[3], Wenchao Zhao[2], Jonas Björk[3], Lifeng Chi[1], Johannes Barth[2], and •Biao Biao[1,2] — [1]Soochow University, Suzhou 215123 (P. R. China) — [2]Technical University of Munich, D-85748 Garching, (Germany) — [3]Linköping University, 58183 Linköping (Sweden)

Aromatic amines are widely used as intermediates to dyes, pharmaceuticals, and agricultural chemicals, whereas the oxidation process is of great significance for their direct application and synthesis of other complex nitrogen-containing compounds. Dehydrogenation of the amino group in aromatic amines on copper surfaces is usually achieved with thermal treatment. However, amino has no reactivity reported on relatively inert surfaces of silver and gold. Herein, we combine STM and XPS studies to explore the reactivity of aromatic amines on silver and gold surfaces with the O2-gas mediation[1]. While oxidative dehydrogenation reaction is achieved on silver surfaces, forming the N-Ag complex with intrinsic Ag adatoms, aromatic amine layers remain intact on the gold surface with the O2-mediated treatment. Complementary insights from density functional theory calculations of reaction pathway, core level shift, and chemisorption energy reveal the thermodynamically substrate-dependent mechanics. The systematic study unveils the mechanism of catalytic oxidative dehydrogenation reaction of amines, enriching the on-surface synthesis toolbox for the production of low-dimensional organic materials. Reference [1] Nan Cao, Biao Yang et al. Nat. Commun. 14, 1255 (2023)

O 87.5 Thu 16:15 HE 101

An alternative on-surface synthesis approach on Ag(111) studied by STM and XPS — •Paul Schweer[1], Christoph Vonnemann[2], Stefan Huber[2], and Karina Morgenstern[1] — [1]Chair of Physical Chemistry I, Ruhr University of Bochum, Germany — [2]Chair of Organic Chemistry II, Ruhr University of Bochum, Germany

On-surface synthesis uses planar precursors containing carbon atoms that are sp2 hybridized and halogen functionalized to construct 2D structures by thermal activation. The mechanism includes the cleavage of the carbon-halogen bond, followed by an intermolecular C-C coupling. In this talk, we will present an alternative approach based on a polyaromatic molecule with a single sp3 hybridized fluorinated carbon atom as the reactive center. The reaction is induced by thermal and photoactivation on Ag(111). We describe the self-assembly of the molecules and the mechanisms of their on-surface reaction by means of scanning tunneling microscopy and X-ray photoelectron spectroscopy investigations. These results give insights into an intramolecular reaction pathway that produces single cyclized structures.

O 87.6 Thu 16:30 HE 101

Functionalization of Surfaces with Ordered Arrays of Endohedral Fullerenes — •Lukas Spree, Caroline Hommel, Luciano Colazzo, and Andreas Heinrich — IBS Center for Quantum Nanoscience, 52 Ewhayeodae-gil, Daehyeon-dong, 03760 Seoul, South Korea

Endohedral fullerenes are hollow, sperical carbon structures, that can encase and protect very exotic configurations of certain elements. Confining several atoms inside the carbon cage leads to a unique combination of desirable physical properties, chemical stability, and tunability. Studies of endohedral fullerenes in bulk have yielded some very promising single molecule magnets and spin-qubit candidates.

Making these fascinating compounds accessible to characterization and manipulation by scanning probe microscopy techniques is challenging. Short range order can often not be achieved, because freely rotating encapsulated species and energetically similar configurations of the cages on the substrate offer high degrees of freedom. This leads to variance in the magnetic and coherent properties of the individual molecules. Chemical functionalization of the fullerene cages and the use of template layers made of tetrapyrroles like porphyrines offer a way to reduce the number of possible configurations.

The combination of different molecular species on metallic substrates in ultrahigh vacuum requires the use of various deposition techniqes. Electrospray deposition, sublimation, and formation of monolayers at substrate-solution interfaces are discussed.

O 87.7 Thu 16:45 HE 101

Low temperature atomic force microscopy with an adaptive tunneling current feedback for simultaneously visualizing chemical structures and adsorption positions of organic molecules — Daniel Martin-Jimenez[1,2,3], Qigang Zhong[1,2], André Schirmeisen[1,2], and •Daniel Ebeling[1,2] — [1]Institute of Applied Physics (IAP), Justus Liebig University Giessen, Germany — [2]Center for Materials Research (LaMa), Justus Liebig University Giessen, Germany — [3]Institut de Ciencia de Materials de Barcelona (ICMAB-CSIC), Campus Bellaterra, Barcelona, Spain

Low temperature atomic force microscopy with CO-functionalized tips allows to visualize the chemical structure of adsorbed organic molecules. This tool became essential for the field of on-surface synthesis as it enables revealing the structures of the products and corresponding reaction mechanisms. Established methods for determining adsorption positions of precursors, intermediates, and products, which are important for a direct comparison with calculated adsorption structures, however, remain rather complex and time-consuming. Here, we illustrate a relatively simple method that can be used to simultaneously visualize the chemical structure of organic molecules and surface atoms in a single scan. The proposed method is based on automatically switching between different tunneling feedback parameters, which are optimized for the two different tasks. Switching between feedback parameters allowed to reliably image highly mobile 2-iodotriphenylenes on a Ag(111) surface with sub-molecular resolution and precisely determine the positions of Ag(111) top sites in their close vicinity.

O 87.8 Thu 17:00 HE 101

NC-AFM characterization of single-atom catalytic sites within metal-coordinated supramolecular networks — Bertram Schulze Lammers[1], Nieves López-Salas[2], Julya Stein Siena[2], Hossein Mirhosseini[3], Damla Yesilpinar[1], Julian Heske[3], Thomas D. Kühne[3], Harald Fuchs[1], Markus Antonietti[2], and •Harry Mönig[1] — [1]Physikalisches Institut, Universität Münster — [2]Max Planck Institute of Colloids and Interfaces, Potsdam — [3]Theoretical Chemistry, Universität Paderborn

For the development of single atom catalysts, non-noble metal-organic layers combine functional variability with cost efficiency. Here we investigated reacted layers of melamine and melem on a Cu(111) surface by NC-AFM with CuOx-functionalized tips [1]. Upon deposition and subsequent heat treatments, the molecular units are completely deprotonated and adsorb flat and strongly chemisorbed. We observe a particularly extreme interaction of the deprotonated N- with single Cu atoms located at intermolecular sites. The NC-AFM measurements and ab-initio simulations indicate a pronounced interaction of O-species at these N-Cu-N sites. To study potential catalytic properties, we performed cyclic voltametry experiments on our samples at ambient pressure within a drop of electrolyte in a controlled O_2 or N_2 environment. Both Cu-nitride structures show a robust activity in irreversibly catalyzing the reduction of oxygen. The activity is assigned to the intermolecular N-Cu-N sites or corresponding oxygenated versions (N-CuO-N, N-CuO_2-N) [2]. [1] H. Mönig, ChemComm 54, 9874 (2018); [2] B. Schulze Lammers et al., ACS Nano 16, 14284 (2022).

O 87.9 Thu 17:15 HE 101

On-Surface Synthesis of Oxygen Annulated Porphyrins — •Joel Deyerling[1], Beatrice B. Berna[2], Dmytro Biloborodov[2], Felix Haag[1], Sena Tömekce[1], Marc G. Cuxart[1], Davide Bonifazi[2], and Willi Auwärter[1] — [1]Physics Department E20, TUM School of Natural Sciences, Technical University of Munich, Garching, Germany — [2]Institute of Organic Chemistry, Faculty of Chemistry, University of Vienna, Vienna, Austria

Recently the on-surface synthesis of fused porphyrins through C-H activation followed by C-C bond formation has generated considerable interest [1-3]. These porphyrins afford various edges and topologies, thus, offering precise control of their functional properties. From a chemistry point of view, the toolbox is currently limited to C-C coupling, while on-surface C-heteroatom fusing to porphyrins is elusive.

In this context we explore the on-surface oxygen annulation of Ni(II)-tetra- and octahydroxyporphyrins on Au(111). Upon thermal activation, the ortho-positions of the meso-aryl substituents (dihydroxyphenyls) are connected by an O-bridge to the β-pyrrolic positions of the porphyrin macrocycle, creating pyran rings. Employing STM, nc-AFM and high-resolution XPS, we provide a comprehensive characterization of the on-surface reaction. Beyond that, we elucidate how the oxygen tailors the electronic structure and drives the assembly of the achiral molecules into chiral lattices.

[1] Sun, Q. et al., JACS, 142, 18109-18117 (2020).
[2] Yang, X.-Q. et al., ACS Nano, 16, 13092-13100 (2022).
[3] Deyerling, J. et al., J. Phys. Chem. C, 126, 8467-8476 (2022).

O 87.10 Thu 17:30 HE 101

A temperature-programmed X-ray photoelectron spectroscopy study of a halogenated porphyrin on Au(111) and Cu(111) — •Michael Clarke[1], Eleanor Frampton[2], Matthew Edmondson[1], Ailish Gray[1], Jonathan Bradford[1], and Alex Saywell[1] — [1]School of Physics & Astronomy, University of Nottingham, UK — [2]MAX IV Laboratory, Lund, Sweden

Halogenated porphyrins on metallic surfaces undergo a variety of reactions when heated, such as Ullmann-type coupling, ring-closing, metalorganic coordination, and self-metalation. These reactions are influenced by various factors, including porphyrin and substrate chemistry.[1] Porphyrins are of interest as precursor monomers in on-surface assembly, due to their tuneable optoelectronic and gas absorption properties.[2] Here, we detail the reaction of a brominated porphyrin (BrxTPP) on Au(111) and Cu(111), via near edge X-ray absorption fine structure (NEXAFS), X-ray photoelectron spectroscopy (XPS) and scanning tunnelling microscopy (STM). Building upon our previous work, [3,4] temperature programmed XPS (TP-XPS) provides a detailed characterisation of the molecular structure to be obtained as the reaction progresses. The chemistry, orientation, and conformation of the porphyrins is explored.

[1] L. Grill, S. Hecht, Nat. Chem. 12, 115 (2020) [2] E. Vesselli, J. Phys. Mater. 3, 022002 (2020) [3] M. Edmondson, E.S. Frampton, C.J. Judd, N.R. Champness, R.G. Jones and A. Saywell, Chem. Commun., 2022, 58, 6247. [4] E.S. Frampton, M. Edmondson et al., Inorganica Chim. Acta, 558, 121718, (2023)

O 87.11 Thu 17:45 HE 101

On-Surface Reaction of Tetraphenylporphyrin with Caesium — •Leonard Neuhaus, Florian Münster, Kassandra Zoltner, Lukas Heuplick, Jan Herritsch, and J. Michael Gottfried — Department of Chemistry, University of Marburg, 35043 Marburg, Germany

Tetrapyrroles such as porphyrins and their metal complexes play important roles in living organisms and in modern technologies, e.g., in energy storage, (electro-)catalysis and sensor systems. While transition metal complexes of tetrapyrroles on surfaces have been well studied in the last decades, much less is known about their complexes with alkali metals. Here, we report studies of the reaction of tetraphenylporphyrin (H2TPP) with vapor-deposited caesium in the multilayer and (sub)monolayer regimes on Ag(111). XPS indicates for the submonolayer and multilayer partial metalation to Cs2TPP already at 300 K, whereas in monolayer regime no reaction is observed at this temperature. STM however, shows that the Cs atoms still coordinate to the porphyrin molecules at 300 K. After annealing to 400 K full conversion is observed. DFT calculations suggest that the Cs2TPP complex has a bipyramidal structure with Cs ions on both sides of the molecular plane, which is in line with STM measurements. In complementary temperature-programmed reaction (TPR) experiments, the desorbing species were clearly identified by mass spectrometry as Cs2TPP. TPR also indicates that Cs2TPP is thermally stable up to at least 700 K.

O 88: Focus Session: Ultrafast Processes in Organic Semiconductors and Perovskites II (joint session O/CPP)

Time: Thursday 15:00–18:00 Location: MA 004

Topical Talk O 88.1 Thu 15:00 MA 004

Time-resolved chiroptical probes to track spin & light polarization in solution-processable semiconductors — •Sascha Feldmann — Harvard University, Cambridge, MA, USA

Chiral solution-processable semiconductors based, for example, on small molecules, polymers or halide perovskites offer an exciting new avenue to simultaneously control charge, spin and light using a single material. This could enable efficient spin-optoelectronic devices ranging from displays and holography to detectors, and even applications in quantum information technology.[1] In this talk, I will give an overview of our recent efforts to understand the underlying mechanisms by developing novel time-resolved chiroptical spectroscopy techniques.

By pushing broadband circular dichroism to diffraction-limited spatial and 15 fs time resolution, we create a spin cinematography technique to witness the ultrafast formation of spin domains in halide perovskite films due to local symmetry breaking and spin-momentum locking.[2] In terms of circularly polarized photoluminescence (CPL), I will first explain the fundamentals and artifacts involved in measuring CPL reliably and introduce an open-access methodology and code to do so [3]. I will conclude by showing our most recent development of a transient broadband full Stokes vector polarimetry with unprecedented time and polarization resolution to track the emergence of chiral light emission [to be submitted].

[1] Nature Reviews Materials 8, 365 (2023)
[2] Nature Materials 22, 977 (2023)
[3] Advanced Materials 44, 2302279 (2023)

O 88.2 Thu 15:30 MA 004

Nonlinear Pathways for Coherent Control of Lead Halide Perovskite Lattices — •Sebastian F. Maehrlein — Fritz Haber Institute of the Max Planck Society, Berlin

Lead halide perovskites (LHPs) constitute a vast and highly diverse library of emerging semiconductors. So far, to tailor their optoelectronic properties, the material science community mainly focused on changing the static design of the perovskite lattice by tuning the chemical composition or morphology. Meanwhile, the full potential for dynamic phonon-driven ultrafast material control, as successfully applied for oxide perovskites, has rarely been exploited.

I will present a summary of linear and nonlinear, phononic and photonic, excitation pathways to coherently control lattice vibrations with the help of intense THz fields. This comprehensive toolset not only allows to drive also non-IR active modes, but additionally unlocks phonon phase-sensitive lattice trajectory control. In prototype MAPbBr3 and CsPbBr3, we demonstrate coherent control of the octahedral twists [1], which act as structural order parameter and modulate the optical bandgap. Moving to more complex systems, we unveil counterintuitive lattice coherences in mixed 4-cation LHPs, proving the versatility of our method and providing active feedback to the design of novel energy materials.

We acknowledge fruitful collaborations with the groups of X.-Y. Zhu (Columbia U.), T. Kampfrath (FU Berlin), and S. Mathur (U. of Cologne).

[1] Frenzel et al., Sci. Adv.9, eadg3856 (2023)

O 88.3 Thu 15:45 MA 004

Confined Excitons in Lead Halide Perovskite Quantum Dots — •Lena Stickel, Anja Barfüsser, Quinten A. Akkerman, and Jochen Feldmann — Chair for Photonics and Optoelectronics, Nano-Institute Munich and Department of Physics, Ludwig-Maximilians-Universität (LMU), Königinstr. 10, 80539 Munich, Germany

In the past years, lead halide perovskite quantum dots (LHP-QDs) have gained massive attention, due to their unique optoelectronic properties.[1] We discuss sphere-like LHP-QDs, which exhibit distinct resonances in their absorption spectra.[2] By comparing experimental data with model calculations based on weak and strong confinement, the origin of these resonances was investigated. Pump-probe experiments show bleaching and induced absorption signals, which we assign to excitonic and biexcitonic transitions.[3] We further use these well-defined excitonic QDs to study the interaction between LHP-QDs and functionalized ligands.

[1] A. Dey et al. ACS Nano 2021, 15, 10775-10981.
[2] Q. A. Akkerman et al. Science 2022, 377, 1406-1412.

[3] A. Barfüßer et al. Nano Lett. 2022, 22, 8810-8817.

O 88.4 Thu 16:00 MA 004

Two-Dimensional Electronic Spectroscopy from First Principles — •Jannis Krumland^{1,2}, Michele Guerrini^{1}, Antonietta De Sio^{1}, Christoph Lienau^{1}, and Caterina Cocchi^{1,2} — ^{1}Carl von Ossietzky Universität Oldenburg, Physics Department and Center for Nanoscale Dynamics (CeNaD), D-26129 Oldenburg, Germany — ^{2}Humboldt-Universität zu Berlin, Physics Department and IRIS Adlershof, D-12489 Berlin, Germany

In recent decades, two-dimensional electronic spectroscopy has been established as an experimental tool that can provide unprecedented insight into ultrafast light-matter interaction. Employing three to four concerted laser pulses, this technique is able to map out the third-order polarizability of molecules and extended systems, which contains rich information about nonlinear optical properties as well as vibronic excited-state dynamics following photoexcitation. Here, we present a first-principles approach to the simulation of two-dimensional spectra based on real-time time-dependent density functional theory and Ehrenfest molecular dynamics, applying it to prototypical molecules such as benzene and pyrene [1]. In contrast to other theoretical schemes, our approach does not require any external parameters and thus offers more predictive power. Several numerical techniques are employed to mitigate the computational complexity, making it manageable even for larger systems that are out of reach with other atomistic methods.

[1] J. Krumland et al., arXiv:2308.09062 (2023)

O 88.5 Thu 16:15 MA 004

Excited state symmetry breaking and solvation in quadrupolar dyes — •Katrin Winte^{1}, Somayeh Souri^{1}, Daniel Lünemann^{1}, Fulu Zheng^{2}, Mohamed Madjet^{2}, Teresa Kraus^{3}, Elena Mena-Osteritz^{3}, Peter Bäuerle^{3}, Sergei Tretiak^{4}, Antonietta De Sio^{1}, and Christoph Lienau^{1} — ^{1}Oldenburg University, Germany — ^{2}Bremen University, Germany — ^{3}Ulm University, Germany — ^{4}Los Alamos National Laboratory, USA

Quadrupolar acceptor-donor-acceptor (A-D-A) dyes represent a versatile and chemically tunable class of prototypical molecular systems for exploring photo-induced charge transfer processes. Their optoelectronic properties are governed by an interplay between electronic tunneling between two arms and vibronic coupling to high frequency modes in each arm. In polar solvents, optical excitation results in a pronounced solvatochromism thought to arise from a light-induced charge localization inside the molecule, induced by an interplay between intramolecular charge separation and solvation[1]. So far, both processes could not be distinguished experimentally. Here, we report the first observation of the vibronic-coupling induced nonadiabatic charge separation in A-D-As. By comparing pump-probe and 2DES spectra recorded with 10fs time resolution in polar and nonpolar solvents, we demonstrate coherent vibrational wavepacket motion during the first 100fs that is largely unaffected by solvation. Our results give evidence for a vibronic-coupling induced double-minimum potential energy surface inducing spontaneous symmetry breaking and charge separation after photoexcitation. [1] E. Vauthey, J. Phys. Chem. Lett. 13,2064 (2022).

O 88.6 Thu 16:30 MA 004

THz Control of Coherent Phonon Dynamics in Low-Dimensional Hybrid Perovskites — •Joanna M. Urban^{1}, Marie Cherasse^{1}, Michael S. Spencer^{1}, Maximilian Frenzel^{1}, Charlotte Berrezueta Palacios^{2}, Gaelle Trippe-Allard^{3}, Abdelaziz Jouaiti^{4}, Sylvie Ferlay^{4}, Emmanuelle Deleporte^{3}, Stephanie Reich^{2}, Martin Wolf^{1}, and Sebastian F. Maehrlein^{1} — ^{1}Fritz Haber Institute of the Max Planck Society, Berlin, Germany — ^{2}Institute of Experimental Physics, Freie Universität Berlin, Germany — ^{3}Université Paris-Saclay, ENS Paris-Saclay, CentraleSupélec, CNRS UMR 9024, LuMIn, 91190 Gif-sur-Yvette, France — ^{4}Université de Strasbourg-CNRS, UMR 7140, France

Charge carrier-phonon interaction governs the outstanding optoelectronic properties of hybrid lead halide perovskites (LHPs). Understanding their complex vibrational dynamics is crucial for harnessing their full potential. Recently, we used intense THz pulses to nonlinearly drive octahedral twist modes in 3D LHPs, demonstrating coherent lattice control at ultrafast timescales [1]. Low-dimensional hybrid LHPs combine the intriguing properties of the soft, polar, and anharmonic lattice with quantum confinement effects arising from the reduced dimensionality. We study quasi-2D layered Ruddlesden-Popper perovskites and 1D hybrid perovskite-like metal halides by THz Kerr Effect spectroscopy and analyze the changes of lattice dynamics with dimensionality reduction, towards a better understanding of exciton self-trapping and charge carrier screening.

References: [1] Frenzel et al., Sci. Adv.9, eadg3856 (2023)

O 88.7 Thu 16:45 MA 004

Self-Trapped Exciton Emission in Two Families of Antimony and Bismuth Halide Perovskites — •Lukas Gümbel^{1}, Philip Klement^{1}, Meng Yang^{2}, Sangam Chatterjee^{1}, and Johanna Heine^{2} — ^{1}Institute of Experimental Physics I and Center for Materials Research, JLU Gießen, Germany — ^{2}Department of Chemistry and Material Sciences Centre, PU Marburg, Germany

Main group metal halide materials are currently explored for a variety of applications including solar cells, but also light emitting devices (LED), lasers, sensing, and photo-catalysis. They promiss less harmful alternatives to the prominent lead halide perovskites. This study investigates the optical properties of antimony and bismuth halide compounds. Temperature-dependent photoluminescence spectroscopy reveals the electron-phonon coupling andyields a Huang-Rhys factor in trhe range of 5 to 22. The broad emission bands and large Stokes shifts suggest self-trapped exciton (STE) emission, linked to an interaction between the photogenerated electron-hole pairs and the lattice. The intricate interplay of factors like ground and excited state distortion, lattice softness, and electron-phonon coupling necessitates deeper understanding. A systematic analysis contributes to establishing the general structure-property relationships for STE emission in such metal halide perovskite-derivate materials. Additionally, the study delves into the diffusion of self-trapped excitons through temporal and spatial resolved photoluminescence, enhancing comprehension of emission processes.

O 88.8 Thu 17:00 MA 004

Direct observation of ultrafast lattice distortions during exciton-polaron formation in lead halide perovskite nanocrystals — •Hélène Seiler^{1,2}, Zahn Daniela^{1}, Victoria C.A. Taylor^{1}, Maryna I. Bodnarchuk^{3}, Yoav W. Windsor^{1,4}, Maksym V. Kovalenko^{3,5}, and Ralph Ernstorfer^{1,4} — ^{1}Fritz Haber Institute, Berlin, Germany — ^{2}Freie Universität Berlin, Berlin, Germany — ^{3}EMPA, Dübendorf, Switzerland — ^{4}Technische Universität Berlin, Berlin, Germany — ^{5}ETHZ, Zürich, Switzerland

We have employed femtosecond electron diffraction (FED) to directly measure the sub-picosecond lattice dynamics of weakly confined CsPbBr3 nanocrystals following above-gap photoexcitation. The data reveal a light-induced structural distortion appearing on a time scale varying between 380 and 1200 fs depending on the excitation fluence. We attribute these dynamics to the effect of exciton-polarons on the lattice and the slower dynamics at high fluences to slower sub-picosecond hot-carrier cooling, which slows down the establishment of the exciton-polaron population. Further analysis and simulations show that the distortion is consistent with motions of the [PbBr3]- octahedral ionic cage, and closest agreement with the data is obtained for Pb-Br bond lengthening.

O 88.9 Thu 17:15 MA 004

Structural Dynamics during Excimer Formation in Fluorinated Zinc-Phthalocyanine Thin Films — •Sebastian Hammer^{1}, Laurenz Kremeyer^{1}, Tristan Britt^{1}, Maximilian Rödel^{2}, Jens Pflaum^{2,3}, and Bradley Siwick^{1} — ^{1}Departments for Physics and Chemistry, McGill University, Montreal, QC H3A 2K6, Canada — ^{2}Experimental Physics VI, University of Würzburg, 97074 Würzburg — ^{3}CAE (Center of Applied Energy Research) Bayern,, 97074 Würzburg

In molecular solids, the coupling of charge-transfer and Frenkel states facilitated by inter-molecular vibrational modes can lead to the formation of so called excimer states [1]. The formation of these energetically low lying states is accompanied by a significant structural deformation of the local inter-molecular geometry.

In this contribution we examine the dynamics of the structural deformation during the excimer formation in polycrystalline α-zinc-phthalocyanine thin films by means of ultrafast electron diffraction. Our findings show that the geometric relaxation follows a two-step process (300 fs and $\approx$ 15 ps) and the final geometry is stable beyond 300 ps. Tuning the inter-molecular interaction by means of molecular fluorination reveals that the time-scales of the structural relaxation process are critically dependent on the local energetic environment.

Funding from the DFG (Project 490894053) is gratefully acknowledged.

[1] Bialas et al. *J. Chem. Phys. C.* **126** 4067-4081 (2022)

O 88.10 Thu 17:30 MA 004

Interrelation between excimer dynamics and crystal structure of molecular solids: The case of perylene and perfluorotetracene — •Dominik Muth, Anton Krüger, Sebastian Anhäuser, Daniel Bischof, Gregor Witte, and Marina Gerhard — Department of Physics and Material Sciences Center, Philipps-Universität Marburg, Germany

Excimer formation in organic semiconductors has been known for approximately 70 years. The phenomenon has, however, recently attracted increased interest due to potential applications in the fabrication of organic white light emitting diodes. Gaining insight into the process of excimer formation in molecular solids and controlling the excimer properties via the intermolecular arrangement is therefore of great interest, also from a fundamental point of view.

In this work, we investigate the excimer dynamics in single crystals of two organic semiconductors, i.e. the prototypical material perylene as well as perfluorinated tetracene via time resolved photoluminescence spectroscopy. For both materials, two different polymorphs are studied, providing insight into the influence of the molecular packing on the formation of excimers. Beside the excimer signatures, resolving the dynamics on a picosecond time scale allows us to explore short-lived higher energetic features, which potentially originate from precursor states of excimers. Varying the temperature from 10 to 295 K as well as determining the polarization of the emitted signatures reveals thermal barriers for the population of certain states and the orientation of the underlying transition dipole moments.

O 88.11 Thu 17:45 MA 004

Calculation of Diffusion Properties of Molecular Excitons based on Static Optical Spectra — •Chris Rehhagen and Stefan Lochbrunner — University of Rostock, Institute of Physics

The understanding of the mobility of molecular excitons is fundamental for the use of molecular systems in organic solar cells. Especially a small diffusion length is a bottleneck still limiting the efficiency of light harvesting devices. Often, the diffusion constant of molecular excitons is determined by experimental methods observing e.g. exciton-exciton interaction or quenching processes. Additionally, advanced theoretical tools are used for prediction. In both cases, the effort is significant. In this work, we present a method to predict the exciton diffusion constant using static absorption and emission spectra in combination with basic geometric information of the system. Based on Försters theory the diffusion constant can be calculated from the spectral overlap and the electronic coupling. Both properties can be determined from static spectra based on the methods given in the scientific literature. We analyze these methods in detail. Then, the results are compared with experimentally determined diffusion constants in four different PBI-based solid state systems investigated in our group previously, including one-dimensional aggregates, extended nanoparticles and single crystals. In all cases, we find consistency between the experimental results and our prediction.

O 89: Focus Session: 2D Transition Metal Carbides, Nitrides and Carbonitrides III (joint session DS/MM/O)

Properties: Catalysis & electrochemistry; physical properties

Time: Thursday 15:00–17:30 Location: A 053

Invited Talk O 89.1 Thu 15:00 A 053

Heterogeneous catalysis with MXenes: the role of the surface passivating groups and the structural defects — •Alexey Fedorov — Department of Mechanical and Process Engineering, ETH Zürich, CH-8092 Zürich, Switzerland

Mo2CTx, two-dimensional (2D) molybdenum carbide of the MXene family (Tx are passivating surface groups), contains only surface Mo sites and is therefore a convenient model catalyst for structure-activity studies. For instance, the catalytic activity of Mo2CTx in Fischer-Tropsch (FT) synthesis increases when a Tx coverage is minimized, the latter achieved via reductive defunctionalization of Tx groups under H2. However, high temperature H2 treatment of Mo2CTx removes also ca. one third of the carbidic lattice carbon, yielding a 2D-Mo2C1-x material that is an active methanation catalyst. The removal of Tx species is also possible in the FT conditions (i.e., in the presence of CO), and this gives 2D-Mo2C without detectable carbon vacancies and Tx groups. 2D-Mo2C material, in contrast to 2D-Mo2C1-x, converts CO to diesel range alkanes. Other examples considered include dry reforming of methane, (reverse) water gas shift as well as electrocatalytic reactions (HER, NO3RR) of Mo2CTx:M, i.e., a material with dopant sites (M = Co, Fe) replacing Mo sites in the lattice of Mo2CTx.

O 89.2 Thu 15:30 A 053

Pt-doped Ti3C2Tx and Mo2Ti2C3Tx MXenes for catalytic hydrogenation — •Yilong Yan[1], Franck Morfin[1], Stéphane Célérier[2], and Laurent Piccolo[1] — [1]IRCELYON, CNRS & Université Lyon 1, 69626 Villeurbanne, France — [2]IC2MP, CNRS & Université de Poitiers, 86073 Poitiers, France

Transition metal carbides can act as efficient metal-like catalysts or catalyst supports, and MXenes offer renewed possibilities to anchor metal atoms and promote catalytic performances. Herein, we report on the elaboration of Pt/MXene single-atom catalysts and their performance in CO2 and alkadiene hydrogenation reactions.

Anchoring of single Pt atoms is favorable at the surface of Ti3C2Tx and Mo2Ti2C3Tx MXenes. According to in situ XAS and XPS, Pt cations partially reduce upon thermal treatment at 400 °C in H2 flow, while forming bonds with surface M atoms of the MXene. This includes the probable location of Pt atoms at M vacancies or Pt-M substitution, consistently with STEM. In addition, XAS, XRD and TPR reveal MXene restructuring together with desorption of chemical intercalants and terminal groups.

While Ti3C2Tx is inactive, Mo2Ti2C3TX exhibits significant catalytic activity for both reactions. The addition of single Pt atoms on Ti3C2Tx leads to unusually high selectivity to 2-butene from butadiene hydrogenation [Mater. Today Catal. 2023, 2, 100010]. For CO2 hydrogenation, Pt/Ti3C2Tx shows a high Pt-molar activity and almost 100% selectivity to CO; Pt/Mo2Ti2C3TX is even more active, while methane and methanol are formed as minority products.

O 89.3 Thu 15:45 A 053

Cobalt based MXene composites for the Oxygen Evolution Reaction — •Michelle Browne — Young Investigator Group Electrocatalysis: Synthesis to Devices, Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Hahn-Meitner-Platz 1, Berlin 14109, Germany

In the Electrocatalysis: Synthesis to Devices Group at HZB, our research is focused on combining MXenes and metal oxides to create the next generation Oxygen Evolution Reaction (OER) catalysts. Metal oxides are known to be active for the OER but lack high conductivity. On the other hand, MXenes are highly conductive but oxidise readily under several conditions due to its termination sites and don't contain OER active sites. To overcome these issues, we employ several strategies in our group to combine these two materials to make one material which is OER active and high conductive. Furthermore, by blocking the MXene termination sites with a metal oxide, this may lead to less oxidation of the MXenes structure. This presentation will focus on the development of Co-based MXene materials for the OER through various fabrication methods and combining Co with other metal oxide materials (e.g. CoCu and CoRu). The OER performance of the MXene/metal oxides composites compared to their parent materials will be discussed. We will also evaluate the various strategies to one another and how the initial activity and stability of the composite materials are affected.

15 min. break

Invited Talk O 89.4 Thu 16:15 A 053

Ultrafast Photoexcitations in 2D MXenes — •Lyubov Titova — Worcester Polytechnic Institute, Worcester, MA, USA

MXenes are 2D transition metal carbides and nitrides with electronic properties that can be tuned by their chemistry and structure. Metallic-like conductivity, flexibility, high optical damage threshold and ease of processing owing to their hydrophilicity, make MXenes candidates for a host of electronic and optical applications. We use ultrafast optical and THz spectroscopic techniques to investigate the nature and behavior of photoexcitations in MXenes of different chemistries. We show that electronic and optical properties of MXenes can be engineered by choices of the transition metals and their order as well as by controlling the intercalants in the interlayer gaps. Furthermore, we demonstrate that MXenes with high free carrier density show promise as polarizers and tunable electromagnetic interference shields in the THz range.

O 89.5 Thu 16:45 A 053

UV-to-IR Broadband Ellipsometry Characterization of Spray-Coated MXenes — •Andreas Furchner[1], Tetiana Hryhorchuk[2], Yury Gogotsi[2], and Tristan Petit[1] — [1]Helmholtz-Zentrum Berlin für Materialien und Energie, Berlin, Germany — [2]Drexel University (Nanomaterials Institute), Philadelphia, USA

The chemical composition of MXenes determines whether they exhibit metal-, semi-metal- or semiconductor-like properties, which is important knowledge regarding optoelectronic applications. We employ broadband ellipsometry to characterize the optical and structural properties of spray-coated MXene layers of different chemical composition on silicon and glass substrates. Measuring from the deep-UV (200 nm) to the mid-infrared (25 μm) provides simultaneous access to the electronic and free-charge-carrier properties of the MXenes, as well as to their vibrational fingerprints. Furthermore, ellipsometry enables the quantification of layer thicknesses, roughnesses and film inhomogeneities. The results are corroborated by Vis microscopy and atomic-force-microscopy (AFM) measurements. The authors acknowledge support from the Federal Ministry of Education and Research in the framework of the project Catlab (03EW0015A/B) and funding from the U.S. National Science Foundation (Grant Number CHE-2318105, M-STAR CCI).

O 89.6 Thu 17:00 A 053

2D to 3D weak localization dimensional crossover in $Ti_3C_2T_x$ MXene induced by thickness and defect engineering — •Sophia Tangui[1], Simon Hurand[1], Lola Loupias[2], Stéphane Célérier[2], Ayoub Benmoumen[1,3], Philippe Moreau[3], Marie-Laure David[1], and Vincent Mauchamp[1] — [1]Université de Poitiers, ISAE-ENSMA, CNRS, PPRIME, Poitiers France — [2]Université de Poitiers, CNRS, IC2MP, Poitiers, France — [3]Nantes Université, CNRS, IMN, Nantes, France

Due to their hydrophilic properties and very good metallic electrical behavior, MXenes are promising materials for numerous applications, including transparent conductive thin films. Therefore, there is a need to unravel the transport mechanism involved in MXene multilayers. Although weak localization (WL) has been proposed as the dominating low-temperature transport mechanism in thin films, there have been however few attempts to model quantitatively temperature and magnetic field dependent resistivity measurements.

In this talk, we will focus on the dimensionality of the low-temperature transport mechanisms in spin coated thin films elaborated with the most-studied and metallic $Ti_3C_2T_x$ MXene. The influence of the thin film thickness on one hand and of defects introduced by ion irradiation at different fluences on the other hand is studied by low temperature and magnetic field dependant resistivity measurements. The data are analyzed in the framework of both 2D and 3D models : we will demonstrate a non-trivial evolution between the two behaviors and discuss the validity of both models.

O 89.7 Thu 17:15 A 053

Non-Covalent Functionalized Schottky Interface at Ti3C2Tx/c-Si Van der Waals Heterojunction — •Eloi Ros Costals, Sergio Giraldo, Marcel Placidi, Cristobal Voz, Joaquim Puigdollers, Edgardo Saucedo, Zacharie Jehl Li Kao, and Kunal Tiwari — Electronics Engineering Department, Polytechnic University of Catalunya (UPC), Barcelona Spain

Synergistic interaction between 2D materials and organic molecules presents an additional dimension for tuning their intrinsic properties. Herein, we aim to finely tune the work function of 2D Ti3C2Tx MXene by introducing ultrathin interlayers of organic dipoles (O.D.) with a defined dipole moment value. Interface engineering is achieved through the inclusion of poly(ethylene)amine (PEI 0.1%) and third generation poly(amido-)amine (PAMAM G3), between the Ti3C2TX and c-Si. Charge transport properties of the fabricated Schottky diodes with a structure of c-Si/O.D./Ti3C2TX were evaluated through systematic analysis of the I-V and C-V characteristics. Our investigations reveal that diodes featuring O.D. as interlayers exhibit substantially reduced reverse saturation current density (J0) and enhanced built-in potential (Vbi). We also report a significant reduction in the work function value of Ti3C2Tx from 5.8 eV to 4.2 eV for Ti3C2Tx/PEI 0.1% and 3.3 eV for Ti3C2Tx/PAMAM-G3 heterostructures. On the basis of inferences drawn from photoemission spectroscopy we ascribe this to formation of oriented interfacial dipoles at the Ti3C2Tx/O.D. interface. Our study introduces an innovative approach for precisely controlling the work function of Ti3C2Tx through the incorporation of O.D.

O 90: 2D Materials VI: Growth, Structure and Substrate Interaction

Time: Thursday 15:00–18:00 Location: MA 005

O 90.1 Thu 15:00 MA 005

Growth of aligned and twisted hexagonal boron nitride on Ir(110) — •Thomas Michely, Jason Bergelt, Affan Safeer, Alexander Bäder, Tobias Hartl, and Jeison Fischer — II. Physikalisches Institut, Universität zu Köln, Köln, Germany

The growth of monolayer hexagonal boron nitride (h-BN) on Ir(110) through low-pressure chemical vapor deposition is investigated using low energy electron diffraction and scanning tunneling microscopy. Growth of aligned h-BN on Ir(110) requires a growth temperature of 1500 K, whereas lower growth temperatures result in coexistence of aligned h-BN with twisted h-BN. The presence of the h-BN overlayer suppresses the formation of the nanofaceted ridge pattern known from clean Ir(110). Instead, we observe the formation of a $(1 \times n)$ reconstruction, with n such that the missing rows are in registry with the h-BN/Ir(110) moiré pattern. Quantitative moiré analysis yields a precise determination of the moiré periodicity and the h-BN lattice parameter on Ir(110).

O 90.2 Thu 15:15 MA 005

Incommensurability and negative thermal expansion of single layer hexagonal boron nitride — •Marko Kriegel[1], Karim Omambac[1], Steffen Franzka[2], Frank Meyer zu Heringdorf[1,2], and Michael Horn-von Hoegen[1] — [1]Faculty of Physics and Center for Nanointegration Duisburg-Essen (CENIDE), University of Duisburg-Essen, Lotharstr. 1, 47057 Duisburg, Germany — [2]Interdisciplinary Center for Analytics on the Nanoscale (ICAN), Carl-Benz-Str. 199, 47057 Duisburg, Germany

The emerging field of straintronics, i.e., the control and utilization of the strain state of a 2D material, is of great importance for their technological development, specifically in view of their future incorporation into van der Waals heterostructures. To gain fundamental insights into structural peculiarities of two-dimensional systems, single layer hexagonal boron nitride (hBN) grown on Ir(111) by chemical vapor deposition was used as a prototypical model system: High-resolution reciprocal space mapping by SPA-LEED reveals the incommensurate nature of the material system by measuring the hBN in-plane lattice parameter, facilitated by the moiré magnification effect. In a growth temperature (T_g) regime of 700-1150 °C an average lattice parameter of 2.496±0.006 was found. Eventually, careful disentanglement of the hBN's and substrate's behavior for rising T_g allowed the determination of a negative thermal expansion coefficient of $\alpha_\mathrm{hBN} = -2.4 \pm 1.2 \times 10^{-6}\,\mathrm{K}^{-1}$.[1]

[1] *M. Kriegel *et al.* *Appl. Surf. Sci.* **624** (2023) 157156

O 90.3 Thu 15:30 MA 005

Growth of high quality hexagonal boron nitride (*h*-BN) on surfaces of transition metals — Adrian Hemmi[1], •Ari Paavo Seitsonen[2], Michael S Altman[3], Marcella Iannuzzi[4], Thomas

Greber[1], and Huanyao Cun[1] — [1]Physik-Institut der Universität Zürich — [2]Département de Chimie, École Normale Supérieure, Paris — [3]Department of Physics, Hong Kong Unversity of Science and Technology — [4]Chemie-Institut der Universität Zürich

The production of high-quality hexagonal boron nitride (*h*-BN) is essential for the ultimate performance of two dimensional (2D) materials-based devices, since it is the key 2D encapsulation material. We are working on the optimisation of the procedures to grow *h*-BN on surface of transition metals. Our recent achievements include the enhanced quality of the *h*-BN on Rh(111) via 2D distillation [*ACS Nano* **15**, 1351-1357 (2021)], and a decisive guideline for fabricating high-quality *h*-BN on Pt(111) [*Small* **18**, 2205184 (2022)]. We have found that it is crucial to exclude carbon from the *h*-BN related process, otherwise carbon prevails over boron and nitrogen due to its larger binding energy, thereupon forming graphene on metals after high-temperature annealing. We introduce the pyrolysis temperature T_P as an important quality indicator for *h*-BN on transition metals.

In order to understand better the underlying physical trends, we have performed systematic density functional theory calculations on 12 different hexagonally oriented transition metals [*Nanoscale Advances*, in Press; DOI: 10.1039/D3NA00472D]. We will discuss the experimental findings in light of the computed results.

O 90.4 Thu 15:45 MA 005

Formation of Europium-Ruthenium surface compound and protection of Eu below hexagonal boron nitride (h-BN) — •Alaa Mohammed Idris Bakhit[1,3] and Frederik Schiller[1,2] — [1]Centro de Física de Materiales CSIC-UPV/EHU-Materials Physics Center, E-20018 San Sebastián, Spain — [2]Donostia International Physics Center, E-20018 Donostia-San Sebastián, Spain — [3]Departamento de Física Aplicada I, Universidad del País Vasco UPV/EHU, E-20018 San Sebastián, Spain

We investigate the structural and electronic properties of Europium (Eu) intercalated beneath a layer of hexagonal Boron nitride (h-BN) atop of transition metal Ruthenium Ru(0001) substrate. The hBN monolayer was grown by the Chemical Vapor Deposition process on clean Ru(0001). To explore the structural aspects, we employed Low-energy electron diffraction (LEED). Our observations from LEED revealed c(4×2) and (2×2) superstructures emerging upon Eu intercalation, varying based on the Eu coverage. Simultaneously, the electronic structure was investigated by X-ray photoelectron spectroscopy (XPS) and angle-resolved photoemission measurements (ARPES). Notably, XPS analysis post-Eu intercalation indicated the presence of Eu in a divalent valence state. Furthermore, we studied the possible protection of Eu by the h-BN layer on the Ru(0001) substrate. We observe that partially Eu can be protected but full protection was incomplete probably due to defects and h-BN growth boundaries which facilitated Eu oxidation.

O 90.5 Thu 16:00 MA 005

Real-Time Multiscale Monitoring and Tailoring of Graphene Growth on Liquid Copper — Valentina Rein[1], Mehdi Saedi[2], Maciej Jankowski[1], Anastasios Manikas[3], Francesco La Porta[1], Christos Tsakonas[3], Costas Galiotis[3], Gilles Renaud[4], Oleg Konovalov[1], and •Irene Groot[2] — [1]ESRF — [2]Leiden University — [3]University of Patras — [4]CEA

The synthesis of large, defect-free two-dimensional materials such as graphene is a major challenge toward industrial applications. Chemical vapor deposition on liquid metal catalysts is a recently developed process for the fast synthesis of high-quality single crystals of graphene. However, up to now, the lack of in situ techniques enabling direct feedback on the growth has limited our understanding of the process dynamics and primarily led to empirical growth recipes. Thus, an in situ multi-scale monitoring of the graphene structure, coupled with a real-time control of the growth parameters, is necessary for efficient synthesis. Here we report real-time monitoring of graphene growth on liquid copper (at 1370 K under atmospheric pressure conditions) via four complementary in situ methods: synchrotron X-ray diffraction and reflectivity, Raman spectroscopy, and radiation-mode optical microscopy. This has allowed us to control graphene growth parameters such as shape, dispersion, and the hexagonal supra-organization with very high accuracy. The presented results have far-reaching consequences for studying and tailoring 2D material formation processes on liquid metals during chemical vapor deposition.

O 90.6 Thu 16:15 MA 005

Imaging of topological defects in graphene grown by bottom-up synthesis — •Benedikt P. Klein[1,2,3], Matthew A. Stoodley[1,2], Luke A. Rochford[1,6], Joel Deyerling[4], David Hopkinson[1], Fulden Eratam[1], Tien-Lin Lee[1], Sam Sullivan-Allsop[7], Sarah J. Haigh[7], Roman Gorbachev[7], Christopher Allen[1,5], Wilhelm Auwärter[4], Reinhard J. Maurer[2], and David A. Duncan[1] — [1]Diamond Light Source, Didcot, UK — [2]University of Warwick, Coventry, UK — [3]Korea Basic Science Institute, Daejeon, ROK — [4]Technical University Munich, Germany — [5]University of Oxford, UK — [6]University of Cambridge, UK — [7]University of Manchester, UK

Introducing defects into graphene often requires post processing, like ion sputtering or plasma etching. We present a bottom-up synthesis method yielding graphene with incorporated topological defects on a copper substrate. In the chemical vapour deposition process we use an aromatic precursor that contains the same structural elements as the desired defect. During the growth of the graphene lattice, the topology of the precursor is partly retained and topological defects are formed in the carbonaceous network. In addition to the spectroscopic characterisation presented in the separate companion talk, we quantified the defect concentration using the direct imaging techniques of nc-AFM and TEM. Our results show that the ratio of ideal to defective graphene can be controlled by varying the substrate temperature during the growth process.

O 90.7 Thu 16:30 MA 005

Spectroscopic study of topological defects in graphene grown by bottom-up synthesis — Benedikt P Klein[1,2], Matthew A. Stoodley[1,2], Luke A. Rochford[1], Dylan B. Morgan[2], Michael Clarke[3], Alexander Generalov[4], Alexei Preobrajenski[4], Leon B. S. Williams[1], Tien-Lin Lee[1], Alex Saywell[3], Reinhard J. Maurer[2], and •David A. Duncan[1] — [1]Diamond Light Source, Didcot, UK — [2]University of Warwick, Coventry, UK — [3]University of Nottingham, UK — [4]MAX IV, Lund, Sweden

Introducing defects into graphene often requires post-processing, e.g. by ion sputtering or plasma etching. We present a bottom-up synthesis method yielding graphene with incorporated topological defects on a copper substrate. In the chemical vapour deposition process we use an aromatic precursor that contains the same structural elements as the desired defect. During the growth of the graphene lattice, the topology of the precursor is partly retained and topological defects are formed in the carbonaceous network. In addition to the direct imaging presented in the separate companion talk, we spectroscopically studied the defective graphene using the techniques of normal incidence X-ray standing waves (NIXSW), near edge X-ray absorption fine structure (NEXAFS) and X-ray photoelectron spectroscopy (XPS) to elucidate the structural and chemical changes induced by varying defect concentration. Structurally, our results indicate that the adsorption height varies inversely with the defect concentration.

O 90.8 Thu 16:45 MA 005

Operando Characterization and Molecular Simulations Reveal the Growth Kinetics of Graphene on Liquid Copper — Valentina Rein[2], •Hao Gao[1], Hendrik H. Heenen[1], Oleg V. Konovalov[2], Karsten Reuter[1], and Maciej Jankowski[2] — [1]Fritz-Haber-Institut der MPG, Berlin — [2]ESRF, Grenoble, France

In recent years, liquid metal catalysts have emerged as a compelling choice for the controllable, large-scale, and high-quality synthesis of two-dimensional materials. At present, there is little mechanistic understanding of the intricate catalytic process and its governing factors. In a combined experimental and computational study, we investigate the kinetics of graphene growth during chemical vapor deposition on a liquid copper catalyst. By monitoring the growing graphene flakes in real time using in situ optical microscopy, we explore the growth morphology and kinetics over a wide range of CH_4-to-H_2 pressure ratios and deposition temperatures. Constant growth rates of the flakes' radius indicate a growth mode limited by precursor attachment, whereas methane-flux-dependent flake shapes point to limited precursor availability. Large-scale free energy simulations enabled by an efficient machine-learning potential trained to density-functional theory data provide quantitative barriers for key atomic-scale growth processes. The experimental and theoretical data can be consistently combined into a microkinetic model that reveals a mixed growth kinetics that is controlled by both precursor availability and attachment. Key mechanistic aspects that explain improved graphene quality are a largely suppressed carbon dimer attachment and a self-healing mechanism.

O 90.9 Thu 17:00 MA 005

Structural and electronic properties of two-dimensional single-layer HfS_2 on Au(111) — •Monika Schied[1], Paolo Lacovig[1], Marco Bianchi[2], Philip Hofmann[2], and Silvano Lizzit[1] — [1]Elettra Sincrotrone Trieste — [2]Department of Physics and Astronomy, Aarhus University

HfS_2 is a promising 2D material for low-power semiconductor devices due to its predicted high electron mobility and low contact resistance for n-type carrier transport. For actual applications, layers with excellent structural and electronic properties are needed. However, films with the necessary quality are only available from exfoliation, which is neither scalable nor very reproducible and only few experimental studies on a single-layer (SL) of HfS_2 have been performed so far. In analogy to the growth of high-quality SL transition metal dichalcogenides such as MoS_2 and WS_2 [1,2] we have epitaxially grown an ordered layer of HfS_2 on Au(111). Monitoring the S 2p and Hf 4f core levels in real time by fast X-ray photoelectron spectroscopy allows the fine-tuning of the relevant parameters – such as the dosing rate and temperature – during the growth. The characterization by X-ray photoelectron diffraction, scanning tunnelling microscopy, and low-energy electron diffraction gives insight into the crystal structure of the film grown in this way. In addition, the electronic structure is investigated by angle-resolved photoemission spectroscopy.

[1] Bana, H., et.al., 2D Mater. 5 035012 (2018)
[2] Bignardi, L. et.al., Phys. Rev. Mat. 3, 014003, (2019)

O 90.10 Thu 17:15 MA 005

Na-doped Borophene/Ir(111) — •Sena Tömekce, Marc G. Cuxart, Knud Seufert, and Willi Auwärter — Physics Department E20, TUM School of Natural Sciences, Technical University of Munich, Garching, Germany

Alkali metal doping has been shown to be an effective strategy to tune the electronic properties of 2D materials, due to their low ionization energy [1,2]. Borophenes, 2D layers of covalently bonded boron atoms, combine many interesting properties such as polymorphism, anisotropy and metallicity, making them promising for various applications [3]. Particularly, when doped by alkali metals, free-standing borophenes have been predicted to be excellent materials for Li/Na ion batteries, offering high charge storage capacity, low diffusion barrier and maintaining metallic conductivity [4,5]. Here, we report on the high-resolution LT-STM (low temperature scanning tunneling microscopy) and XPS (X-ray photoelectron spectroscopy) characterization of Na-doped $\chi 6$ borophene/Ir(111). We demonstrate that Na atoms adsorb on borophene without forming clusters, after room temperature deposition. The Na adsorption sites on the borophene lattice were investigated. XPS reveals a binding energy shift of the B 1s core level upon increasing Na coverage, implying charge transfer.

[1] J. Cai et al., Phys. Rev. B. 2018, 98, 195443
[2] D. Marchiani et al., Nano Lett. 2023, 23, 170-176
[3] A. J. Mannix et al., Nat. Nanotechnology. 2018, 13, 444-450
[4] X. Zhang et al., Nanoscale. 2016, 8, 15340-15347
[5] J. Li et al., J. Electrochem. Soc. 2020, 167, 090527

O 90.11 Thu 17:30 MA 005

A buckled honeycomb lattice of Fe atoms grown on a clean Be(0001) surface — •Stefan Krause, Hermann Osterhage, Karoline Oetker, Radek Dao, and Roland Wiesendanger — Department of Physics, University of Hamburg, Germany

The Be(0001) surface is considered to be an ideal model system to host a 2D electron gas with pronounced electron-electron and electron-phonon interactions that are decoupled from the bulk [1,2]. Consequently, growing magnetic adlayers on Be(0001) raises expectations for the emergence of novel electronic and magnetic phases that result from the interactions between the 2D electron gas and the magnetic atoms. Here we report on our experimental studies of the epitaxial growth of Fe on a clean Be(0001) surface using scanning tunneling microscopy at low temperature. Individual Fe atoms are found to replace Be atoms in the topmost Be(0001) surface layer, driven by an atomic exchange. Increasing the Fe coverage results in the self-terminated growth of ultrathin films consisting of atomically well-ordered patches with a p(2×2) superstructure. Based on the atomic structure of the $FeBe_2$ bulk alloy we develop an atomistic growth model, where the dosing of Fe transforms the topmost Be layer into a Kagome lattice that supports the formation of a 2D buckled honeycomb lattice of Fe atoms. The experimental results will be presented and discussed in terms of the atomic lattice structure and spectroscopic characteristics in comparison with the clean Be(0001) surface.

[1] P. T. Sprunger *et al.*, Science **275**, 1764 (1997).
[2] H.Osterhage *et al.*, Phys. Rev. B **103**, 155428 (2021).

O 90.12 Thu 17:45 MA 005

Growth and structure of Chromium sulfide: a new magnetic 2D material — •Affan Safeer[1], Mahdi Ghorbani Asl[2], Wouter Jolie[1], Hannu Pekka Komsa[3], Arkady V. Krasheninnikov[2], Thomas Michely[1], and Jeison Fischer[1] — [1]Universität zu Köln, Köln, Germany — [2]Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany — [3]University of Oulu, Oulu, Finland

We combine scanning tunneling microscopy (STM), low-energy electrons diffraction (LEED) and density functional theory (DFT) calculations to investigate growth, structure and properties of chromium sulfides fabricated by molecular beam epitaxy on graphene. The thinnest chromium sulfide formed has threefold symmetry, an in-plane lattice parameter of 0.340 +/- 0.002 nm, apparent STM height of 0.78-0.87 nm, and 2 Cr atoms per unit cell. Upon annealing of the thinnest, phase pure layers, compounds with same symmetry and lattice parameter, but with increased height in steps of 0.24 nm are formed. In combination with DFT calculations we find a five-layer stack S-Cr-S-Cr-S, namely Cr2S3, to yield excellent agreement with the experimental observations. Systematic variation of the stacking sequence in the DFT calculations of five- layer stacks makes plain, that Cr2S3 grows in the NiAs-structure. Thicker layer, as observed after annealing, are formed by covalent growth of multiples of additional CrS layers. Spin-polarized STM of Cr2S3 at 1.7 K displays hysteresis of the differential conductance signal as a function of out-of-plane magnetic field. This hysteresis is consistent with a ferromagnetic response of the top Cr-layer and antiferromagnetic alignment between Cr layers in Cr2S3.

O 91: Scanning Probe Microscopy: Light Matter Interaction at Atomic Scales III

Time: Thursday 15:00–17:45 Location: MA 041

Topical Talk O 91.1 Thu 15:00 MA 041

SPM on the path to direct nano-optical measurements — •Martin Svec — Institute of Physics, CAS, CZ — Institute of Organic Chemistry and Biochemistry, CAS, CZ

Exploration of essential photophysics at the level of individual molecules and atoms requires highly specialized optical spectroscopies that work at the very limit of instrument sensitivity or have to use plasmonic nanostructures - in order to overcome the fundamental resolution limits achievable with visible and infrared light. Recent developments emerging in the field of Scanning Probe Microscopy (SPM) bring the unique opportunity to pursue intriguing, often hard-to-access interactions between light and matter with plasmonic tips at submolecular scale. The modes of operation can be switched between the electroluminescence, photoluminescence and tip-enhanced Raman spectroscopies, and applied to probe the eigenmodes, charges, vibronics and temporal evolution of the transient states (TCSPC) in molecular emitters at nanoscopic scales.

References

[1] ACS Nano 16, 1082 (2022)
[2] Nature Comm. 13, 6008 (2022)
[3] arXiv:2310.12546 (2023)
[4] arXiv:2309.04416 (2023)

O 91.2 Thu 15:30 MA 041

Triplet emitter electroluminescence from Pd-octaethylporphyrin (PdOEP) in a scanning tunneling microscope (STM) — Li-Qing Zheng[1], Fábio Costa[1,2], Abhishek Grewal[1], Anna Rosławska[1], •Klaus Kuhnke[1], and Klaus Kern[1,3] — [1]Max-Planck Institut für Festkörperforschung, 70569 Stuttgart, Germany — [2]Gleb Wataghin Institute of Physics, UNICAMP, Campinas 13083-859, Brazil — [3]Institut de Physique, École Polytechnique Fédéral de Lausanne, 1015 Lausanne, Switzerland

Triplet emitters produce significant emission (phosphorescence) even below their lowest singlet transition (fluorescence) due to spin-orbit coupling by a heavy metal center. While this property makes them interesting for OLEDs, the study of their luminescence at the single molecule level in high resolution STM is challenging.

We investigate isolated PdOEP molecules decoupled from Ag(100) and Ag(111) by an ultrathin NaCl layer. Singlet and triplet emission lines are observed in the STM at visible wavelengths, only about $100nm$ apart from each other. The singlet S_1 state of PdOEP emits photons even when the energy of one tunneling electron is lower than the emitted S_1 photon energy. The mechanism requires a relay (or shelving) state in which energy is stored in the molecule for the interval between two tunneling electrons. Emission is then expected to follow a quadratic current dependence. The S_1 emission, in contrast, exhibits a linear dependence, suggesting a relay state with nanoseconds life time. We discuss possible scenarios for the S_1 overbias emission.

O 91.3 Thu 15:45 MA 041

Tip-enhanced Raman spectroscopy of physisorbed molecular hydrogen — Akitoshi Shiotari[1], Shuyi Liu[2], Heiko Appel[3], Adnan Hammud[1], Toshiki Sugimoto[4], •Jun Yoshinobu[5], Martin Wolf[1], Mariana Rossi[3], and Takashi Kumagai[4] — [1]Fritz-Haber Institute of the Max-Planck Society, Berlin, Germany — [2]Huazhong University of Science and Technology, Wuhan, Republic of China — [3]Max-Planck-Institute for Structure and Dynamics of Matter, Hamburg, Germany — [4]Institute for Molecular Science, Okazaki, Japan — [5]Institute for Solid State Physics, University of Tokyo, Kashiwa, Japan

Hydrogen molecules on metals is an important model system for understanding physisorption. However, characterizing such weakly adsorbed molecules has been technically challenging. We demonstrate that tip-enhanced Raman spectroscopy (TERS), when combined with scanning tunneling microscopy (STM) at cryogenic temperatures, is capable of detecting physisorbed adsorbates. The rotational and vibrational modes of hydrogen molecules on a Ag(111) surface are characterized using TERS, revealing the nature of the nearly-free rotor. Additionally, we find that the electromagnetic mechanism predominantly contributes to the enhancement of Raman scattering, thus enabling sensitive detection and providing local information on the physisorption system.

O 91.4 Thu 16:00 MA 041

Tip-enhanced Raman spectroscopy in plasmonic nanocavities: enhancement factors and applications — •Borja Cirera[1], Martin Wolf[2], and Takashi Kumagai[3] — [1]Material Science Institute of Madrid, Madrid, Spain — [2]Fritz Haber Insitute, Berlin, Germany — [3]Institute for Molecular Science, Okazaki, Japan

Tip Enhanced Raman Spectroscopy (TERS) in plasmonic nanocavities has reached subnanometer resolution, visualizing chemical heterogeneities and vibrations of adsorbates in the real space. The capabilities of TERS at the atomic scale are a great complement to the existing scanning probe microscopy (SPM) techniques to properly investigate the local physico-chemical properties of the adsorbates. The required Raman sensitivity generally relies on atomically-confined electromagnetic fields in (sub-)nanoscale gaps. Here, the latest results of a single C60 between a silver tip and various metallic and semiconductor substrates are presented. These model systems allow us to discuss the diverse factors contributing to the observed intensity in single molecule junctions, finding a drastic increase in the signal upon molecular point contact (MPC) formation. The resulting exceptionally chemical sensitivity is operative even for weak and non-plasmonic substrates such as Pt(111) and Si(111). These results open the possibility to probe adsorbates on catalytically relevant substrates and a wide range semiconductors, increasing the versatility of TERS beyond coinage metals. The simultaneous access to Stokes and anti-Stokes bands can also be used to investigate heating-cooling processes in non-equilibrium quantum transport systems, of great relevance in molecular electronics.

O 91.5 Thu 16:15 MA 041

Activating the fluorescence of a Ni(II) complex by energy transfer — Tzu-Chao Hung[1,2], Yokari Godinez-Loyola[3], Manuel Steinbrecher[1], Brian Kiraly[1], Alexander A. Khajetoorians[1], Nikos L. Doltsinis[3], Cristian A. Strassert[3], and •Daniel Wegner[1] — [1]Radboud University, Nijmegen, The Netherlands — [2]University of Regensburg — [3]University of Münster

Luminescence of open-shell 3d metal complexes is often quenched due to ultrafast intersystem crossing (ISC) and cooling into a dark metal-centered excited state. We demonstrate successful activation of fluorescence from individual nickel phthalocyanine (NiPc) molecules in the junction of a scanning tunneling microscope (STM) by resonant energy transfer from other metal phthalocyanines at low temperature. By combining STM, STS, STM-induced luminescence and time-dependent density functional theory, we provide evidence that there is an activation barrier for the ISC, which in most experimental conditions is overcome. We demonstrate that, when placing a donor MPc (M = Zn, Pd, Pt) molecule close to NiPc by means of STM atomic manipulation, resonant energy transfer can excite the acceptor NiPc without overcoming the ISC activation barrier, leading to Q-band fluorescence. This work demonstrates that the thermally activated population of dark metal-centered states can be avoided by a designed local environment at low temperatures paired with a directed molecular excitation into vibrationally cold electronic states.

O 91.6 Thu 16:30 MA 041

Gating single-molecule photoluminescence with electrons — •Anna Rosławska[1,2], Katharina Kaiser[1,3], Michelangelo Romeo[1], Fabrice Scheurer[1], and Guillaume Schull[1] — [1]Université de Strasbourg, CNRS, IPCMS, UMR 7504, F-67000 Strasbourg, France — [2]Max-Planck-Institut für Festkörperforschung, D-70569, Stuttgart, Germany — [3]IV. Physikalisches Institut, Georg-August-Universität Göttingen, Friedrich-Hund-Platz 1, 37077 Göttingen

The ability to switch an optically-active molecule from a bright to a dark state by modifying its redox state is one of the most striking modifications of optical properties of a chromophore. Here, we show that this mechanism can be applied to a single molecule located in the double barrier tunneling junction of a scanning tunneling microscope (STM). We achieve nearly full tip-enhanced photoluminescence (TEPL) quenching by controlling the population of a transiently charged state by adjusting the tip-molecule distance, the applied bias voltage, and the in-plane position of the tip with respect to the molecule. This versatile gating of the fluorescence yield of a single molecule with sub-nm precision is explained using a straightforward many-body description of our system.

O 91.7 Thu 16:45 MA 041

Electrically driven cascaded photon-emission in a single molecule — •Katharina Kaiser[1,2], Anna Rosławska[1,3], Michelangelo Romeo[1], Fabrice Scheurer[1], Tomáš Neuman[4], and Guillaume Schull[1] — [1]CNRS, Institut de Physique et Chimie des Matériaux de Strasbourg, France — [2]IV. Physikalisches Institut, Georg-August-Universität Göttingen, Germany — [3]MPI für Festkörperforschung, Stuttgart, Germany — [4]Institute of Physics of the Czech Academy of Sciences, Prague, Czechia

In STM-induced luminescence (STML) of single molecules, the intricate interplay between charging dynamics and excited state formation and decay can be accessed by studying correlations between consecutively emitted photons. This not only provides access to the time constants of the processes involved, but also sheds light on the quantum mechanical nature of a single molecule emitter. Until now, such studies were mainly focused on one transition only. For molecules adsorbed on thin insulating films, however, there are many accessible states, depending on the STM setpoint. Taking advantage of this, we investigate the interplay between the exciton and trion formation and decay in individual ZnPc molecules. We show that, upon trion formation, we can drive the correlated emission of photons from two different excited states within one molecule. In addition, by tuning the rate at which the molecule is charged we can control the population dynamics of the states involved. This allows us to disentangle which states play a role in the formation of excited states in a molecule in STML, and provides a direct means to manipulate excited state dynamics.

O 91.8 Thu 17:00 MA 041

Statistical properties of light emission in current-driven single-molecule STM-junctions — •Andrés Bejarano[1,2], Rémi Avriller[2], Thomas Frederiksen[1,3], and Fabio Pistolesi[2] — [1]Donostia International Physics Center (DIPC), E-20018 Donostia-San sebastián, Spain — [2]Univ. Bordeaux, CNRS, LOMA, F-33405 Talence, France — [3]Ikerbasque, Basque Foundation for Science, E-48013 Bilbao, Spain

The atomic resolution of the scanning tunneling microscope (STM) enables fluorescence on the scale of single molecules. Recent experiments demonstrate the change from a broad plasmonic resonance to a sharp peak in the photon emission spectrum, by moving the tip laterally from the bare substrate towards the molecule [1]. These systems are of particular interest to the quantum cryptography community because they have been reported to emit non-classical light (antibunching) [2, 3]. We propose a microscopic model based on quantum master equation approach for the reduced density matrix of the central system. In particular, we focus on the description of the emission spectrum, conductivity and photon coherence. Additionally, by using full counting statistics, we calculate the Fano factor and correlations between emission and currents. The model provides a simple framework to explain the features observed experimentally in the photon spectrum and the electronic conductance. [1] B. Doppagne et al., Science 361, 251 (2018) [2] P. Merino et al., Nat. Commun. 6, 8461 (2015) [3] L. Zhang et al., Nat. Commun. 8, 580 (2017)

O 91.9 Thu 17:15 MA 041

Vibronically coupled excited states of asymmetric phthalocyanines from STM luminescence spectroscopy — •Sofia Canola[1], Kirill Vasilev[2], Fabrice Scheurer[2], Alex Boeglin[2], Fanny Lotthammer[3], Frederic Cherioux[3], Tomas Neuman[1], and Guillaume Schull[2] — [1]Institute of Physics of the Czech Academy of Sciences, Cukrovarnicka 10, 16200 Praha, Czech Republic. — [2]Universite de Strasbourg, CNRS, IPCMS, UMR 7504, F-67000 Strasbourg, France. — [3]Universite de Franche-Comte, CNRS, FEMTO-ST, F-25000 Besancon, France.

Interactions between molecular excited states, mediated by vibrations, rule the internal dynamic of the molecular electronic structure following excitation. Information on these interactions can be accessed via the vibronic features of the spectra that arise from different mechanisms of coupling between the electronic excitation and molecular vibrations (e.g. Franck-Condon or Herzberg-Teller). In this presentation, I will discuss a systematic experimental and computational study of the vibronic properties for a series of related zinc (II) phthalocyanine derivatives employing electroluminescence in a scanning tunneling microscope (STML). The optical properties of phthalocyanines have been widely studied due their biological and technological relevance, but many fundamental aspects are still to be clarified. From high-resolution vibronic spectra and the associated spatially resolved light emission maps, we can selectively probe different mechanisms of vibronic coupling and access novel microscopic details otherwise not accessible to conventional spectroscopy.

O 91.10 Thu 17:30 MA 041

Resonant TERS of a Single-Molecule Kondo System — •Rodrigo Cezar de Campos Ferreira[1], Amandeep Sagwal[1,2], Jiří Doležal[1,3], Sofia Sofia[1], Pablo Merino[4], Tomáš Neuman[1], and Martin Švec[1,5] — [1]Institute of Physics, Czech Academy of Sciences; Czech Republic — [2]Faculty of Mathematics and Physics, Charles University; Czech Republic — [3]Institute of Physics, École Polytechnique Fédérale de Lausanne; Switzerland — [4]Instituto de Ciencia de Materiales de Madrid; Spain — [5]Institute of Organic Chemistry and Biochemistry, Czech Academy of Sciences; Czech Republic

Single-molecule TERS under UHV and cryogenic conditions enables exploration of the relations between the adsorption geometry, electronic state, and vibrational fingerprints[1,2]. Here we precisely lift a PTCDA molecule from a metal surface to bring it into an open-shell spin one-half anionic state. We reveal a correlation between the Kondo resonance in the differential conductance spectroscopy and concurrent characteristic changes captured by the TERS measurements. Through the investigation of various adsorbed and tip-contacted PTCDA scenarios, we infer that the Raman scattering on the suspended PTCDA is resonant with a higher excited state. Theoretical simulation of the vibrational spectra enables a precise assignment of the individual TERS peaks to high-symmetry Ag modes, including the fingerprints of the observed spin state[3]. [1] Y. Zhang et. al. National Science Review, 6, 1169-1175 (2019).[2] R. Jaculbia et. al. Nat. Nanotechnol. 15, 105-110 (2020).[3] R.C.C. Ferreira et. al. arXiv:2310.12546 (2023).

O 92: Nanostructures at Surfaces II

Time: Thursday 15:00–17:45 Location: MA 042

O 92.1 Thu 15:00 MA 042

Shape control in 2D molecular nanosheets by tuning anisotropic intermolecular interactions and assembly kinetics — •Maximilian Dreher[1], Pierre Dombrowski[1], Matthias Tripp[2], Niels Münster[2], Ulrich Koert[2], and Gregor Witte[1] — [1]Philipps-University of Marburg, Physics Department — [2]Philipps-University of Marburg, Chemistry Department

The fabrication of organic nanostructures is hardly applicable by traditional lithographic pattering techniques due to the limited stability of molecular materials. In this study, we present a methodology for achieving controlled mesoscopic shapes in 2D molecular nanosheets through kinetic controlled self-organization. Using van-der-Waals bound molecules, we demonstrate precise control over the shape of nanosheets without modification of their intrinsic molecular packing. Our study focuses on anisotropic attractive Coulomb forces between partially fluorinated pentacenes, evaluating their role in promoting the growth of distinctly elongated nanosheets. Notably, we observe anisotropic elongation between nanosheets grown directly and those fabricated through partial desorption of a complete molecular monolayer. Monte Carlo simulations reveal that lateral intermolecular interactions predominantly drive the different kinetics of nanosheet growth and desorption. By comparing the behavior of differently fluorinated molecules experimentally and computationally, we identify design rules for molecules for effective shaping of organic nanostructures.[1]

[1] M. Dreher et al., Nat Commun (2023), 14, 1554

O 92.2 Thu 15:15 MA 042

Fe-coordinated $PcZn(OH)_8$ Frameworks on Ag(111) by Electrospray-controlled Ion Beam Deposition — •Pengfei Zhao[1], Wei Ran[1], Andreas Walz[1], Annette Hüttig[1], Hartmut Schlichting[1], Renhao Dong[2], Xinliang Feng[2], Anthoula C. Papageorgiou[1,3], Joachim Reichert[1], and Johannes V. Barth[1] — [1]Technical University of Munich, Germany — [2]Dresden University of Technology, Germany — [3]National and Kapodistrian University of Athens, Greece

Molecular engineering of two-dimensional metal-organic frameworks (2D-MOFs) has garnered significant interest for their structural and functional tunability.[1] However, challenges persist in preparing model systems suitable for investigations at the sub-nanometer scale in a well-defined environment. Here, we present a versatile approach to fabricate 2D-MOFs from an un-sublimable metal-phthalocyanine complex ($PcZn(OH)_8$) via electrospray-controlled ion beam deposition (ESCIBD) on a coinage metal surface.[2] By scanning tunneling microscopy, we find that upon adsorption on Ag(111), $PcZn(OH)_8$ features close-packed molecular assemblies stabilized by lateral intermolecular interactions. After dosing Fe and annealing at 423 K, $PcZn(OH)_8$ exhibits Fe-coordinated 2D-MOFs. These results confirm that ESCIBD can expand the on-surface synthesis of MOF-based materials to thermolabile, highly functionalized organic compounds.

Reference

[1] H. Zhong, et al., Nat Commun, 2020, 11, 1409.

[2] A. Walz, et al., Anal Chem, 2022, 94, 7767-7778.

O 92.3 Thu 15:30 MA 042

2D Metal-organic frameworks on the topological insulator Bi_2Se_3(0001) surface — •Matthias Blatnik[1], Anna Kurowská[1], Pavel Procházka[1], and Jan Čechal[1,2] — [1]CEITEC, Brno University of Technology, Brno, CZ — [2]IPE, Brno University of Technology, Brno, CZ

Topological insulators (TI) and other exotic quantum matter have recently drawn much attention due to their fascinating electronic properties. 3D TIs show strong spin-orbit splitting and time-reversal symmetry (TRS) leading to topologically protected surface states with linear, Dirac-like dispersion. Heterostructures of TIs with other materials are highly interesting candidate structures for a variety of applications in new quantum devices and thus of considerable interest for quantum computing, but also optoelectronics or spintronics. Periodic arrays of ferromagnetically coupled transition metal (TM) atoms on the surface of a TI are predicted to spontaneously break TRS and induce a band gap opening at zero magnetic field (i.e., a quantum anomalous Hall effect, QAHE). A 2D metal-organic framework (MOF) of spin-coupled TM atoms ordered by the right organic linkers could be such a candidate. Here, we report on the self-assembly of dicyanoanthracene (DCA) molecules on the topological insulator Bi_2Se_3(0001) surface as a first step, and then the first-time realized 2D Fe-DCA MOFs on the same surface. We apply a variety of surface science techniques (e.g. LEEM, STM, LEED) to investigate and characterize the formation of molecular islands from small to monolayer coverages and study the chemical environment and electronic properties by XPS and ARPES.

O 92.4 Thu 15:45 MA 042

Hot electron lifetime of confined image potential state in a metal-coordinated organic porous network — •Martin Mitkov, Lukas Kau, Ralf Hemm, Martin Anstett, Lu Lyu, Martin Aeschlimann, and Benjamin Stadtmüller — University of Kaiserslautern-Landau and Research Center OPTIMAS, Erwin-Schrödinger Str. 46, 67663 Kaiserslautern, Germany

Two-dimensional metal-organic porous networks (2DMOPNs) on noble metal surfaces are highly ordered quantum boxes for confining surface state electrons. For a long time, this approach has mainly been used for the confinement of surface electrons in Shockley-type surface states and has only recently been explored for excited electrons at surfaces[1] , i.e., image potential state (IPS) electrons. In this contribution, we focus on the hot electron dynamics of quantum confinement IPS electrons in a 2D porous network. As a model system, we study the Cu-coordinated T4PT network on the Cu(111) surface for which the quantum confinement leads to a substantial increase in the effective band mass of the IPS electrons[1]. We use time-resolved two-photon photoemission spectroscopy to quantify the fs timescale hot electron dynamics and quasi-particle lifetimes of the confined electrons in the flat IPS bands. Our findings provide insight into the role of the quantum confinement for the hot electron dynamics in the 2DMOPNs.

[1] Lyu, Lu, et al. arXiv preprint arXiv:2307.06814 (2023).

O 92.5 Thu 16:00 MA 042

On-surface Synthesis of Tunnable Dinuclear Organometallic Networks — •Wenchao Zhao, Nan Cao, Mathias O. Senge, Ethem Akturk, Johannes V. Barth, and Biao Yang — Physics Department E20, School of Natural Sciences, Technical University of Munich, 85748 Garching, Germany

Engineering organometallic networks (OMNs) obtains great efforts due to their potential applications in catalysis, optoelectronic devices etc[1]. Dinuclear metal complexes have multifunctional properties with their tunable structure and hence various applications in the field of OLEDs, chemosensors and smart materials[2]. The relatively complex structure makes it difficult to form thin films for advanced devices application. Therefore, on-surface synthesis of dinuclear organometallic networks serves as a powerful strategy to directly obtain the thin functional layers. Herein, with a designed bi-component precursor and O2-mediated treatment, we manage to construct ordered Ag-Ag dinuclear OMNs containing C-Ag-N bridging linkers. Moreover, it is feasible to tune the the thermal stability and electronic property by replacing the dinuclear metal centers via post metal-exchange method, transferring to Cu-Cu dinuclear with C-Cu-N linkages. This advanced strategy will greatly expand the diversified synthesis and application of dinuclear organometallic supramolecular materials. Reference. 1. D. Ecija et al., Acc. Chem. Res. 2018, 51, 365-375. 2. J. Lv et al., Nature 2023, 622, 754-760.

O 92.6 Thu 16:15 MA 042

Tailoring the ferromagnetic surface potential landscape by a templating two-dimensional metal-organic porous network — •Lu Lyu, Martin Anstett, Ka Man Yu, Azadeh Kadkhodazadeh, Martin Aeschlimann, and Benjamin Stadtmüller — Department of Physics and Research Center OPTIMAS, Rheinland-Pfälzische Technische Universität Kaiserslautern-Landau, Erwin-Schrödinger-Straße 46, 67663 Kaiserslautern, Germany.

Two-dimensional metal-organic porous networks (2D-MOPNs) have emerged as versatile nanoarchitectures for tailoring surface electronic and magnetic properties on noble metals. In this context, we demonstrate the manipulation of the potential landscape of a ferromagnetic surface using a 2D-MOPN structure. As a model surface, we use ultrathin cobalt (Co) films grown on Au(111) which exhibit well-ordered triangular dislocation loops. On this ferromagnetic surface, 2,4,6-tris(4-

pyridyl)-1,3,5triazine (T4PT) molecules can be coordinated with the surface Co atoms, forming a large-scale Co-T4PT porous network. The periodicity of the network acts as a template for the ferromagnetic surface potential and guides the subsequent growth of isolated C60 molecules on the network. For larger submonolayer coverage, the structure of the molecular film is further influenced by the intermolecular interactions between C60 molecules. Our findings underline the potential of 2D-MOPN to template the electronic and spin properties of molecular films on magnetic surfaces.

O 92.7 Thu 16:30 MA 042

Isomorphism of Supramolecular vs. Covalent Binding Motifs in Molecules Forming Two-Dimensional Nanopatterns — •David Ari Hofmeister, Lucas Rothe, Sigurd Höger, and Stefan-Sven Jester — Kekulé-Institut für Organische Chemie und Biochemie, Rheinische Friedrich-Wilhelms-Universität Bonn, Gerhard-Domagk-Str. 1, 53121 Bonn, Germany

The isomorphic packing of different molecules is an established concept in crystallography. Established for three-dimensional (3D) crystals, the concept has recently been extended to two-dimensional (2D) supramolecular patterns of organic molecules on a solid surface, such as highly-oriented pyrolytic graphite (HOPG). The formation of a physisorbed 2D nanopattern is driven by noncovalent interactions. Thereby, different types of interactions (*e.g.* hydrogen bonds and van der Waals interactions) determine the packing, while the balance of interactions is crucial for 2D self-assembly. Here, we substitute a noncovalent interaction motif (*i.e.* the hydrogen bond interaction between two carboxylic acids, R-COOH) with a covalent linkage (a butadiynylene unit). N molecules with COOH groups at opposite ends and an n-mer (in which $n - 1$ R-COOH...HOOC-R units are substituted by a butadiynylene unit) pack isomorphously. *In situ* scanning tunneling microscopy at the 1,2,4-trichlorobenzene/HOPG interface provides a submolecularly resolved insight into the nanopatterns formed.

O 92.8 Thu 16:45 MA 042

Dimensional reconstruction of nanoscale grating profile in hybrid approach — •Leonhard Merlin Lohr, Richard Ciesielski, Vinh-Binh Truong, Frank Scholze, and Victor Soltwisch — Physikalisch-Technische Bundesanstalt (PTB), Abbestraße 2-12, 10587 Berlin, Germany

Quality assessment of nanoscale test structures in the semiconductor industry is crucial for the fabrication of devices, such as advanced transistors. The shape of test structures, like nanoscale line gratings, needs to be described with sub-nanometer accuracy. A fast and non-destructive characterization technique is scatterometry, which uses short-wavelength radiation to resolve features of the nanoscale grating. Synchrotron radiation, with its high beam quality probing the structured surface, can be employed to develop the characterization method. A nanoscale grating can be characterized through both soft X-ray scattering and fluorescence. The soft X-ray standing wave field at the structure profile from radiation-surface interaction can be solved using a finite element Maxwell solver and Markov chain Monte Carlo sampling to derive the grating's shape, including uncertainties.

This method is suitable for investigating the potential to enhance reconstruction accuracy by combining complementary measurements in a hybrid measurement scheme. This is illustrated through measurements conducted with a recently presented experimental scattering chamber [1].

[1] R. Ciesielski, L. M. Lohr, A. Fernández Herrero, et al., Rev. Sci. Instrum. 1 Jan 2023; 94 (1): 013904.

O 92.9 Thu 17:00 MA 042

Supramolecular nanopatterns of caltrop-shaped molecular platforms on graphite — •Jakob Gabriel, David Ari Hofmeister, Georgiy Poluektov, Tristan Johannes Keller, Sigurd Höger, and Stefan-Sven Jester — Kekulé-Institut für Organische Chemie und Biochemie, Universität Bonn, Gerhard-Domagk-Str. 1, 53121 Bonn, Germany

Two-dimensional supramolecular nanopatterns of arylene-alkynylenes carrying long alkyl/alkoxy side chains are investigated at the solid/liquid interface on highly oriented pyrolytic graphite (HOPG). Scanning tunneling microscopy provides a submolecularly resolved insight into the nanopatterns formed. In many cases, the exact interdigitation motifs of the side chains are clearly resolved. We have recently reported on molecular spoked wheel architectures with intraannular pillars carrying large entities, such as a perylene monoimide or a fullerene unit, pointing into the volume phase.[1] Here, we focus on investigating a caltrop-shaped tetraphenylmethane derivative with three "foot" units (instead of a ring-shaped platform). Each of the "feet" carries two pairs of $OC_{16}H_{33}$ side chains. We evaluate the abundance of different polymorphs that form, when different concentrations in the supernatant liquid phase as well as different solvents are used (1,2,4-trichlorobenzene and 1-phenyloctane).

[1] G. Poluektov, T. J. Keller, A. Jochemich, A. Krönert, U. Müller, S. Spicher, S. Grimme, S.-S. Jester, S. Höger *Angew. Chem. Int. Ed.* **2021**, *60*, 27264.

O 92.10 Thu 17:15 MA 042

Revisiting the realization of artificial graphene by fullerenes on gold and copper surfaces — •Dmitry Marchenko[1], Maxim Krivenkov[1], Maryam Sajedi[1], Alexander Fedorov[1,2], Jaime Sánchez-Barriga[1], Oliver Clark[1], Emile Rienks[1], Oliver Rader[1], and Andrei Varykhalov[1] — [1]Helmholtz-Zentrum Berlin für Materialien und Energie — [2]IFW Dresden

There has been significant progress in the creation of artificial graphene based on molecular networks. For a layer of C60 molecules on Au(111) and Cu(111) surfaces, graphene-like Dirac cones were reported by angle-resolved photoelectron spectroscopy (ARPES). The explanation given is based on a hexagonal network of hollow sites in the 2D surface electron gas, due to the expulsion potential of adsorbed molecules, with the corresponding formation of an electronic honeycomb structure.

Using ARPES and DFT methods we demonstrate [1,2] that the observed bands are neither Dirac cones nor hybridization states. They are formed by final state photoelectron diffraction on a regularly arranged C60 molecular layer. This effect creates a highly coherent pattern of replicated bands, which mimics Dirac cones.

In addition, we discuss in detail the nature of well-known steeply dispersing states in the Au(111) and Cu(111) electronic structure. Traditionally these states are believed to be bulk sp-bands, but we demonstrate that they are a manifold of 2D surface resonance bands leaking into the bulk.

[1] M. Krivenkov et. al., Nanoscale 14, 9124 (2022).

[2] D. Marchenko et. al., Phys. Rev. B 108, 115155 (2023).

O 92.11 Thu 17:30 MA 042

Supramolecular nanopatterns of pentagonal arylene-alkynylenes on graphite — •Israa Abushawish, Vanessa Bobbe, Christian Krewer, Anna Krönert, Georgiy Poluektov, Kristin Gratzfeld, Sigurd Höger, and Stefan-S. Jester — Kekulé-Institut für Organische Chemie und Biochemie, Bonn, Germany

The physisorption of organic molecules at solid surfaces is an effective way to produce highly ordered two-dimensional (2D) nanoscale functional arrays. Shape-persistent arylene- alkynylenes with flexible alkyl/alkoxy substituents decorate the solid/liquid interface of highly oriented pyrolytic graphite (HOPG) and a solution of the compound of interest in 1,2,4- trichlorobenzene (TCB). The so-formed nanopatterns are imaged *in situ* by scanning tunneling microscopy (STM, with the tip immersed into the solution), providing submolecularly resolved insights into the nanopatterns formed. Recently, a series of macrocycles with 3- to 6-fold symmetry has led to the concept of molecular polygons [1,2]. Here, we report on the influence of symmetry, substitution patterns, and the influence of different corner units (i.e. carbazole, dibenzofurane, and dithiophene [1]) on the exact shapes of the molecular pentagons and their different supramolecular packings.

References: [1] S.-S. Jester, E. Sigmund, S. Hoeger, *J. Am. Chem. Soc* 2011, 133, 29, 11062.

[2] P. Wilhelm, J. Vogelsang, G. Poluektov, N. Schönfelder, T. J. Keller, S.-S. Jester, S. Höger, J. M. Lupton, *Angew. Chem. Int. Ed* . **2017**, *56*, 1234.

O 93: Scanning Probe Techniques: Method Development

Time: Thursday 15:00–18:00 Location: MA 043

O 93.1 Thu 15:00 MA 043

Tip-to-tip approach for multi-probe scanning probe microscopy utilizing long-distance all-optical methods — •Philipp Lindner, Raluca Boltje, Jonas Koch, Stefan Krause, and Roland Wiesendanger — Department of Physics, University of Hamburg, Germany

In multi-probe scanning probe microscopy, scanning electron microscopy (SEM) is conventionally used to support the individual positioning of the probes. However, SEM is incompatible with the application of high magnetic fields, eventually contaminates high-quality sample surfaces, and is limited to close working distances. Here we present the employment of a Maksutov-Cassegrain telescope (MCT) and a high-irradiance focused fiber-optic illumination system to achieve a diffraction-limited optical resolution of 3 micrometers at a working distance greater than 50 cm for three-tip spin-polarized scanning tunneling microscopy experiments at low temperatures and high magnetic fields. The oblique angle between the MCT objective lens and the sample plane, in combination with Airy pattern interference between the probe tips and their mirror images reflected from the specular sample surface, is utilized for the three-dimensional positioning of the probe tips into overlapping scan ranges. In contrast to SEM-based approaches, our method is applicable to compact scanning probe microscopes installed into the confined geometries of a superconducting magnet cryostat.

O 93.2 Thu 15:15 MA 043

Multi-Probe RF Scanning Tunneling Microscopy in UHV, at Low Temperatures, and in Magnetic Fields — •Jonas Koch, Philipp Lindner, Raluca Boltje, Stefan Krause, and Roland Wiesendanger — Department of Physics, University of Hamburg, Germany

We present a three-tip scanning tunneling microscope designed for multi-probe investigations on the atomic scale. It was developed for the operation in an ultra-high vacuum chamber system [1], at temperatures between 1.5 K and 100 K, and in an external magnetic field of up to 3 T. Three independent scanning units are capable of spin-polarized tunneling with atomic spatial and picosecond time resolution. Ultrasharp tunneling tips are brought into overlapping scan ranges. One of the potential applications is the localized generation of high-density surface current at low total currents, thereby minimizing harming effects like Joule heating. We will present first proof-of-concept experiments on the example of a local surface current application, along with respective numerical modelling of the electrical current on the sample surface. [1] J. Friedlein *et al.*, Rev. Sci. Instrum. **90**, 123705 (2019).

O 93.3 Thu 15:30 MA 043

Spin-Polarized Transport at the Atomic Limit — •Markus Leisegang, Patrick Härtl, Jens Kügel, and Matthias Bode — Physikalisches Institut, Experimentelle Physik II, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany

Transport measurements that are sensitive to the band structure of a material require techniques that operate on the length scale of the charge carrier's mean free path. In order to get real space access to charge carrier transport at distances of the mean free path and thus in the ballistic regime, we developed and established the molecular nanoprobe (MONA) technique [1,2].

Hereby, we use a single molecule as a detector for charge carriers, which are injected into the substrate under investigation by the STM tip a few nanometers away from the molecule. The high spatial resolution of MONA combined with the small size of the molecular detector allows for transport paths which can be controlled at the atomic level. In a very recent experiment, we merged the MONA technique with spin-polarized STM to SP-MONA. By using the Rashba-split surface state of the $BiAg_2$ surface as a test sample, we proof that this technique allows to detect spin-polarized transport at the atomic limit [3].
[1] J. Kügel *et al.*, Nano Lett. **17**, 5106 (2017)
[2] M. Leisegang *et al.*, Nano Lett. **18**, 2165–2171 (2018)
[3] P. Härtl *et al.*, arXiv:2303.00393 (2023)

O 93.4 Thu 15:45 MA 043

Autonomous chemical reactions in scanning tunneling microscope — •Nian Wu[1], Markus Aapro[1], Alexander Ilin[2], Robert Drost[1], Joakim Jestilä[1], Zhijie He[2], Peter Liljeroth[1], and Adam S. Foster[1,3] — [1]Applied Physics, Aalto University, Espoo, Finland — [2]Computer Science, Aalto University, Espoo, Finland — [3]WPI Nano Life Science Institute, Kanazawa University, Kanazawa, Japan

Several breakthrough studies have harnessed scanning probe microscopy (SPM) manipulations to control chemical reactions in on-surface molecular synthesis. In general, for scanning tunnelling microscope (STM) manipulations, they are predominantly controlled via parameters of the tip position, pulse voltages and tunneling conductance. However, the selection of proper parameters requires extensive domain knowledge, which is time consuming and not necessarily transferable to new systems. Recent research has allowed the automation of a wide range of challenges in SPM, including image quality assessment, lateral and vertical manipulation. However, the automation for breaking or forming covalent bonds, which is an indispensable step during chemical synthesis is, as yet, unexplored. To address this problem, we build on our deep reinforcement learning approach to automate bromine removal from 5,15-bis(4-bromo-2,6-methyl-phenyl)porphyrin (Br2Me4DPP) through learning manipulation parameters in STM. We further explore the potential of automated STM to then controllably react the resultant fragments into larger molecular structures.

O 93.5 Thu 16:00 MA 043

Single crystal diamond needles as sensor in scanning probe microscopy — •Stefan Schulte[1,2], Sven Just[1,3], Victor I. Kleshch[4], F. Stefan Tautz[1,3], and Ruslan Temirov[1,2] — [1]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, Germany — [2]II. Physikalisches Institut, Universität zu Köln, Cologne, Germany — [3]Fundamentals of Future Information Technology, Jülich Aachen Research Alliance (JARA), Jülich, Germany — [4]Moscow, Russia

It was recently demonstrated that an SPM tip carrying a quantum dot allows for the measurement of electrostatic surface potentials, in technique that is referred to as scanning quantum dot microscopy (SQDM) (Wagner et al., Phys. Rev. Lett., 2015). Here, we report on an attempt to fabricate a SQDM tip with a CVD-grown single crystal diamond needle (Kleshch et al., Phys. Rev. B, 2020; Kleshch et al., Carbon, 2021). Using the micro-manipulator inside a focussed ion beam microscope the diamond needle is attached to a needle sensor. The diamond tip needle sensor is then characterized by performing non-contact AFM and field emission experiments at room temperature UHV conditions. At elevated field emission currents of several hundred nanoamperes, a surface conductive layer forms on the diamond needle, that eventually allows us to observe electron tunneling between the tip and the surface.

O 93.6 Thu 16:15 MA 043

Systematic protocol to prepare SPM tips for Scanning Quantum Dot Microscopy — •Tim Dierker, Paul Laubrock, and Philipp Rahe — Universität Osnabrück

Scanning probe microscopy (SPM) is an established family of techniques for high-resolution measurements of surfaces. One of its variants is scanning quantum dot microscopy (SQDM) [1], a technique that enables the quantitative mapping of the electrostatic potential at the atomic scale [2]. Central for SQDM is the controlled functionalization of the SPM tip with a single molecule that acts as a quantum dot. In this work we perform low-temperature manipulation experiments with 3,4,9,10-perylene-tetracarboxylic-dianhydride (PTCDA) molecules on Ag(111) with a combined scanning tunneling and atomic force microscope system. From the classification of manipulation and pick-up data we develop a routine to isolate single molecules from the edges of PTCDA islands and to attach them to the tip. The experimental observations allow to condense the physical complexity of the tip-molecule system into an instructive flowchart for an efficient preparation of SQDM tips.

[1] C. Wagner et al., PRL 115, 026101 (2015)
[2] C. Wagner et al., Nat. Mater. 18, 853 (2019)

O 93.7 Thu 16:30 MA 043

Van der Waals scanning probe tips — •Abhisek Kole[1,2,4], Tobias Wichmann[1,2,4], Keda Jin[1,2,3], Jia Grace Lu[5], Xiaosheng Yang[6,7], F. Stefan Tautz[1,2,4], Markus Ternes[1,2,3], Jose Martinez Castro[1,2,3], and Felix Lüpke[1,2] — [1]Peter Grünberg Institut

(PGI-3), Forschungszentrum Jülich, 52425 Jülich, Germany — [2]Jülich Aachen Research Alliance, Fundamentals of Future Information Technology, 52425 Jülich, Germany — [3]Institut fur Experimentalphysik II B, RWTH Aachen, 52074 Aachen, Germany — [4]Institut fur Experimentalphysik IV A, RWTH Aachen, 52074 Aachen, Germany — [5]Department of Physics/Electrophysics, University of Southern California, Los Angeles, CA 90089, USA — [6]Wuhan National Laboratory for Optoelectronics and School of Optical and Electronic Information, Huazhong University of Science and Technology, Wuhan 430074, China — [7]Optics Valley Laboratory, Hubei 430074, China

Van der Waals materials are known for their intriguing emergent 2D physics, such as correlated phenomena and topological effects. Here, we report the methodical fabrication of van der Waals scanning tunnelling tips from exfoliated graphite flakes with a graphene-like edge as a scanning tunnelling tip. The principle of STM is based on the quantum mechanical tunneling between the tip and sample, revealing the convoluted underlying electronic structure. We characterize the tip by performing atomically resolved STM of an Ag(111) surface. The tip-sample differential conductance dI/dV reveals direct evidence of tunnelling through a graphene nanoribbon-like zigzag edge state, which we support by tight binding calculations.

O 93.8 Thu 16:45 MA 043

A versatile Peak Force IR variation for correlative nanoscale chemical and mechanical AFM-IR — •Martin Wagner, Qichi Hu, Chunzeng Li, Shuiqing Hu, Chanmin Su, and Peter Dewolf — Bruker Nano Surfaces, Santa Barbara CA 93117, USA

Nanoscale infrared (nano-IR) microscopy enables label-free chemical imaging and spectroscopy at the nanometer scale by combining atomic force microscopy (AFM) with infrared radiation. Over the last years, AFM-IR has been developed with different AFM modes: the original photothermal induced resonance enhanced mode and the recently developed surface sensitive technique are based on contact mode, while Tapping AFM-IR is built on tapping mode [1]. Peak Force Tapping based Peak Force infrared (PFIR) microscopy [2] has lately joined as another AFM-IR mode. All these nano-IR variations inherit the advantages and limitations of their respective AFM base mode. In this work, we focus on the capabilities offered by a PFIR-related approach, and illustrate those with examples on a variety of polymer samples. We discuss (1) the capability to perform simultaneous multimodal imaging collecting both mechanical properties such as elastic modulus together with chemical information, (2) methods to understand and decouple artifacts induced by variations in mechanical properties from the AFM-IR data, and (3) the combination with other AFM-IR techniques such as the surface sensitive mode while, at the same time, minimizing lateral forces to allow one to study soft & fragile samples.

[1] J. Mathurin et al., J. Appl. Phys. 131, 010901 (2022).

[2] L. Wang et al., Chem. Soc. Rev. 51, 5268 (2022).

O 93.9 Thu 17:00 MA 043

Approaching unstable periodic states in dynamic Atomic Force Microscopy — •Lukas Böttcher[1], Hannes Wallner[2], Niklas Kruse[2], Anna Dittus[2], Wolfram Just[2], Ingo Barke[1], Jens Starke[2], and Sylvia Speller[1] — [1]Institute of Physics, University of Rostock — [2]Institute of Mathematics, University of Rostock

Bistable states, caused by the nonlinear tip-sample interaction, are frequently encountered during dynamic Atomic Force Microscopy (AFM) measurements, sometimes leading to characteristic image artifacts. These states are characterized by two stable states, one at low, one at high amplitude, flanking an unstable branch at intermediate amplitudes. This unstable branch is usually not accessible experimentally. Utilizing fast, minimally invasive control schemes we were able to approach such states of instability. The aim is to reveal the whole resonance curve of our cantilever including fold bifurcation points and a cusp, while interacting with the surface. This may bring new insights into AFM as well as in the tip-sample contact.

O 93.10 Thu 17:15 MA 043

Total variation denoising for microscopy images — •Marco Corrias[1,2], Thomas Pock[4], and Cesare Franchini[1,3] — [1]University of Vienna, Faculty of Physics and Center for Computational Materials Science, Vienna, Austria — [2]University of Vienna, Vienna Doctoral School in Physics, Vienna, Austria — [3]University of Bologna, Department of Physics and Astronomy, Bologna, Italy — [4]Institute of Computer Graphics and Vision, Graz University of Technology, Graz, Austria

Experimentally acquired images are affected by the ubiquitous presence of noise, which degrades their quality and hides their features. With the increase of image acquisition rate in recent years, modern denoising solutions have become necessary. This study focuses on microscopy image denoising, specifically those obtained through atomic force microscopy (AFM), scanning tunneling microscopy (STM), scanning electron microscopy (SEM), and scanning transmission electron microscopy (STEM). A total variation (TV)-based workflow is presented to automatically denoise microscopy images with different types of noise, proving that the Huber-ROF and TGV-L1 are effective to accomplish this task. Our results suggest a wider applicability of this method in microscopy, not only restricted to STM, AFM, SEM, and STEM images. The Python code used for this study will be made publicly available as part of the AiSurf package. It is designed to be integrated into experimental workflows for image acquisition or can be used to denoise previously acquired images.

O 93.11 Thu 17:30 MA 043

Image interpretation methods for high-resolution SPM — •Lauri Kurki[1], Niko Oinonen[1,2], and Adam S. Foster[1,3] — [1]Aalto University, Finland — [2]Nanolayers Research Computing Ltd., UK — [3]WPI-NanoLSI, Kanazawa University, Japan

Scanning tunnelling microscopy (STM) and atomic force microscopy (AFM) functionalized with a CO molecule on the probe apex capture sub-molecular level detail of the electronic and physical structures of a sample from different prespectives [1]. However, the produced images are often difficult to interpret. To accelerate the analysis, we propose automated machine learning image interpretation tools to extract sample properties directly from SPM images.

In recent years, there has been rapid development in image analysis methods using machine learning, with particular impact in medical imaging. These concepts have been proven effective also in SPM in general and in particular for extracting sample properties from AFM images [2,3,4]. We build upon these models and show that we can extract atomic positions directly from STM images. We also further explore how the accuracy of these predictions varies with the use of a simultaneous AFM signal. Finally, we establish the limits of the approach in an experimental context by predicting atomic structures from STM images of 2D ice structures.

[1] Cai et al. J. Am. Chem. Soc. 2022, 144, 44, 20227-20231 [2] Alldritt et al., Sci. Adv. 2020; 6 : eaay6913 [3] Carracedo-Cosme et al., Nanomaterials 2021, 11, 1658. [4] Oinonen et al., MRS Bulletin 2022, 47, 895-905

O 93.12 Thu 17:45 MA 043

Automated prediction of three-dimensional molecular structures from Atomic Force Microscopy images — •Joakim S. Jestilä[1], Shuning Cai[1], Niko Oinonen[1], Peter Liljeroth[1], and Adam S. Foster[1,2] — [1]Department of Applied Physics, Aalto University, 00076 Aalto, Espoo, Finland — [2]Nano Life Science Institute (WPI-NanoLSI), Kanazawa University, Kanazawa 920-1192, Japan

Identification of three-dimensional features in surface-adsorbed molecules imaged by Atomic Force Microscopy (AFM) represents a great challenge. While the structures of planar molecules can often be recognised by human users, deviation from planarity contributes to images that are non-intuitive and difficult to interpret, even for experts. Fortunately, neural networks are well-suited for extracting the embedded information in such images. Still, the latter cannot directly determine the placement of atoms that do not contribute to the image contrast, such as atoms eclipsed by those closest to the AFM-tip. In an attempt to access the hidden atoms in the lower layers, we supplement the prediction of the upper atoms with an algorithm that provides candidate structures based on their physical feasibility, evaluated hierarchically in terms of chemical connectivity and the corresponding density functional theory energy. We demonstrate the applicability of the method in a case study of a model system for surface-adsorbed lignocellulosic molecules: 4-nitrophenyl-α/β-D-galacturonide on Au(111).

O 94: Focus Session: Proximity Effects in Epitaxial Graphene I

Proximity-induced correlation effects in low dimensional electron gases are essential for the design of new quantum materials with tailored electronic, magnetic and optical properties. The recent reports on superconductivity in twisted bilayer graphene is an intriguing example. Inherently, proximity coupling often comes along with further effects, e.g. hybridization, charge transfer, etc., so that intended quantum phenomena become easily quenched. Epitaxial graphene (EG) resembles a truly 2D electron gas system and is known for its ability of manifold and flexible functionalization schemes at its vacuum and interface site, e.g. by doping, encapsulation with high-Z materials as well as formation of superlattices. This Focus Session will provide an overview of the current state of research and possible perspectives.

Organizer: Christoph Tegenkamp (TU Chemnitz)

Time: Thursday 15:00–18:00 Location: MA 141

Topical Talk O 94.1 Thu 15:00 MA 141

Tailoring the electronic structure of epitaxial graphene on SiC — •Kathrin Küster — Max-Planck-Institut für Festkörperforschung, Stuttgart, Germany

Inducing correlation effects in low dimensional electron gases by proximity coupling is a powerful concept for the design of new quantum materials with tailored electronic, magnetic and optical properties. The DFG research unit FOR5242 investigates spin-orbit interaction and electronic correlations of 2D electron gas systems using epitaxial graphene grown on SiC substrates. After a brief introduction to the objectives of the research unit, I will present two different projects of our group. (i) By intercalation with lanthanides like Gd [1] and Yb [2,3], graphene can be driven to extreme doping levels where the system deviates from the low-energy limit of massless Dirac fermions. (ii) In contrast, intercalation of graphene with Pb leads to the formation of almost charge neutral graphene. In general intercalation with heavy elements is an interesting research area since those might induce spin-orbit coupling effects and exotic collective phenomena. Besides the influence of the intercalants on the electronic properties of the graphene, also the investigation of the electronic and structural properties of the intercalants is an exciting topic on its own since they often strongly deviate from their 3D counterparts.

[1] S. Link et al. PRB 100, 121407(R) (2019); [2] P. Rosenzweig et al. PRB 100, 035445 (2019), [3] P. Rosenzweig et al. PRL 125, 176403 (2020); [4] B. Matta et al. Phys. Rev. Research 4, 023250 (2022); [5] P. Schädlich et al. Adv. Mater. Interfaces 2300471 (2023).

O 94.2 Thu 15:30 MA 141

Investigating intercalated Lead layers underneath epitaxial graphene — •Philip Schädlich[1,2], Franziska Schölzel[1], Peter Richter[1,2], and Thomas Seyller[1,2] — [1]Technische Universität Chemnitz, Institut für Physik, 09126 Chemnitz — [2]Center for Materials, Architectures and Integration of Nanomembranes (MAIN), 09126 Chemnitz

Intercalation is a promising approach for tailoring the electronic structure of epitaxial graphene on SiC. Beyond that, it enables the formation of otherwise unstable two-dimensional (2D) phases of elements and opens a route to investigate the interplay between the two 2D materials and the substrate [1,2].

We have studied in detail the Pb intercalation process itself, as well as the structure and electronic properties of the 2D Pb layer by means of low-energy electron microscopy and photoelectron spectroscopy (PES). Probing the electronic structure using angle-resolved PES reveals a strict (1×1) periodicity of the intercalated Pb with respect to the substrate. The quasi-freestanding graphene turns out to be effectively screened from the doping influence of the substrate leading to charge-neutrality. In fact, the 2D Pb layer is compensating the spontaneous polarization of the substrate. Our results experimentally quantify the interaction between the 2D Pb layer, the substrate and the graphene layer and demonstrate a first step towards controlling the diversity of 2D Pb phases.

[1] C. Ghosal, Phys. Rev. Lett. 129, 116802 (2022), [2] P. Rosenzweig, Phys. Rev. B 101, 201407(R) (2020)

O 94.3 Thu 15:45 MA 141

Pb-intercalated epitaxial graphene on SiC: Charge transfer mechanism and spin-split interlayer bands — •Bharti Matta[1], Philipp Rosenzweig[1], Kathrin Küster[1], Craig Polley[2], and Ulrich Starke[1] — [1]Max-Planck-Institut für Festkörperforschung, Stuttgart, Germany — [2]MAX IV Laboratory, Lund, Sweden

Intercalation of epitaxial graphene is a robust way of modifying its properties and stabilizing two-dimensional (2D) interlayers at the graphene/SiC interface. Pb being a heavy element superconductor has a great potential as an intercalant due to possible proximity effects in graphene. In this work, angle-resolved photoelectron spectroscopy (ARPES) shows that the near charge-neutrality of Pb-intercalated quasi-freestanding monolayer graphene on SiC (Pb-QFMLG) involves charge transfer from both interlayer-Pb and SiC. This is based on the observation that at 20 K, the p-doping of Pb-QFMLG increases by $\approx 9 \times 10^{10}$ cm^{-2} compared to room temperature, which can be attributed to the freezing out of SiC bulk n-dopants. The bands of interlayer-Pb cross the Fermi level, confirming its metallic nature. ARPES in the repeated Brillouin zone confirms the (1×1) alignment of Pb relative to SiC. Constant initial state mapping as a function of photon energy reveals dispersionless Pb bands, corroborating their 2D nature. Potassium adsorption results in predominant charge transfer into graphene, inducing a strong n-doping of $\approx 10^{14}$ cm^{-2}. Further, spin-resolved ARPES uncovers a strong spin splitting of the interlayer bands. However, any proximity-induced spin splitting in graphene is still ambiguous. This ongoing work is supported by DFG through FOR 5242.

O 94.4 Thu 16:00 MA 141

Electronic structure of intercalated epitaxial graphene: A first principles study — •Andres Unigarro[1], Florian Günther[2], Franziska Schölzel[1], Niklas Witt[3], Thomas Seyller[1], Tim Wehling[3], and Sibylle Gemming[1] — [1]Institute of physics, TU Chemnitz, Chemnitz, Germany — [2]UNESP, Rio Claro, Brazil — [3]Universität Hamburg, Hamburg, Germany

Two-dimensional materials such as graphene are fascinating because they combine unique mechanical and electronic properties. Proximity effects introduced from the interaction with intercalates and intercalation layers change the electronic, optical and transport properties of epitaxial graphene (EG) while preserving the regular honeycomb structure. Elements of the IV group such as Pb can be used as intercalants below a graphene sheet, leading to partly well-defined hetero bilayer with different functionalities. Heavy atoms like Pb furthermore, promise to introduce additional effects such as spin-orbit coupling to the electron gas of graphene. Using a density functional based tight binding approach, we investigate the modifications in the electronic structure of EG due to proximity effects induced by intercalation.

O 94.5 Thu 16:15 MA 141

Mott states under cover: Silicon intercalation of epitaxial graphene — •Niclas Tilgner, Zamin Mamiyev, and Thomas Seyller — TU Chemnitz Institut für Physik

Mott-Hubbard bands are well known from different superstructures on SiC(0001). Typical examples are the siliconrich (3×3) and $(\sqrt{3} \times \sqrt{3})R30°$ reconstruction, but also the unreconstructed (1×1) surface. In all cases the electrons occupying the dangling bonds show a non-negligible interaction among themselves, which gives rise to a Mott-Hubbard metal-insulator transition. The dispersion and the energetic splitting of the upper and lower Hubbard band was found to scale with the distance between the dangling bonds and the dielectric properties of the environment, respectively. It could be achieved to prepare an analogous system underneath graphene by intercalation of silicon. The talk focuses on the major results, which were acquired. Experiments using diffraction techniques (LEED, SPA-LEED) allow the evaluation of the silicon arrangement, that is found to consist of different patches with $(\sqrt{3} \times \sqrt{3})R30°$, (2×2) and (3×3) ordering. Furthermore, investigations of the prepared samples with photoemission techniques (ARPES, XPS) reveal several surface states, which can partly be attributed to the dangling bonds of the silicon adatoms and therefore to one of the Hubbard bands. Unintuitively, no avoided crossing with the Dirac cone is observed. Possible reasons for this circumstance will be discussed.

Topical Talk O 94.6 Thu 16:30 MA 141

Proximity spin-orbit coupling and topological interfaces in graphene / alloyed transition metal dichalcogenide heterostructures — •Stephen Power[1] and Zahra Khatibi[2] — [1]School of Physical Sciences, Dublin City University, Ireland — [2]School of Physics, Trinity College Dublin, Ireland

Spin-orbit coupling (SOC) in graphene can be enhanced by proximity effects in stacked graphene/transition metal dichalcogenide (TMDC) heterostructures. The TMDC layer composition determines the nature and strength of the resultant SOC induced in graphene.

Here, we demonstrate the evolution of the SOC in graphene with the composition of an alloyed TMDC layer ($W_xMo_{1-x}Se_2$). Density functional theory is used to simulate systems with different compositions and distributions, and allows local and global signatures of the metal-atom alloying to be clarified. The low-energy spin and electronic behavior follow a effective medium model which depends only on the composition ratio x.

While graphene/$MoSe_2$ has a simple band gap, valley-Zeeman-driven inverted bands are present in graphene/ WSe_2. The topological state of mixed systems can thus be tuned by varying x, with band gap closure occurring at a critical value. Furthermore, heterostructures with W- and Mo-domains can host boundary states similar to those between AB- and BA-stacked domains in bilayer graphene. Finally, we trace the electronic evolution of alloyed heterostructures from homogeneous effective medium to domain-localised bands as the domain size increases.

O 94.7 Thu 17:00 MA 141

Non-equilibrium carrier dynamics and band structure of graphene on 2D tin — •Maria-Elisabeth Federl[1], Niklas Hofmann[1], Leonard Weigl[1], Johannes Gradl[1], Niklas Witt[2], Tim Wehling[2], Biao Yang[3], Neeraj Mishra[4], Camilla Coletti[4], and Isabella Gierz[1] — [1]University of Regensburg — [2]University of Hamburg — [3]Technical University Munich — [4]Istituto Italiano di Tecnologia, Pisa

Van der Waals heterostructures are novel artificial materials with tailored electronic properties that might enable new technologies in the fields of optoelectronics, spintronics, and quantum computing. Wafer-scale heterostructures with clean interfaces are easily obtained by confinement heteroepitaxy on SiC(0001) [1] where various elements are intercalated below the covalently bound carbon buffer layer on SiC. Confinement heteroepitaxy has recently been used to stabilize a 2D layer of Sn with exotic electronic properties [2-4]. A possible interaction with the quasi-freestanding graphene layer on top remains unexplored. Using time-and angle-resolved photoemission spectroscopy, we find a surprisingly short-lived non-thermal carrier distribution inside the Dirac cone of graphene as well as a rigid down-shift of the whole π-band that we attribute to interlayer hybridization revealed by density functional theory.

[1] Nat. Mater. 19, 637 (2020)
[2] J. Phys. D: Appl. Phys. 49, 135307 (2016)
[3] Appl. Phys. Express 11, 015202 (2018)
[4] Phys. Rev. Lett. 122, 126403 (2019)

O 94.8 Thu 17:15 MA 141

Spatially Resolved Sn Intercalation of Epitaxial Buffer Layer of Graphene on SiC — •Benno Harling and Martin Wenderoth — IV. Physical Institute, Georg-August-University Göttingen, Germany

Intercalation, the insertion of atoms into a layered material such as graphene, has been shown to be a versatile means of altering electronic properties through proximity. While intercalation is typically discussed as a spatially homogeneous process, our study focusses on the inhomogeneities on the mesoscopic scale. An investigation of a partially intercalated epitaxial graphene buffer layer with tin was conducted using Kelvin Probe Force Microscope (KPFM). $\sqrt{3} \times \sqrt{3}$ and 1×1 phases of the intercalated tin can be discriminated as doping on the graphene layer corresponding to a specific shift in the work function. This allows to locate and quantify the different intercalation phases as well as the transition to the pristine buffer layer on a sub-micrometer scale. This opens insight into the diffusion of the intercalated atoms, i.e., the intercalation dynamics on this scale. Gradients from fully intercalated areas to the pristine buffer layer are observed and correlated with the topographic information. Our results show the strong impact of substrate steps as a significant boundary for the diffusion process, more specifically diffusion of Sn mainly happens on a terrace or via pin hole like channels across steps.

O 94.9 Thu 17:30 MA 141

Low energy plasmons in Sn-intercalated quasi-free-monolayer-graphene — •Zamin Mamiyev and Christoph Tegenkamp — Institut für Physik, Technische Universität Chemnitz

Graphene plasmons, particularly confined within structured and sandwiched graphene, represent a fascinating area of research. They hold the potential to facilitate the manipulation of light at sub-wavelength scales while providing a platform for investigating the many-body electronic interactions. Our study focuses on low-energy plasmonic excitations in epitaxial quasi-free monolayer graphene, created through the intercalation of Sn beneath the buffer layer on 4H-SiC(0001). [1]

The quantitative examination of the sheet plasmon dispersion reveals that the Sn-induced (1×1) interface exhibits metallic properties, leading to the formation of charge-neutral graphene. The dispersion of the sheet plasmon (activated by mild doping with K) is slightly redshifted with w.r.t. epitaxial graphene (EG). On the other hand, the Sn-diluted ($\sqrt{3} \times \sqrt{3}$) reconstruction forms intrinsically n-type doped graphene with a dispersion similar to the epitaxial monolayer graphene on the carbon buffer layer. Moreover, it appears that the dipolar coupling of longitudinal charge density fluctuations in EG to the interface layer triggers the formation of a plasmonic multipole component at higher frequencies. Our study shows that angle-resolved plasmon spectroscopy is a promising technique to investigate proximity effects of excitations in electronically weakly coupled 2D heterostructures.

[1] Z. Mamiyev and C. Tegenkamp, Surf. & Int. 34, 102304 (2022)

O 94.10 Thu 17:45 MA 141

Evidence of Sn-induced Mott states coupled to Dirac electrons in epitaxial graphene — •Chitran Ghosal[1], Zamin Mamiyev[1], Siheon Ryee[2], Niklas Witt[2,3], Tim Wehling[2,3], and Christoph Tegenkamp[1] — [1]Institute of Physics, Technische Universität Chemnitz, Reichenhainer Str. 70, Germany — [2]I. Institute of Theoretical Physics, University of Hamburg, Notkestraße 9-11, 22607 Hamburg, Germany — [3]The Hamburg Centre for Ultrafast Imaging, Luruper Chaussee 149, 22607 Hamburg, Germany

The adsorption of 1/3 ML of Sn on SiC(0001) was shown to reveal a robust 2D Mott state. In this work we studied the electronic structure of intercalated Sn below the buffer layer on SiC(0001) coming along with the formation of n-type doped graphene. SPALEED clearly revealed new $\sqrt{3}$-reconstruction spots after the intercalation process. By means of EELS and STS we analyzed in detail the electronic structure of this heterostructure. We found strong evidence of a hybridization between Sn-induced Mott states and the graphene π-bands. This leads to a gap opening of around 200 meV at the Dirac point. Moreover, a new state at around 1.2 eV emerged which we assign to the upper Hubbard band. The formation of narrow bands is supported by EELS measurements, showing besides the sheet plasmon also a pronounced interband transition at 1.5 eV. Combined DFT and dynamical mean field theory calculations support the emergence of strong Mott Hubbard correlations in this system. The emerging correlation effects show a pronounced stacking and charge transfer dependence and lead to spectral functions in good agreement with the experiments.

O 95: Electronic Structure of Surfaces II: Spectroscopy, Surface States

Time: Thursday 15:00–17:30 Location: MA 144

O 95.1 Thu 15:00 MA 144

Towards Robust Dichroism in Angle-Resolved Photoemission Spectroscopy — •Jakub Schusser[1], Hibiki Orio[1], Maximilian Ünzelmann[1], Johannes Hessdörfer[1], Muthu P.T. Masilamani[1], Florian Diekmann[2,3], Kai Rossnagel[2,3], and Friedrich Reinert[1] — [1]Experimentelle Physik VII and Würzburg-Dresden Cluster of Excellence ct.qmat, Universität Würzburg, D-97074 Würzburg, Germany — [2]Ruprecht Haensel Laboratory, Deutsches Elektronen-Synchrotron DESY, D-22607 Hamburg, Germany — [3]Institute of Experimental and Applied Physics, Kiel University, D-24098, Germany

Dichroic techniques in angle-resolved photoemission spectroscopy are highly relevant in the field of topological materials, layered systems, etc. Dichroism is per se a matrix-element effect that depends on the initial and final states as well as the light-matter interaction. However, accessing the information about the initial state of the solid directly has so far not been possible. By comparing both experimental and theoretical SPR-KKR soft X-ray data in bulk WSe_2 we show the robustness of the newly introduced dichroic technique against variation of photon energy, light polarization and angle of incidence. Such robustness of the matrix-element effect hints at a leap towards accessing the initial state properties directly and encourages further investigation.

O 95.2 Thu 15:15 MA 144

Dichroic Photoemission Tomography of Orbital Vortex Lines in a Topological Semimetal — •Maximilian Ünzelmann[1], Tim Figgemeier[1], Philipp Eck[2], Begmuhammet Geldiyev[1], Philipp Kagerer[1], Jakub Schusser[1], Domenico Di Sante[3], Giorgio Sangiovanni[2], Friedrich Reinert[1], and Hendrik Bentmann[4] — [1]Experimentelle Physik 7 and Würzburg-Dresden Cluster of Excellence ct.qmat, Universität Würzburg — [2]ITPA Würzburg and Würzburg-Dresden Cluster of Excellence ct.qmat, Universität Würzburg — [3]Department of Physics and Astronomy, Univerity of Bologna — [4]Center for Quantum Spintronics, Department of Physics, NTNU, Norway

In this talk, we report on the discovery of orbital vortex lines (OVL) in the three-dimensional (3D) band structure of a topological semimetal. Using dichroic photoemission tomography, i.e., linear and circular dichroism applied at bulk-sensitive soft x-ray photon energies [1], we directly observe vortices of atomic orbital angular momentum (OAM) and trace their trajectories in full 3D momentum space. In the core of the OAM vortex we resolve a two-fold spin-degenerate Weyl nodal line [2]. The experimental data is supported by density functional theory as well as photoemission intensity calculations. Taken together, our experiments achieve the first imaging of non-trivial quantum-phase winding at line nodes.

[1] M. Ünzelmann et al., Nat. Commun., 12, 3650 (2021)
[2] M. Hirschmann et al. Phys. Rev. Mat., 5, 054202 (2021)

O 95.3 Thu 15:30 MA 144

Circular dichroism by core-level angle-resolved photoemission: Application of multiple and single-site scattering theory — •Trung-Phuc Vo[1], Olena Tkach[2,3], Sylvain Tricot[4], Didier Sébilleau[4], Olena Fedchenko[2], Hans-Joachim Elmers[2], Gerd Schönhense[2], and Ján Minár[1] — [1]New Technologies - Research Center, Univ West Bohemia, 30100 Pilsen, Czech Republic — [2]Johannes Gutenberg-Universität, Institut für Physik, 55128 Mainz, Germany — [3]Sumy State Univ, 40007 Sumy, Ukraine — [4]Univ Rennes, CNRS, IPR (Institut de Physique de Rennes), F-35000, Rennes, France

Photoelectron diffraction (PED) is a powerful and driving experimental technique for resolving surface structures with sub-ångstrom resolution, namely bonding geometries of atoms and the local environment of impurity or dopant atoms inside surfaces. In high energy regime, PED effects are found in ARPES measurements beside other obstacles (low cross-sections, large photon momentum transfer, non-negligible phonon scattering). Here, to disentangle these diffraction influences and pronounced Kikuchi patterns, we present a PED implement for SPRKKR package which makes use of multiple scattering theory and one-step model in photemission process. In contrast to the other real space implementations of the multiple scattering PED formalism, we propose to use k-space implementation based on the layer KKR method. The main advantage is that we can without convergence problems (wrt. the angular momentum and cluster size) address very broad kinetic energy range (20-8000eV). A so-called alloy analogy model can be utilized to simulate XPD at finite temperatures.

O 95.4 Thu 15:45 MA 144

VUV Polarimeter for Inverse Photoemission — •Pascal Jona Grenz, Patrick Geers, Lennard Strompen, and Markus Donath — Physikalisches Institut, Universität Münster, Germany

In photoemission (PE) experiments, the use of polarized light for excitation provides access to orbital information of the electronic states under investigation. In inverse photoemission (IPE), the equivalent is the analysis of the polarization of the emitted light. So far, this light-polarization analysis was neglected due to the much lower cross-section in IPE compared with PE and the lack of suitable optics in the VUV (vacuum ultraviolet) spectral range.

In this contribution, we present a VUV polarimeter for IPE. A mirror with a polarizing coating of high reflectivity in Brewster angle geometry is used. With the high reflectivity and polarization power, this provides an easy-to-use attachment to the established photon detector [1]. We performed IPE measurements of the well-studied Cu(111) surface state by detecting p- and s-polarized light separately. The data show a strong dependence of the photon intensity on the polarization directions. Our results are in accordance with polarization-dependent PE measurements of the occupied part of the surface state [2].

[1] Thiede *et al.*, Meas. Sci. Technol. **29**, 065901 (2018)
[2] Mulazzi *et al.*, Phys. Rev. B **79**, 165421 (2009)

O 95.5 Thu 16:00 MA 144

Double Photoemission of C_{60} on $SrTiO_3(001)$ with pulsed laser radiation — •Kathrin Plass[1], Robin Kamrla[1], Frank O. Schumann[2], and Wolf Widdra[1] — [1]Institute of Physics, Martin-Luther-Universität Halle-Wittenberg, Halle (Saale), Germany — [2]Max Planck Institute of Microstructure Physics, Halle (Saale), Germany

Via photoelectron spectroscopy, important insights into the electronic structure of solids were obtained. However, correlation effects can only be addressed indirectly. With double photoemission spectroscopy (DPE), such phenomena can be observed directly by detecting pairs of correlated photoelectrons emitted upon absorption of a single photon [1]. C_{60} is classified as a strongly correlated material with a highly structured valence band. In this contribution we present DPE data for C_{60} thin films on $SrTiO_3(001)$, obtained by a laboratory high-order harmonic (HHG) light source, operating at MHz repetition rates [2]. For different photon energies, we analyze the 2D energy maps of correlated electron pairs and compare the resulting sum energy spectra to simulations using a two-electron density of states [3,4]. This allows us to determine orbital-resolved two-electron binding energy shifts in C_{60} for the first time.

[1] J. Berakdar et al., Phys. Rev. Lett. **81**, 3535 (1998)
[2] A. Trützschler et al., Phys. Rev. Lett. **118**, 136401 (2017)
[3] M. Cini, Solid State Communications **24**, 681 (1977)
[4] G. A. Sawatzky, Phys. Rev. Lett. **39**, 504 (1977)

O 95.6 Thu 16:15 MA 144

On the unoccupied electronic structure of $Fe_3O_4(100)$ — •Jan Bieling and Markus Donath — Universität Münster, Germany

The (100) surface of magnetite (Fe_3O_4) undergoes a $(\sqrt{2}\times\sqrt{2})R45^\circ$ reconstruction. While its occupied states have been extensively investigated, studies addressing the empty ones are lacking.

We examined the unoccupied electronic structure of a pristine and well-characterized $Fe_3O_4(100)$ surface by angle-resolved inverse photoemission. A threefold split, almost non-dispersive $3d$-related spectral feature is observed. It is attributed to band groups originating from tetrahedrally and octahedrally coordinated iron atoms. The latter give rise to two spectral features since the t_{2g} and e_g orbitals are non-degenerate due to crystal-field splitting.

Based on our present study, we showcase the inherent limitations of using the second derivative to analyze electron spectroscopical data. Extracted peak positions can significantly deviate from the underlying ones, as demonstrated by synthetically generated spectra.

O 95.7 Thu 16:30 MA 144

Unusual Behavior of the Spin-Split L-Gap Surface Resonance at Pt(111) — •Fabian Schöttke, Peter Krüger, and Markus Donath — Universität Münster, Germany

We investigate the unoccupied electronic structure of Pt(111) with focus on the surface resonance (SR) at the bottom of the L gap. SR is discussed in the literature to be either typically free-electron like or hybridized with bulk states [1]. With different approaches within density-functional theory, we demonstrate that the lattice constant is crucial for the energetic position and dispersion behavior of SR. For experimentally obtained structural parameters as derived from low-energy electron diffraction studies [2], the hybridization-induced shape of SR results. In the case of hybridization, close to $\overline{\Gamma}$, a downward dispersing branch of SR splits off with predominantly one spin direction. This behavior is a distinct deviation of the usual parabolic upward dispersing L-gap surface states. By spin- and angle-resolved inverse photoemission, we measure the dispersion of SR in close vicinity to $\overline{\Gamma}$ and find a scenario of spin-split spectral features in agreement with the aforesaid theoretical results. Furthermore, we elucidate how the position of SR at the bottom of the L gap leads to hybridization with the d bands in comparison with L-gap surface states/resonances at other fcc(111) surfaces [3].

[1] A. Dal Corso, Surface Science **637-638**, 106 (2015).
[2] L. Hammer, T. Kißlinger, A. Schneider, private communication.
[3] Braun & Donath, Europhys. Lett. **59**, 592 (2002).

O 95.8 Thu 16:45 MA 144

On the unoccupied electronic structure of Hf(0001) — •Lennard Strompen[1], Vincent Reinartz[1], Sven Schemmelmann[1], Karol Hricovini[2], Saleem Khan[3], Jan Mínar[3], and Markus Donath[1] — [1]Universität Münster, Muenster, Germany — [2]CY Cergy Paris University, Paris, France — [3]University of West Bohemia, Pilsen, Czech Republic

Spin-orbit-induced effects are of particular importance in the surface electronic structure of high-Z materials. In our study, we focus on the unoccupied electron states at the so far unexplored Hf(0001) surface. The first challenge was to prepare a well-ordered and clean Hf(0001) surface, which, according to the literature, is known to show considerable impurities, in particular oxygen. After having developed a successful preparation recipe, we used spin- and angle-resolved inverse photoemission to determine the energy vs wavevector dispersion of the unoccupied states along the high-symmetry directions $\overline{\Gamma M}$ and $\overline{\Gamma K}$. Besides bulk-related spectral features and an image-potential-induced surface state, we identify surface-related features, which are interpreted on the basis of calculations of the electronic structure.

O 95.9 Thu 17:00 MA 144

Electronic structure of the $(\sqrt{3}\times\sqrt{3})R30^\circ$ TlAg$_2$ surface alloy on Ag(111) — •Sven Schemmelmann[1], Patrick Härtl[2], Peter Krüger[3], Matthias Bode[2], and Markus Donath[1] — [1]Physikalisches Institut, Universität Münster — [2]Physikalisches Institut, Experimentelle Physik II, Universität Würzburg — [3]Institut für Festkörpertheorie, Universität Münster

The BiAg$_2$ surface alloy on Ag(111) exhibits a giant Rashba splitting which is one order of magnitude larger than the splitting of the L-gap surface state on Au(111) [1]. We investigate the unoccupied electronic structure of the similar $(\sqrt{3}\times\sqrt{3})R30^\circ$ TlAg$_2$ surface alloy by scanning tunneling spectroscopy (STS) and angle-resolved inverse photoemission (IPE) [2]. We observe two dominant empty-state electronic features, which are attributed to downward dispersing s, p_z-derived states and to states with p_z orbital symmetry, respectively. On the basis of bandstructure and charge distribution calculations, we discuss the variation of the binding energies of the respective electronic features observed by STS and IPE.

[1] Ast et al., Phys. Rev. Lett. 98, 186807 (2007)
[2] Härtl et al., Phys. Rev. B 107, 205144 (2023)

O 95.10 Thu 17:15 MA 144

Insights into the dominant confinement mechanism and a spatially inhomogeneous broadening effect in quantum corrals — •Marco Weiss, Michael Schelchshorn, Fabian Stilp, Alfred J. Weymouth, and Franz J. Giessibl — Institute of Experimental and Applied Physics, University of Regensburg, Universitätsstraße 31, 93053 Regensburg

Understanding the factors influencing the lifetime of electronic states in artificial quantum structures is of great significance for advancing quantum technologies. This study focuses on CO-based quantum corrals on a Cu(111) surface. Tunneling spectroscopy measurements revealed a strong correlation between the size of the quantum corral and spectral width, characterized by a predominant Gaussian line shape. We attribute this dominant Gaussian-shaped lifetime broadening to the interaction of surface state electrons with the corral boundary.

To further investigate this phenomenon, we constructed corrals of the same size but varying wall densities. Our findings indicate that the energetic behavior of resonant eigenstates in a quantum corral is predominantly dictated by elastic processes, such as tunneling, rather than lossy interactions with the wall, like bulk coupling or inelastic scattering.

Due to our characterization of the interaction between resonant electrons states and their confining boundary, the observations made in our work enhance the understanding of lifetime limiting factors in artificial quantum structures, paving the way for more predictive simulations and more controllable quantum systems.

O 96: Solid-Liquid Interfaces IV: Reactions and Electrochemistry

Time: Thursday 15:00–18:00 Location: TC 006

O 96.1 Thu 15:00 TC 006

How to exploit the electrochemical driving forces to understand electrochemical CO(2) reduction — •Georg Kastlunger[1], Hendrik Heenen[2], and Nitish Govindarajan[3] — [1]Technical University of Denmark, Fysikvej, Kongens Lyngby, Denmark — [2]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — [3]Materials Science Division, Lawrence Livermore National Laboratory, Livermore, California, USA

The first-principles understanding of complex reaction mechanisms in electrocatalysis aids not only the discovery of improved catalytic materials but also the choice the ideal reaction environment for tailored products. For the latter, an appropriate understanding of the influence of the electrochemical driving forces, such as potential and pH, is key.

In my talk, I will present density functional theory-based studies on electrocatalytic reaction mechanisms with a special focus on electrochemical CO(2) reduction (eCO(2)R). I will describe how the combination of constant-potential DFT approaches[I] and transition state theory-based considerations allow us to explicitly study the potential and pH dependence of multistep reaction networks.[II] I will further discuss the kinetic characteristics of the competing elementary reactions within eCO(2)R and their consequences on the potential and pH response of the product selectivity.[III]

I. Kastlunger et al., J. Phys. Chem. C 122, 12771-12781 (2018).
II. Kastlunger et al., ACS Catal. 12, 4344-4357 (2022).
III. Kastlunger et al., ACS Catal., 13, 7, 5062-5072 (2023)

O 96.2 Thu 15:15 TC 006

Multiscale modeling reveals mass transport-controlled product selectivity in electrochemical CO2 reduction on Cu — •Adith Ramakrishnan Velmurugan[1], Youngran Jung[2], Dae-Hyun Nam[3], and Stefan Ringe[1] — [1]Korea University, Seoul, Republic of Korea — [2]Seoul National University, Seoul, Republic of Korea — [3]DGIST, Daegu, Republic of Korea

Electrochemical CO2 reduction, one of the most promising processes for a sustainable closure of the artificial carbon cycle, is severely limited by the lack of a catalyst that can reduce CO2 to higher-reduced chemicals actively and selectively. Cu is the only catalyst found to produce considerable amounts of C2 products, albeit at high overpotentials. The conversion mechanism is unclear, with different active sites and rate-determining steps being proposed. In addition, the mass transport of CO2 has been suggested to significantly impact the product selectivity. Gas-diffusion-layer (GDL)-based electrolyzers have become a state-of-the-art solution to circumvent these mass transport limitations. In this work, we present a new multi-scale model based on first-principles kinetics, and a modification of a recently reported gas diffusion electrode model. From this model, we show that even in GDL systems, mass transport is the limiting factor governing all experimentally observed trends in product selectivity, irrespective of the reaction mechanism or product pathway. We further find indications of C2/C1 product selectivity being dependent on the pore size and depth. This work provides strong evidence for the importance of mass transport in designing CO2 electrolyzers.

O 96.3 Thu 15:30 TC 006

The mechanism of electrochemical CO_2 reduction to post CO and C_{2+} products over single atom catalysts — Reza Khakpour[1], Kaveh Farshadfar[1], Kari Laasonen[1], and •Michael Busch[2,3] — [1]Aalto University, Esbo, Finland — [2]Luleå University of Technology, Luleå, Sweden — [3]Wallenberg Initiative Materials Science for Sustainability (WISE), Luleå, Sweden

Electrochemical reduction of CO_2 to CO or post CO products is of central importance for energy storage and conversion. A promising class of catalysts for CO_2 reduction are single atom catalysts (SACs) which consist of a single metal atom embedded into graphene. These materials are generally believed to only form C_1 compounds. However, recent experiments indicate, that methane together with minor amounts of products with 2 or more carbon atoms are formed over Fe phthalocyanine complexes, which are structurally similar to classical SACs.[1]

In this contributions we will explore the reaction routes from CO_2 to methane and C_{2+} compounds using density functional theory (DFT) computations [2]. Our results indicate, that the selectivity between different products mainly depends on activation barriers and is strongly influenced by the CO and proton concentration close to the electrode.

[1] S.-T. Dong, C. Xu, B. Lassalle-Kaiser *Chem. Sci.* **14** (2023) 550.

[2] R. Khakpour, K. Farshadfar, M. Busch et al. *submitted.*

Topical Talk O 96.4 Thu 15:45 TC 006

Importance of charge transfer descriptor for the computational screening of electrocatalysts — •Stefan Ringe — Korea University, Seoul, Rep. of Korea

Electrochemistry has become a key player in establishing a global sustainable energy landscape. Unfortunately, most electrochemical processes are limited in their efficiency and selectivity which has prevented them from replacing carbon-intensive industrial processes. Computational simulations have the potential to conquer the vast chemical space and reveal so far unconsidered new electrocatalysts and boost the performance of these devices. This, however, requires the knowledge of computationally accessible activity and selectivity descriptors. In this work, and by the example of electrochemical CO2 reduction, it is shown that the conventionally considered adsorption energy descriptor is not enough to distinguish product selectivity among catalysts. Including specifically the electrochemical environment in the quantum chemical calculations, we find that a charge-transfer descriptor is separately required, such as the work function or the potential of zero charge. With this, experimental trends in product selectivity can be well described, thus providing a new set of descriptors for high-throughput screening of electrocatalysts.

O 96.5 Thu 16:15 TC 006

Potentially Uncontrolled Barrier Calculations for Electrocatalysis — •Simeon D. Beinlich[1,2], Georg Kastlunger[3], Karsten Reuter[1,2], and Nicolas G. Hörmann[1] — [1]Fritz-Haber-Institut der MPG, Berlin — [2]Technische Universität München — [3]Technical University of Denmark, Kongens Lyngby, Denmark

Does computing electrochemical barriers necessarily require the use of a potentiostat? Here, we present a novel set of potentiostat-free methods for computing grand canonical activation barriers at constant potential from common canonical DFT calculations [1]. Making most efficient use of the underlying DFT data, these methods can, by construction, re-create potentiostat-based results. Moreover, they offer several systematic approximations that cover the potential-induced electronic and geometric responses, while only requiring a single transition state search at the point of zero charge, i.e. at zero excess charge.

Besides offering new pathways for efficiently calculating electrochemical activation barriers, our analysis highlights the importance of including geometric effects and more generally sheds light on the similarities and dissimilarities between a canonical and a grand canonical treatment of electrochemical interfaces. [1] S.D. Beinlich *et al.*, J. Chem. Theory Comput., https://doi.org/10.1021/acs.jctc.3c00836

O 96.6 Thu 16:30 TC 006

Relation between Electrocatalyst Morphology and Product Selectivity from Multi-Scale Reaction Models — •Hemanth S. Pillai, Hendrik H. Heenen, Karsten Reuter, and Vanessa J. Bukas — Fritz-Haber-Institut der MPG, Berlin

Electrocatalytic selectivity has shown a puzzling dependence on experimental parameters related to catalyst morphology. We argue that such effects can often be rationalized on the basis of mesoscopic mass transport. Basis for the underlying mechanism is the competition that arises from exchanging surface-bound, yet volatile, reaction intermediates between the electrode and the bulk electrolyte. The catalyst morphology can be decisive in driving this competition since its surface area directly affects the probability that a diffusing species will return to the surface for continued conversion, rather than escape as an early intermediate product. It remains unclear, however, exactly how catalyst morphology is predicted to affect the resulting selectivity and neither is the level of detail that needs to be considered. In this study, we specifically demonstrate this competition for the electrochemical CO_2 reduction on Cu. For this purpose, we develop a simple multi-scale model that couples diffusion to the electrochemical surface kinetics. This allows to predict selectivity towards the early CO product, while systematically improving the representation of catalyst morphology within our model: from an effective one-dimensional descriptor of surface roughness, to an explicit two-dimensional consideration of surface corrugation.

O 96.7 Thu 16:45 TC 006

Converging Divergent Paths: Constant Charge vs. Constant Potential Energetics in Computational Electrochemistry — •Nicolas G. Hörmann, Simeon D. Beinlich, and Karsten Reuter — Fritz-Haber-Institut der MPG, Berlin

Using the example of a proton adsorption process, we analyze and compare two prominent modelling approaches in computational electrochemistry at metallic electrodes – electronically canonical, constant-charge and electronically grand-canonical, constant-potential calculations. We first confirm that both methodologies yield consistent numerical results for the differential free energy change in the infinite cell size limit. This validation emphasizes that, fundamentally, both methods are equally valid and precise. In practice, the grand-canonical, constant-potential approach shows superior interpretability and size convergence as it aligns closer to experimental ensembles and exhibits smaller finite-size effects. On the other hand, constant-charge calculations exhibit greater resilience against discrepancies, such as deviations in interfacial capacitance and absolute potential alignment, as their results inherently only depend on the surface charge, and not on the modeled charge vs. potential relation. The present analysis thus offers valuable insights and guidance for selecting the most appropriate ensemble when addressing diverse electrochemical challenges.

O 96.8 Thu 17:00 TC 006

Approximating Grand-Canonical Energetics of Electrified Semiconductor-Electrolyte Interfaces – A Benchmark Study — •Hedda Oschinski[1,2], Karsten Reuter[1,2], and Nicolas G. Hörmann[1] — [1]Fritz-Haber-Institut der MPG, Berlin — [2]Technische Universität München

Computational studies of the electrosorption of adsorbates centrally target the grand canonical (GC) energetics $\mathcal{G}^{\mathrm{s}}(\phi_E)$ with respect to an applied potential ϕ_E. For metal electrodes, $\mathcal{G}^{\mathrm{s}}(\phi_E)$ can be well described by a parabolic form that incorporates the interfacial capacitance C at the potential of zero charge. Unfortunately, for semiconductors (SC), this appealing approximation breaks down due to the existence of the band gap. Here, we propose a simple extension that explicitly incorporates the electrode density of states (DOS) and thus allows to describe C for both metals and SCs. For SCs, C is thereby partitioned into a DOS capacitance and a solution capacitance using a straightforward DOS shift picture that combines the filling of electronic states with the shift of the electrostatic potential drop across the electrode-electrolyte interface. Using density-functional theory calculations in an implicit solvation environment, we benchmark the model for a set of conducting and semiconducting/insulating 2D materials. The GC energetics is reliably reproduced across the entire materials space, with only minor deviations but at a fraction of the computational cost. The analysis underlines the importance of the position of the band gap and the response of the solution, in line with classical macroscopic ideas of semiconductor electrochemistry.

O 96.9 Thu 17:15 TC 006

Thermodynamic Cyclic Voltammograms from First Principles — •Nicolas Bergmann, Nicolas G. Hörmann, and Karsten

Reuter — Fritz-Haber-Institut der MPG, Berlin

Computationally, the predictive-quality modeling of cyclic voltammograms (CVs) is complicated by the need to accurately account for the interactions and reactive chemistry at the liquid electrolyte/solid electrode interface, as well as for the electrostatic interactions of the diffuse double layer at applied potential conditions [1]. For sufficiently small scan rates, *ab initio* thermodynamics approaches help to meet these challenges.

Here, we compare different approximations commonly employed in such approaches [1]. Using the well-studied model system of Ag(100) in a Br-containing electrolyte as a show case, we analyze the influence of statistical sampling of the adlayer, how implicit solvent models affect the surface energetics, and the benefits of augmenting traditional zero-field calculations - aka the computational hydrogen electrode (CHE) - with capacitive double layer energetics (CHE+DL) [2]. Finally, we illustrate the possibility to expand our methodology to more complex systems by examining the CVs of Cu(100) electrodes in alkaline, I-containing electrolytes.

[1] N. Bergmann, N.G. Hörmann, and K. Reuter, J. Chem. Theory Comput. (in press) DOI:10.1021/acs.jctc.3c00957.

[2] N.G. Hörmann and K. Reuter, J. Chem. Theory Comput. **17**, 1782 (2021).

O 96.10 Thu 17:30 TC 006

On the Origin of Electrocatalytic Selectivity during the Oxygen Reduction Reaction on Au(111) — •Elias Diesen, Karsten Reuter, and Vanessa J. Bukas — Fritz-Haber-Institut der MPG, Berlin

A puzzling observation during the oxygen reduction reaction (ORR) on Au electrodes is the preference to form hydrogen peroxide (H_2O_2), instead of the thermodynamically favored water product. This selectivity cannot be explained on the basis of thermodynamic reaction models that simply assume a series of proton-coupled electron transfers (PCETs). Here, we use *ab initio* molecular dynamics along with umbrella sampling to obtain free energy profiles for competing key ORR steps on Au(111). Our comparison includes not only PCETs, but also “chemical” reaction steps that do not include a Faradaic charge transfer, such as desorption or surface dissociation. This allows us to explore favorable reaction paths, while varying the capacitive charging to represent realistic ORR potentials. Our results show that all reaction steps competing with H_2O_2 formation have sizeable kinetic barriers and are thus prohibited, even though they may be thermodynamically favored. We find that this situation does not change under more reducing conditions and specifically determine the “nobleness” of Au as playing a decisive role in preventing O-O bond scission. It is thus not the applied potential, but the underlying chemistry that drives the ORR selectivity. In general, our study thus highlights the kinetic competition between PCET and non-PCET steps that cannot be resolved via simple Brønsted-Evans-Polanyi (BEP) scaling relations.

O 96.11 Thu 17:45 TC 006

Computational chemistry analysis of passive layer formation and breakdown mechanisms in ferritic stainless steels — •Vahid Jamebozorgi[1,2], Karsten Rasim[3], and Christian Schröder[1,2] — [1]Bielefeld Institute for Applied Materials Research, Bielefeld University of Applied Sciences and Arts, 33619 Bielefeld, Germany — [2]Faculty of Physics, Bielefeld University, 33615 Bielefeld, Germany — [3]Miele \& Cie. KG, Center for Materials (CFM), Carl-Miele-Straße 29, 33332 Gütersloh

Despite extensive research on passive layer formation and breakdown, several questions still remain unanswered. These include the reasons behind the bilayer nature of the passive layer, the decrease in hydrogen and oxygen diffusivity upon entering the passive layer, the underlying mechanisms of passive layer formation and breakdown, and the influence of microstructure on passive layer formation and function in stainless steels. In this study, we employed ReaxFF molecular dynamics to investigate passivation and depassivation of stainless steel in a polycrystalline structure. Through static and dynamic calculations, we elucidated the mechanisms of passive layer formation, which were primarily governed by clustering. Our analysis also highlighted the significant role of hydrogen diffusion and its reaction with metallic compounds in depassivation. We have identified several physical phenomena involved in the processes of passivation and depassivation, which can provide explanations for the aforementioned unresolved points.

O 97: Focus Session: SrTiO$_3$: A Versatile Material from Bulk Quantum Paraelectric to 2D Superconductor II (joint session TT/KFM/MA/O)

Strontium titanate (SrTiO$_3$) is a paradigmatic material that plays an important role in various fields of solid-state physics, surface science and catalysis: The pure bulk phase is a wide-band-gap semiconductor that upon cooling becomes a textbook quantum paraelectric. When slightly doped, SrTiO$_3$ turns into a Fermi-liquid-type metal that becomes superconducting at extremely low charge carrier density. SrTiO$_3$-based surfaces and interfaces host un-conventional electronic states such as quasi-two-dimensional electron liquid, magnetism and superconductivity. Despite intensive studies over the past decades, SrTiO$_3$ continues to reveal surprising new phenomena that challenge the established views on this material. To this end achieving light-induced nonequilibrium states and the recent preparation of a 2D oxide based on SrTiO$_3$ opens new playgrounds for research. This Focus Session will present exciting developments in the study of electronic states that are based on the peculiar properties of SrTiO$_3$.

Please note that this Focus Session comprises four parts: Posters are presented within the TT poster session TT58 (Wed 15:00-18:00, poster area E). Invited talks are compiled in the session TT62 (Thursday, 9:30 to 12:45, H0104), Contributed talks will be presented in sessions TT72 (Thursday 15:00-18:00, H0104) and TT83 (Fri 9:30-12:30, H0104).

Organizers: Rossitza Pentcheva, University of Duisburg-Essen, Marc Scheffler, University of Stuttgart

Time: Thursday 15:00–18:00 Location: H 0104

O 97.1 Thu 15:00 H 0104

Origin of unconventional normal-state transport and superconductivity in electron-doped SrTiO$_3$ — •Stephen Rowley — Cavendish Laboratory, University of Cambridge, J. J. Thomson Avenue, Cambridge, CB3 0HE, United Kingdom

Quantum phase transitions may be reached in many ferroelectric systems by supressing the Curie temperature to absolute zero using a control parameter such as chemical substitution or hydrostatic pressure. In electron-doped specimens of quantum critical ferroelectrics such as SrTiO$_3$, unconventional superconductivity and unusual normal-state transport have been detected. In the latter case, a resistivity varying as temperature-squared is observed over a wide range of temperatures above the Fermi temperature. We present new experimental and model results that provide insight into the nature of the mechanisms for both superconductivity and normal-sate transport. We find in experiments and quantitative models without adjustable parameters, that both effects are connected and enhanced in samples tuned to the ferroelectric quantum critical point. Superconductivity appears to arise near the critical point due to the virtual exchange of longitudinal hybrid-polar-modes, even in the absence of a direct coupling to the transverse-optical phonon modes.

O 97.2 Thu 15:15 H 0104

Dilute superconductivity in the vicinity of a ferroelectric quantum critical point coupled via the "vector coupling": The case of $SrTiO_3$ — •Sudip Kumar Saha[1,2], Avraham Klein[1], Jonathan Ruhman[2], and Maria Navarro Gastiasoro[3] — [1]Ariel University, Israel — [2]Bar-Ilan University, Israel — [3]Donostia International Physics Center, Spain

Lightly doped $SrTiO_3$ (STO) is one of the most studied examples of quantum ferroelectric metal (QFEMs), where superconductivity coexists with ferroelectric order. Pristine STO is paraelectric naturally close to a ferroelectric quantum critical point (QCP). Strain or chemical substitution (for example, doping with Ba/Ca instead of Sr) drives STO through the QCP to the ferroelectric phase, which manifests itself in the softening of the transverse optical (TO) phonon mode. Doped samples are superconducting, where the T_c vs. density dome extends to very low density. To date, there is no consensus on the mechanism leading to superconductivity at such low density. Edge et al. have proposed that the ferroelectric QCP and dilute superconductivity are related [Phys. Rev. Lett. 115, 247002 (2015)]. In this work we explore the possible origin of low-density superconductivity from coupling linearly to the TO mode via a "vector coupling". We solve the critical-Eliashberg theory numerically, including fermionic and bosonic self-energy corrections, which allows us access all the way to the QCP. Notably, all our calculations are justified within standard approaches. We find the existence of a superconducting dome with magnitude and dependence on the distance from the QCP that resembles experiments.

O 97.3 Thu 15:30 H 0104

Dislocation-based filamentary superconductivity in reduced $SrTiO_3$ — •Christian Rodenbücher[1], Gustav Bihlmayer[2], Carsten Korte[1], and Kristof Szot[3] — [1]Forschungszentrum Jülich GmbH, Institute of Energy and Climate Research (IEK-14), 52425 Jülich, Germany — [2]Forschungszentrum Jülich GmbH, Peter Grünberg Institut (PGI-1), 52425 Jülich, Germany — [3]University of Silesia, Institute of Physics, 41-500 Chorzów, Poland

Exposure of $SrTiO_3$ single crystals to reducing conditions at elevated temperatures leads to the generation of metallic filaments forming along of dislocations, which act as preferential reduction sites. This effect can be enhanced when stimulating the local deoxidation by electric fields. This results in an agglomeration of metallic filaments in nano-bundles, which are embedded in the insulating surrounding crystal matrix. Despite removing only 10^{14-15} oxygen atoms from the dislocation network, electro-reduced crystals are superconducting with a transition temperature of 0.2 K, and their residual resistance is lower than that of purely thermally-reduced crystals. As the total amount of oxygen removed during electro-reduction is much smaller than the smallest reported carrier concentration for superconducting $SrTiO3_{-x}$ so far, our findings challenge traditional explanations of superconductivity in metal oxides. Combining conductivity characterization by atomic force microscopy with theoretical analysis of the dislocation cores, we propose a model explaining the superconducting properties by the coexistence of metallic dislocation cores with polar insulating regions allowing for polaronic coupling in the bundles.

O 97.4 Thu 15:45 H 0104

Dislocation-Induced Photoconductivity Enhancement in Fe-Doped $SrTiO_3$: compensation of low mobility by high carrier density through the emergence of a sub-band gap level — •mehrzad soleimany[1,2], till frömling[1], jürgen rödel[1], and marin alexe[2] — [1]Department of Materials and Earth Sciences, Technical University of Darmstadt, Darmstadt, Germany — [2]Department of Physics, University of Warwick, Coventry, UK

Owing to the remarkable properties of $SrTiO_3$ (STO), such as quantum paraelectric state below 37 K, negative differential resistance under illumination, and significant alteration of properties by doping, STO stands out among perovskite oxides. Until recently, little attention had been paid to the tunability of its properties - especially optical properties -via the introduction of dislocations. In this study, we introduce the method of dislocation imprint, which allows us to induce high densities of dislocations (> 1 x 10^{14} m^{-2}) into a large volume of Fe-doped STO. Low-temperature I-V measurements indicated an about one order of magnitude increase in the photoconductivity of dislocation-rich samples. Photo-Hall measurements revealed that while dislocations might decrease the mobility, they could enhance the photoconductivity by increasing the number of carriers. Spectral responsivity measurements demonstrated that the higher carrier density could stem from the emergence of a sub-band gap level. Complementary C-AFM measurements conducted under illumination confirmed the local enhancement of photoconductivity at dislocations, which fitted well to the Electron Channeling Contrast Images of dislocations.

O 97.5 Thu 16:00 H 0104

IR and THz studies on $(Ba_{0.45}Sr_{0.55}TiO_3)_{24}Ba_{0.45}Sr_{0.55}O$ and $(Ba_{0.45}Sr_{0.55}TiO_3)_8Ba_{0.45}Sr_{0.55}O$ thin films — Veronica Goian[1], Matthew Barone[2], Natalie Dawley[2], Christelle Kadlec[1], •Darrell Schlom[2,3], and Stanislav Kamba[1] — [1]Institute of Physics ASCR, Prague, Czech Republic — [2]Department of Materials Science and Engineering, Cornell University, Ithaca, NY, USA — [3]Kavli Institute at Cornell for Nanoscale Science, Ithaca, NY, USA

$(SrTiO_3)_nSrO$ and (n=1..6) films crystallizing in the Ruddlesdden-Popper (RP) structure are well known for low dielectric loss and large microwave permittivities which are highly tunable with electric field.[1,2] Bulk $(SrTiO_3)_nSrO$ is paraelectric, but the tensile strained thin films deposited on $(110)DyScO_3$ with $n \geq 3$, become ferroelectric at low temperatures. $(ATiO_3)_{24}AO$ and $(ATiO_3)_8AO$, A= $Ba_{0.45}Sr_{0.55}$ films deposited on $(110)DyScO_3$ exhibit no strain and yet become ferroelectric. Here we performed infrared and THz studies of phonon dynamics down to 10 K and compared it with above mentioned thin films and $(Sr,Ba)TiO_3$. The effect of soft mode and central mode on microwave dielectric properties and electric field tunability of permittivity will be discussed.

[1] C. H. Lee et al., Nature, 502 (2013) 532

[2] N. M. Dawley et al., Nat. Mater. 19 (2020) 176

O 97.6 Thu 16:15 H 0104

Polar phonon behaviour in polycrystalline Bi-doped strontium titanate thin films — •Oleksandr Tkach[1], Olena Okhay[2], Dmitry Nuzhnyy[3], Jan Petzelt[3], and Paula M. Vilarinho[1] — [1]Department of Materials and Ceramic Engineering, CICECO, University of Aveiro, Aveiro, Portugal — [2]TEMA-Centre for Mechanical Technology and Automation, Department of Mechanical Engineering, University of Aveiro, Aveiro, Portugal — [3]Institute of Physics of the Czech Academy of Sciences, Prague, Czechia

Among strontium titanate (STO) based materials, Bi-doped STO have been intensively studied as for dielectric as for resistance-switching memory and thermoelectric applications. Here, we enhance the dielectric characterisation by a lattice dynamics study of sol-gel-derived $Sr_{1-1.5x}Bi_xTiO_3$ thin films with x = 0.0053 and 0.167, deposited on Al_2O_3 substrates, using a variable-temperature far-infrared spectroscopy in a transmittance mode. Bi doping, known to induce a low-frequency dielectric relaxation in STO ceramics and films, due to off-centre dopant ion displacements generating electric dipoles, is shown to affect the polar phonon behaviour of thin films. We show that in weakly Bi-doped films, the low-frequency polar TO1 mode softens on cooling but less than in undoped STO. In heavily Bi-doped STO films, this mode displays no significant frequency variation with temperature from 300 to 10 K. The polar phonon behaviour of polycrystalline Bi-doped STO thin films is comparable with that of Bi-doped STO ceramics, which exhibit dielectric relaxations and harden soft-mode behaviour instead of the ferroelectric phase transition.

15 min. break

O 97.7 Thu 16:45 H 0104

Emergence of strain-Induced magnetism in plastically-deformed SrTiO3 at low temperature — •Anirban Kundu[1], Xi Wang[2], Avraham Klein[1], and Beena Kalisky[2] — [1]Department of Physics, Ariel University, Israel — [2]Institute of Nanotechnology & Advanced Materials, Bar-Ilan University, Israel

It is well established that SrTiO3 (STO) can possess ferroelectric states alongside observed superconducting states. However, so far, the phenomena of magnetism have not been established. In our collaborative work, in a plastically deformed bulk STO sample; SQUID measurements reveal strong magnetic signals which are completely absent in pristine samples. This strain-induced magnetism has two salient features. First, the magnetic moment is seen only with applied strain and increases with applied strain. Second, it also increases with temperature. Using Ginzberg Landau theory we show that these properties may be the result of coupling terms between strain, polar, and magnetic orders centered around dislocation walls induced by the plastic deformation. Our analysis implies that deformed STO is a quantum multiferroic.

O 97.8 Thu 17:00 H 0104

Mobility in $SrTiO_3$ Mediated by Machine Learning Predicted Anharmonic Phonons — •Luigi Ranalli[1], Carla Verdi[2], and Cesare Franchini[1] — [1]University of Vienna, Vienna, Austria — [2]University of Queensland: Brisbane, Queensland, Australia

The anharmonic corrections to ionic motion play a crucial role in influencing the electron-phonon interaction, a phenomenon typically addressed through harmonic dynamical matrices at the ground state. By combining machine learning methodologies [1] and the stochastic self-consistent harmonic approximation [2], we achieve a precise depiction of the temperature-dependent evolution of phonon frequencies and the onset of ferroelectricity in the quantum paraelectric perovskites $SrTiO_3$ [3] and $KTaO_3$ [4]. In this presentation, anharmonic dynamical matrices are incorporated into the Boltzmann transport equation calculations for $SrTiO_3$ up to 300K using the EPW code [5] and fixing the derivatives of the Kohn-Sham potential computed through density functional perturbation theory [6]. This approach yields a coherent interaction vertex, ensuring that the temperature-dependent ferroelectric soft mode explains and recovers the observed trend in experimental mobility, akin to the behavior observed in $KTaO_3$.

[1] R. Jinnouchi et al., Phys. Rev. Lett. 122 (2019) 225701
[2] L. Monacelli et al., J. Phys.: Condens. Matter 33 (2021) 363001
[3] C. Verdi et al., Phys. Rev. Materials 7 (2023) L030801
[4] L. Ranalli et al., Adv. Quantum Technol. 6 (2023) 2200131
[5] H. Lee et al., 10.1038/s41578-021-00289-w (2023)
[6] J. Zhou et al., Phys. Rev. Research 1 (2019) 033138

O 97.9 Thu 17:15 H 0104

Machine-learning-backed evolutionary exploration of the $SrTiO_3(110)$ surface phase diagram — •Ralf Wanzenböck[1], Florian Buchner[1], Michele Riva[2], Jesús Carrete[3,1], and Georg K. H. Madsen[1] — [1]Institute of Materials Chemistry, TU Wien, A-1060 Vienna, Austria — [2]Institute of Applied Physics, TU Wien, A-1040 Vienna, Austria — [3]Instituto de Nanociencia y Materiales de Aragón (INMA), CSIC-Universidad de Zaragoza, 50009 Zaragoza, Spain

We use Clinamen2, a modern functional-style Python implementation of the covariance matrix adaptation evolution strategy (CMA-ES), to gain insights into the lesser-known regions of the complex $SrTiO_3(110)$ surface phase diagram. To speed up the process, we leverage the transferability of a neural-network force field (NNFF) implemented on top of the state-of-the-art JAX framework.

Starting from smaller reconstructions in well-explored phases, such as the 4×1 surface reconstruction [Wanzenböck et al., Digit Discov 1, 703-710 (2022)], the NNFF is iteratively refined using an active learning workflow that relies on uncertainty estimation techniques [Carrete et al., J. Chem. Phys 158, 204801 (2023)]. We show how this workflow and the underlying uncertainty metric lead to a flexible NNFF, highlighted by the exploration of out-of-sample $SrTiO_3(110)$-$(2 \times n)$ reconstructions.

O 97.10 Thu 17:30 H 0104

Quasiparticle and excitonic properties of monolayer $SrTiO_3$ — •Lorenzo Varrassi[1], Peitao Liu[2], and Cesare Franchini[1,3] — [1]Dipartimento di Fisica e Astronomia, Università di Bologna, Bologna — [2]Shenyang National Laboratory for Materials Science, Institute of Metal Research — [3]University of Vienna, Faculty of Physics and Center for Computational Materials Science, Vienna.

Recently, a breakthrough has been achieved with the synthesis of freestanding $SrTiO_3$ ultrathin films down to the monolayer limit[1]; its optical and excitonic properties remain however largely unexplored. This talk will provide insights on the quasiparticle and excitonic properties of monolayer $SrTiO_3$, employing many-body perturbation theory.

Our analysis[2] emphasizes the need to go beyond the diagonal GW approximation and include off-diagonal self-energy elements in order to obtain correct description of the orbital hybridizations. A fully satisfying description is achieved by treating non-locality in both exchange and correlation.

The optical properties are studied through the solution of the Bethe-Salpeter equation. We observe a significant enhancement of the excitonic effects with respect to the bulk phase, with a binding energy at the optical gap about four times greater. Furthermore, the two-dimensional polarizability at the long wavelength limit is dominated by two strongly bound excitonic peaks; their character is determined through the analysis of the excitonic wavefunctions.

[1] D. Ji et al., Nature 570 (2019) 87
[2] L. Varrassi et al., arxive:2303.14830 (2023)

O 97.11 Thu 17:45 H 0104

$SrTiO_3$: Thoroughly investigated but still good for surprises — •Annette Bussmann-Holder[1], Reinhard K. Kremer[1], Krystian Roleder[2], and Ekhard K. H. Salje[3] — [1]Max-Planck-Institute for Solid State Research, Heisenbergstr. 1, D-70569 Stuttgart, Germany — [2]Institute of Physics, University of Silesia, ul. 75 Pułku Piechoty 1, 41-500 Chorzów, Poland — [3]Department of Earth Sciences, University of Cambridge, Downing Street, Cambridge CB2 3EQ, United Kingdom

For decades $SrTiO_3$ is in the focus of research with seemingly neverending new insights regarding its ground state properties, its application potentials, its surface and interface properties, the superconducting state, the twin boundaries and domain functionalities, etc. Here, we focus on the already well-investigated lattice dynamics of STO and show that four different temperature regimes can be identified which dominate the elastic properties, the thermal conductivity and the birefringence. These regimes are the low-temperature quantum fluctuation dominated one, followed by an intermediate regime, the region of the structural phase transition at 105 K and its vicinity, and at high temperatures a regime characterized by precursor and saturation effects. They can all be elucidated by lattice dynamical aspects. The relevant temperature dependencies of the soft modes are discussed and their relationship to lattice polarizability is emphasized.

O 98: Members' Assembly

Topics:
- Report of the Chairman
- Election of a new Vice-Chair
- Presentation of the Gerhard Ertl Young Investigator Award
- Miscellaneous

Time: Thursday 19:00–19:30 Location: H 0105

All members of the Surface Science Division are invited to participate.

O 99: Post-Deadline Session

Time: Thursday 19:30–20:30 Location: H 0105

Contributed Post-Deadline Talks

O 100: Overview Talk Marcel Reutzel

Time: Friday 9:30–10:15 Location: HE 101

Invited Talk O 100.1 Fri 9:30 HE 101
Momentum microscopy & 2D materials: Excitons in space and time — •Marcel Reutzel — I. Physikalisches Institut, Georg-August Universität Göttingen, Germany

The isolation of atomically thin van-der-Waals materials and their stacking to artificial heterostructures enables unprecedented possibilities for material control. Because of the 2D nature of the materials, surface science techniques have strongly contributed to the advancement of the field. Here, I will focus on the model system of semiconducting transition metal dichalcogendies (TMDs) and discuss how femtosecond momentum microscopy - a new variant of time- and angle-resolved photoemission spectroscopy - is capable of studying excitonics in space and time. I will show how the formation and thermalization dynamics of bright and dark electron-hole pairs (excitons) can be probed on the femtosecond time- and the nanometer length-scale.

First, I will provide an overview on how the momentum-resolved photoelectron detection scheme facilitates the direct access to the energy landscape and dynamics of bright and dark excitons in monolayer and twisted heterobilayer TMDs. Second, by combining momentum microscopy with dark-field imaging techniques, I will outline how distinct features of the photoemission spectral function can be imaged on the nanoscale, facilitating, e.g., access to the ultrafast exciton formation dynamics in laterally inhomogeneous TMD heterostructures.

O 101: Focus Session: $SrTiO_3$: A Versatile Material from Bulk Quantum Paraelectric to 2D Superconductor III (joint session TT/KFM/MA/O)

Strontium titanate ($SrTiO_3$) is a paradigmatic material that plays an important role in various fields of solid-state physics, surface science and catalysis: The pure bulk phase is a wide-band-gap semiconductor that upon cooling becomes a textbook quantum paraelectric. When slightly doped, $SrTiO_3$ turns into a Fermi-liquid-type metal that becomes superconducting at extremely low charge carrier density. $SrTiO_3$-based surfaces and interfaces host un-conventional electronic states such as quasi-two-dimensional electron liquid, magnetism and superconductivity. Despite intensive studies over the past decades, $SrTiO_3$ continues to reveal surprising new phenomena that challenge the established views on this material. To this end achieving light-induced nonequilibrium states and the recent preparation of a 2D oxide based on $SrTiO_3$ opens new playgrounds for research. This Focus Session will present exciting developments in the study of electronic states that are based on the peculiar properties of $SrTiO_3$.

Please note that this Focus Session comprises four parts: Posters are presented within the TT poster session TT58 (Wed 15:00-18:00, poster area E). Invited talks are compiled in the session TT62 (Thursday, 9:30 to 12:45, H0104), Contributed talks will be presented in sessions TT72 (Thursday 15:00-18:00, H0104) and TT83 (Fri 9:30-12:30, H0104).

Organizers: Rossitza Pentcheva, University of Duisburg-Essen, Marc Scheffler, University of Stuttgart

Time: Friday 9:30–12:30 Location: H 0104

O 101.1 Fri 9:30 H 0104
High-mobility two-dimensional electron gases based on strain engineered ferroelectric $SrTiO_3$ thin films — •Ruchi Tomar[1], Tatiana Kuznetsova[2], Srijani Mallik[1], Luis M. Vicente-Arche[1], Fernando Gallego[1], Maximilien Cazayous[3], Roman Engel-Herbert[2,4], and Manuel Bibes[1] — [1]Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767 Palaiseau, France. — [2]Pennsyvlania State University, University Park, PA 16802, USA. — [3]Laboratoire Matériaux et Phénomènes Quantiques (UMR 7162 CNRS), Université de Paris, 75205 Paris Cedex 13, France. — [4]Paul Drude Institute for Solid State Electronics, Leibniz Institute within Forschungsverbund Berlin eV, Hausvogteiplatz 5-7, 10117, Berlin, Germany.

Two-dimensional electron gases (2DEGs) based on the quantum paraelectric $SrTiO_3$ display fascinating properties such as large electron mobilities, superconductivity, and efficient spin-charge interconversion owing to their Rashba spin-orbit coupling. Here, we use oxide molecular beam epitaxy to grow high-quality strain-engineered $SrTiO_3$ films that are ferroelectric up to 170 K. We then generate a 2DEG by sputtering a thin Al layer and demonstrate an increase in mobilities compared to earlier literature. Furthermore, through Raman spectroscopy and magneto-transport measurements, we show that the ferroelectric character is retained after 2DEG formation. These results thus qualify our samples as ferroelectric 2DEGs up to temperatures well above previous results based on Ca-$SrTiO_3$ substrates, opening the way towards ferroelectric 2DEGs operating at room temperature.

O 101.2 Fri 9:45 H 0104
Two-dimensional electron liquids at truly bulk-terminated $SrTiO_3$ — •Igor Sokolović[1,2], Eduardo B. Guedes[3], Thomas van Waas[4], Samuel Poncé[4,5], Craig M. Polley[6], Michael Schmid[2], Ulrike Diebold[2], Milan Radović[3], Martin Setvín[2,7], and J. Hugo Dil[3,8] — [1]Institute of Microelectronics, TU Wien, Vienna, Austria — [2]Institute of Applied Physics, TU Wien, Vienna, Austria — [3]Photon Science Division, PSI, Villagen, Switzerland — [4]ETSF, Institute of Condensed Matter and Nanosciences, UCLouvain, Louvain-la-Neuve, Belgium — [5]WEL Research Institute, Wavre, Belgium — [6]MAX IV laboratory, Lund University, Lund, Sweden — [7]Faculty of Mathematics and Physics, Charles University, Prague , Czech Republic — [8]Institut de Physique, ÉPFL, Lausanne, Switzerland

A truly bulk-terminated $SrTiO_3(001)$ surface prepared by cleaving *in situ* was investigated with angle-resolved photoemission spectroscopy (ARPES) and noncontact atomic force microscopy (ncAFM). The (1×1) $SrTiO_3(001)$ surfaces were achieved through our cleaving procedure that exploits the strain-induced ferroelectric transition in $SrTiO_3$, and provides both possible surface terminations, TiO_2 and SrO. Each hosts a specific two-dimensional electron liquid (2DEL): the first with split and the other with degenerate bands. The origin of the 2DELs and the band-splitting mechanisms are elucidated by correlating the observed reciprocal- and real-space electronic and atomic structure.

O 101.3 Fri 10:00 H 0104
Low-energy excitations at $SrTiO_3(001)$ surfaces in absence and presence of a two-dimensional electron gas — •Hannes Herrmann, Anne Oelschläger, and Wolf Widdra — Martin-Luther-Universität Halle-Wittenberg, 06120 Halle, Germany

The low-energy excitations of $SrTiO_3$, a large-bandgap oxide perovskite, are dominated by phonons and phonon polaritons. At the surface they couple to dipole-active surface phonon polaritons that are bound to the $SrTiO_3$-vacuum interface. These excitations can be addressed by surface vibrational spectroscopy techniques as, e.g., high-

resolution electron energy loss spectroscopy (HREELS).
Here we will present HREELS studies that identify all $SrTiO_3$(001) dipole-active excitations, including their specific line shapes and will discuss the electron-phonon coupling to a two-dimensional electron gas. The latter are prepared with variable charge-carrier concentrations either by annealing under ultrahigh-vacuum condition or by growth of an ultrathin layers of EuO on top. With formation of the 2DEGs, the discrete surface phonon polaritons couple to the electron-hole pair continuum as is witnessed by a substantial line broadening and asymmetric Fano-like line shapes. A quantitative description that accounts for all details of the line shape paves the way for an in-situ analysis of the 2DEG charge carrier dynamics.

O 101.4 Fri 10:15 H 0104

Confined ionic-electronic systems based on $SrTiO_3$ — •Felix Gunkel, Marcus Wohlgemuth, Moritz L. Weber, and Regina Dittmann — Peter Gruenberg Institute, Forschungszentrum Juelich

$SrTiO_3$ reflects a a prototype ionic-electronic oxide, in which the physical properties are significantly affected by the ionic defect structure. [Gunkel et al., APL 2020] At the same time, spatial confinement of electronic charge carries led to unexpected electronic and magnetic phenomena, including 2DEG formation, magnetoresistance and localization phenomena. Here we will discuss, how spatial confinement also affects the ion-dynamics and defect-equilibria of SrTiO3, yielding interfacial defect structures and ion-dynamics that significantly differ from the bulk. [Rose et al., Adv. Mater. (2023); Weber et al., Nature Mater., to be published (Jan 2 2024)]. New opportunities to tailor such confined ionic-electronic systems arise from synthesis advances in generating transferable, free-standing $SrTiO_3$ sheets. These reflect ideally-confined nanosheets of $SrTiO_3$ and can serve as model system for ionic-electronic confinement phenomena as well as template for the synthesis of functional bilayer structures. We discuss the state-of-the-art of controlled bilayer synthesis and derive the required finite-size corrections in the thermodynamic description of the defect chemistry of $SrTiO_3$, indicating that the average reduction enthalpy of $SrTiO_3$ can be effectively reduced via confinement.

O 101.5 Fri 10:30 H 0104

Origin of spin-polarized 2DEG at the $EuTiO_3$(001) surface and $LaAlO_3/EuTiO_3/SrTiO_3$(001) interface — •Manish Verma and Rossitza Pentcheva — Department of Physics, Universität Duisburg-Essen

Since the discovery of a two-dimensional electron gas (2DEG) at the interface between the $LaAlO_3$ and $SrTiO_3$ band insulators, studies on oxide surfaces and interfaces uncovered an intriguing and rich physics, such as possible magnetism in 2DEG. Using density functional theory with an on-site Coulomb repulsion term U, we find a spin-polarized 2DEG at the $EuTiO_3$(001) surface arising from the interplay of ferromagnetic (FM) order of Eu-$4f$ magnetic moments and the localization of electrons released from oxygen divacancies at the surface Ti sites, in agreement with in situ high-resolution angle-resolved photoemission [1]. The 2DEG at the $LaAlO_3/EuTiO_3/SrTiO_3$(001) interface is formed due to the polar discontinuity. The spin-polarization is due to the FM exchange interaction between Eu $4f$ and Ti $3d$ states and steers the occupation of d_{xz}/d_{yz} orbitals [2].
[1] R. Di Capua *et al.*, Phys. Rev. Research **3** (2021) L042038
[2]. R. Di Capua *et al.*, npj Quantum Mater. **7** (2022) 41

O 101.6 Fri 10:45 H 0104

A multiferroic STO-based 2D-electron gas — •Marco Salluzzo[1], Yu Chen[1], Martando Rath[1], Daniela Stornaiuolo[2], Julien Brehin[3], Manuel Bibes[3], Julien Varignon[4], and Cinthia Piamonteze[5] — [1]Cnr-Spin Complesso Monte S. Angelo via Cinthia 80126, Napoli, Italy — [2]Università "Federico II" di Napoli, Dipartimento di Fisica "Ettore Pancini", Complesso Monte S. Angelo via Cinthia 80126, Napoli, Italy — [3]Unité Mixte de Physique, CNRS, Thales, Université Paris Saclay, Palaiseau, France — [4]Crismat, CNRS , Ensicaen, Normandie Université, Caen, France — [5]Swiss Light Source, Paul Scherrer Institut, Villigen, Switzerland.

The fabrication of artificial materials combining different functional properties is a powerful method to create novel quantum states. Here we demonstrate the realization of a 2D electron gas exhibiting a coexistence of ferroelectric and ferromagnetic order parameters, by heteroepitaxy.

The novel 2DEG is realized by inserting few unit cells of the antiferromagnetic insulator $EuTiO_3$ between a $LaAlO_3$ band insulating thin film (10 unit cells) and a Ca-doped $SrTiO_3$ single crystal.

By using Ti-$L_{2,3}$ and Eu $M_{4,5}$ edges x-ray linear dichroism and x-ray magnetic circular dichroism, we provide evidences of a switchable polarization, non-volatile tuning of Ti3d orbital splitting, and of a modulation of Eu-4f magnetic moment of the 2DEG by the FE-polarization[1]. The result is of interest for quantum spin-orbitronic applications.
[1] J. Bréhin et al., Nat. Phys. 19 (2023) 823

O 101.7 Fri 11:00 H 0104

Magnetotransport properties of a spin polarized STO-based 2D electron system tuned by visible light — Maria D'Antuono[1,2], Yu Chen[2], Roberta Caruso[1,2,3], Benoit Jouault[4], Marco Salluzzo[2], and •Daniela Stornaiuolo[1,2] — [1]Department of Physics, University of Naples Federico II, Italy. — [2]CNR-SPIN, Naples, Italy. — [3]Condensed Matter Physics and Materials Science Division, Brookhaven National Laboratory, NY, USA. — [4]Laboratoire Charles Coulomb, CNRS,Université de Montpellier, France

Two-dimensional electron systems (2DES) developing in STO-based heterostructures possess a wide range of properties which are largely tunable thanks to the systems band structure and carrier density. In $LaAlO_3/EuTiO_3/SrTiO_3$ (LAO/ETO/STO) heterostructure, for instance, the charge carriers, above a critical value, start to fill Ti-3d bands with $d_{xz,yz}$ character, leading to the stabilization of a ferromagnetic order of Ti and Eu magnetic moments, and to a spin polarization of the 2DES. In this work we show that such mechanism can be achieved not only using electric field effect, but also using visible light irradiation. Furthermore, the analysis of the Anomalous Hall effect and of magnetocodnuctance curves demonstrate that visible light irradiation leads to enhanced stabilization of ferromagnetic correlations in the 2DES. Our results establishs the combined use of visible light and gate voltage as a straightforward way to access unexplored regions of the LAO/ETO/STO 2DES phase diagram.

15 min. break

O 101.8 Fri 11:30 H 0104

All-electrical measurement of the spin-charge conversion effect in nanodevices based on $SrTiO_3$ two-dimensional electron gases — •Fernando Gallego[1], Felix Trier[1,2], Srijani Mallik[1], Julien Brehin[1], Sara Varotto[1], Luis Moreno[1], Tanay Gosavy[3], Chia-Ching Lin[3], Jean-René Coudevylle[4], Lucía Iglesias[1], Fèlix Casanova[5,6], Ian Young[3], Laurent Vila[7], Jean-Philippe Attané[7], and Manuel Bibes[1] — [1]Unité Mixte de Phys, CNRS-Thales, Univ. Paris-Saclay, 91767 Palaiseau, France. — [2]Dept of Energy Conservation and Storage, Univ. of Denmark, 2800 Kgs. Lyngby, Denmark. — [3]Comp. Res. Intel Corp., Hillsb., OR 97124, USA. — [4]Centre de Nanosciences et de Nanotech., CNRS, Université Paris-Sud, Université Paris-Saclay, France. — [5]CIC nanoGUNE BRTA, 20018 Donostia, Spain. — [6]IKERBASQUE, Basque Foundation for Science, 48009 Bilbao, Spain. — [7]Univ. Grenoble Alpes, CNRS, CEA, SPINTEC, Grenoble, France.

We report all-electrical spin-injection and spin-charge conversion experiments in nanoscale devices harnessing the inverse Edelstein effect of $SrTiO_3$ 2DEGs. We have designed, patterned and fabricated nanodevices in which a spin current injected from a cobalt layer into a $LaAlO_3/SrTiO_3$ 2DEG is converted into a charge current. We optimized the spin-charge conversion signal by back-gating. We further disentangled the inverse Edelstein contribution from spurious effects. The combination of non-volatility and high energy efficiency of these devices could potentially lead to new technology paradigms for beyond-CMOS computing architectures.

O 101.9 Fri 11:45 H 0104

Effect of confinement and coulomb interactions on the electronic structure of the (111) $LaAlO_3/SrTiO_3$ interface — •Mattia Trama[1,2,3], Vittorio Cataudella[4,5], Carmine Antonio Perroni[4,5], Francesco Romeo[1], and Roberta Citro[1,2] — [1]Universitá degli Studi di Salerno, Fisciano, Italy — [2]INFN Sezione di Napoli, Naples, Italy — [3]Institute for Theoretical Solid State Physics, IFW Dresden, Dresden, Germany — [4]Universitá degli Studi di Napoli Federico II, Naples, Italy — [5]CNR-SPIN Napoli Unit, Naples, Italy

A tight-binding supercell approach is used for the calculation of the electronic structure of the (111) $LaAlO_3/SrTiO_3$ interface. The confinement potential at the interface is evaluated solving a discrete Poisson equation by means of an iterative method. In addition to the effect of the confinement, local Hubbard electron-electron terms are included at the mean-field level within a fully self-consistent procedure. The calculation carefully describes how the two dimensional electron gas arises from the quantum confinement of electrons near the interface due to the band bending potential. The resulting electronic sub-bands and Fermi surfaces show full agreement with the electronic structure determined by angle-resolved photoelectron spectroscopy experiments. In particular, we analyse how the effect of local Hubbard interactions change the density distribution over the layers from the interface to the bulk. Interestingly, the two-dimensional electron gas at the interface is not depleted by local Hubbard interactions which indeed induce an enhancement of the electron density between the first layers and the bulk.

O 101.10 Fri 12:00 H 0104

Enhanced Non-linear Response by Manipulating the Dirac Point in the (111) $LaTiO_3/SrTiO_3$ Interface — •Yoram Dagan, Gal Tuvia, Amir Burshtein, Itai Silber, Amnon Aharony, Ora Entin-Wohlman, and Moshe Goldstein — School of Physics and Astronomy, Tel Aviv University

Tunable spin-orbit interaction (SOI) is an important feature for future spin-based devices. In the presence of a magnetic field, SOI induces an asymmetry in the energy bands, which can produce non-linear transport effects ($V \sim I^2$). Here, we focus on such effects to study the role of SOI in the (111) $LaTiO_3/SrTiO_3$ interface. This system is a convenient platform for understanding the role of SOI since it exhibits a single-band Hall-response through the entire gate-voltage range studied. We report a pronounced rise in the non-linear longitudinal resistance at a critical in-plane field H_{cr}. This rise disappears when a small out-of-plane field component is present. We explain these results by considering the location of the Dirac point formed at the crossing of the spin-split energy bands. An in-plane magnetic field pushes this point outside of the Fermi contour, and consequently changes the symmetry of the Fermi contours and intensifies the non-linear transport. An out-of-plane magnetic field opens a gap at the Dirac point, thereby significantly diminishing the non-linear effects. We propose that magnetoresistance effects previously reported in $SrTiO_3$-based interfaces could be comprehended within our suggested scenario.

O 101.11 Fri 12:15 H 0104

Tunable 2D Electron- and 2D Hole States Observed at $Fe/SrTiO_3$ Interfaces — •Pia Maria Düring[1], Paul Rosenberger[1,2], Lutz Baumgarten[3], Fatima Alarab[4], Frank Lechermann[5], Vladimir N. Strocov[4], and Martina Müller[1] — [1]FB Physik, Universität Konstanz, 78457 Konstanz — [2]TU Dortmund, 44221 Dortmund — [3]FZ Jülich, PGI-6, 52425 Jülich — [4]PSI, SLS, CH-5232 Villingen — [5]TP III, RU Bochum, 44780 Bochum

Oxide electronics provide the key concepts and materials for enhancing silicon-based semiconductor technologies with novel functionalities. However, a crucial property of semiconductor devices remains undisclosed in their oxide counterparts: the ability to set or even switch between negatively (n) charged electrons or positively (p) charged holes. Using resonant angle-resolved photoelectron spectroscopy, we provide direct evidence for individually emerging n- or p-type 2D band dispersions in $SrTiO_3$ (STO)-based heterostructures [1]. The key to setting the carrier character is the oxidation state of a Fe-based interface layer: For Fe and FeO, hole bands emerge in the empty band gap region of STO, while for Fe_3O_4 overlayers, an 2D electron system (2DES) is formed. Unexpected oxygen vacancy characteristics arise for the hole-type interfaces, which as of yet had been exclusively assigned to the emergence of 2DESs. In general, this study unveils the potential to seamlessly alter the conductivity type at STO interfaces by manipulating the oxidation state of a redox overlayer.

[1] P. M. Düring et al., Adv. Mater. (accepted)

O 102: Focus Session: Wetting on Adaptive Substrates III (joint session CPP/DY/O)

The focus session aims to discuss recent developments in the wetting dynamics of adaptive, deformable, and switchable surfaces.

Time: Friday 9:30–12:45 Location: H 0110

Invited Talk O 102.1 Fri 9:30 H 0110

Condensation on soft substrates — •Ambre Bouillant[1,3], Bruno Andreotti[2], and Jacco H. Snoeijer[3] — [1]Laboratoire MSC, CNRS UMR 7057, Université Paris Cité, FR — [2]Laboratoire LPENS, CNRS UMR 8023, Université PSL, FR — [3]Physics of Fluids, Twente Unversity, NL

Vapor molecules can nucleate on cool substrates, provided the surrounding humidity is high enough. Dew formation has been investigated on both rigid (and rough!) solids as well as on liquids. However, how substrate elasticity affects the condensation process remains elusive. In this talk, I will present how water condenses on soft, elastic gels that are smooth at the nanometer scale. We prepare PDMS gels whose softness varies between that of a rigid substrate and an un-crosslinked polymeric liquid.

Although elasticity should be marginal at the nanometric scale at which drops form, we report that the nuclei density is highly sensitive to the substrate softness. Throughout this talk, I will delve into the intricate dynamics of condensation and try to explain some of the intriguing characteristics we have observed. Among these are the influence of softness on nucleation; the sub-diffusive growth of droplets; and the absence of secondary nucleation events (unlike what is classically observed in heterogeneous nucleation). Later, when neighboring drops get closer, they attract each other due to interactions mediated by substrate deformations. Drops then gather into clusters that seem reluctant to coalesce. This ultimately results in the formation of a persistent, ordered, honeycomb-patterned liquid film.

O 102.2 Fri 10:00 H 0110

Modelling droplets on substrates with travelling-wave deformations — •Josua Grawitter and Holger Stark — Technische Universität Berlin, Institut für Theoretische Physik, Hardenbergstr. 36, 10623 Berlin, Germany

Motivated by strategies for targeted microfluidic transport of droplets, we investigate how sessile droplets can be steered toward a preferred direction using travelling-wave deformations of the substrate. To perform our numerical study, we develop a formalism to apply the boundary-element method to dynamic wetting. It solves the governing Stokes equations for the fluid flow field inside the droplet that is pushed forward by the deforming substrate.

We find two distinct modes of droplet motion. For small wave speed the droplet surfs with a constant velocity on the wave, while beyond a critical wave speed a periodic wobbling motion occurs, the period of which diverges at the transition. In the related case of a flat substrate with travelling waves in wettability, such an observation is well rationalized by the *nonuniform oscillator* model and the transition described by a SNIPER bifurcation. Here, however, the mean droplet velocity in the wobbling state is proportional to the wave speed at large speed values since the droplet always has to move up and down. To rationalize this behavior, the nonuniform oscillator model has to be extended. Since the critical wave speed of the bifurcation depends on the droplet radius, this dependence can be used to sort droplets by size.

O 102.3 Fri 10:15 H 0110

High Voltages Generated by Moving Water Drops — •Stefan Weber[1,2], Pravash Bista[2], Aaron Ratschow[3], and Hans-Jürgen Butt[2] — [1]Institute for Photovoltaics, University of Stuttgart — [2]MPI for Polymer Research, Mainz — [3]Institute for Nano- and Microfluidics, TU Darmstadt

Water drops on insulating, hydrophobic substrates can generate electric potentials of kilovolts upon sliding for a few centimeters. We show that the drop saturation voltage corresponds to an amplified value of the solid-liquid surface potential at the substrate. The amplification is given by the substrate geometry, the drop and substrate dielectric properties and the Debye length within the liquid. Next to enabling an easy and low-cost way to measure surface- and Zeta potentials, the high drop voltages have implications for energy harvesting, droplet microfluidics and electrostatic discharge protection.

O 102.4 Fri 10:30 H 0110

Electro(de)wetting with Photoswitches: Control of wetting by electric fields and light — •Billura Shakhayeva and Björn Braunschweig — University of Münster, Institute of Physical Chemistry, 48149 Münster, Germany

Electro-dewetting (EDeW) was introduced as a new method in order to change the wetting properties of surfaces. The underlying mechanism is, however, not fully understood and additional experiments are needed. For that, we have replaced the DTAB surfactants used in the original work [1] by arylazopyrazole triethylammonium bromide (AAP-TB) which is a cationic surfactant that can change the surface tension at the air-water interface to a large extend through E/Z photoisomerization [2]. This offers to fine tune the contact angle during EDeW by E/Z photoisomerization of AAP-TB and we find a further increase in contact angle by ∼6° when the samples were irradiated by UV light that triggers photoisomerization from the E to the less surface-active Z isomer. Li et al.[1] suggested that surfactants are deposited on the silicon oxide surface through the EDeW process. In order to study the possible deposition of surfactants and the role of a possible prewetting layer outside of the drop and at some distance from the 3-phase contact line we have done sum-frequency generation (SFG) spectroscopy and find that surfactants are deposited even several mm adjacent to the drop and that their structure as well as the structure of the accompanying water layer changes drastically close to the 3-phase contact line. [1] Li et al. Nature, 572, 507-510 (2019) [2] Schnurbus et al. J. Phys. Chem. B 124, 6913 (2020).

O 102.5 Fri 10:45 H 0110

Light-Triggered Manipulations of Droplets All in One: Reversible Wetting, Transport, Splitting, and Merging — •Maren Umlandt, Nino Lomadze, and Svetlana Santer — University of Potsdam, Potsdam, Germany

We report on light-triggered droplet manipulation such as reversible wetting, splitting, merging, and transport [1]. The unique feature is that the changes in the wetting properties of anisotropic liquids adsorbed on photo-switchable films can be triggered by application of optical stimuli, which lead to changes in the morphology of the surfaces. The adaptive films consist of an azobenzene-containing surfactant attached to oppositely charged polymer chains. Under exposure to irradiation with light, the azobenzene photo-isomerizes between trans- and cis-states, resulting in changes in surface energy and orientation of surfactant tails in film [2]. The increase in the surface temperature due to absorption of light by the azobenzene groups [3] enables diverse processes of droplet manipulation. Using a moving light spot, we demonstrate the locomotion of the droplet over macroscopic distances. Our findings could lead to the application of a programmable workbench for manipulating and operating an ensemble of droplets.

We thanks for funding DFG and RFBR.

[1] Umlandt, M. et al. ACS Applied Materials & Interfaces 2022 14 (36), 41412-41420 [2] Arya, P. et al. J. Chem. Phys. 2020, 152, 024904 [3] Zakharov, A. V.et al. Phys. Rev. E 2017, 96, 052705

15 min. break

Invited Talk O 102.6 Fri 11:15 H 0110

Of Singularities and Controversies: The Soft Wetting Enigma — •Stefan Karpitschka — University of Konstanz

The surface mechanics of soft solids are ubiquitously important in nature and technology. Static and dynamic wetting of soft polymer gels by simple liquids has emerged as an archetypical model system: The singular capillary traction at the edge of a droplet creates a microscale wetting ridge which slows down dramatically the macroscopic dynamics. Capillary, elastic, viscous and osmotic effects all contribute simultaneously in a highly non-linear regime, which has sparked controversial discussions of the underlying theoretical description. In this talk I will present recent measurements on ultra-thin to ultra-soft materials, along with new theoretical and numerical results, scrutinizing sources of non-linear behavior in both statics and dynamics.

O 102.7 Fri 11:45 H 0110

Dynamics of Moving Droplets on Lubricated Polymer Brushes — •Rodrique Badr[1], Lukas Hauer[2,3], Doris Vollmer[3], and Friederike Schmid[1] — [1]Johannes Gutenberg University, Mainz — [2]Humboldt-Universität zu Berlin, Berlin — [3]Max Planck Institute for Polymer Research, Mainz

The interaction of liquid droplets with lubricated substrates is rich with interesting physics combining thermodynamics, polymer science, and fluid dynamics, and has the potential for various industrial applications. In this work we focus on the dynamics of droplets moving on lubricated polymer brushes. In previous work, we showed the existence of a cloaking transition where the lubricant covers the surface of the droplet at equilibrium. Here, we investigate the influence of this cloak on the dynamics, in addition to its properties during the dynamical steady state. In addition, we investigate the characteristic of the wetting ridge, as well as the flow of the droplet, and quantify the friction forces felt by the droplet during its motion.

O 102.8 Fri 12:00 H 0110

Modeling the growth of biofilms on soft substrates — •Anthony Pietz[1], Karin John[2], and Thiele Uwe[3] — [1]Institute for theoretical physics, University of Münster — [2]Institute for theoretical physics, University of Münster — [3]Research Associate at the CNRS Laboratoire Interdisciplinaire de Physique LIPhy Grenoble - France

We investigate the influence of substrate softness on biofilm growth amending the thin-film model developed by Trinschek et al for rigid solid substrates [1] by the inclusion of a simple description of an elastic substrate [2]. Wettability (described in the mesoscopic model by a wetting energy) is a key factor in the transition between arrested and continuous spreading on rigid substrates [1]. Our focus are changes in the spreading process with changing character of the substrate studied by time simulations of 2d drops/biofilms at fixed surface tension and initial drop volume. We find that with increasing softness from rigid via elastic to liquid-like substrate the spreading velocity of the biofilm decreases at fixed biofilm growth rate and wettability. Further, we discus how these changes depend on wettability and growth rate. In particular, we show that the transition between arrested and continuous spreading is for increasing softness shifted towards larger wettability and larger growth rate.

[1] S. Trinschek, K. John, S. Lecuyer, and U. Thiele, Phys. Rev. Lett. 119, 078003 (2017).

[2] C. Henkel, J. H. Snoeijer, and U. Thiele, Soft Matter 17, 10359 (2021).

O 102.9 Fri 12:15 H 0110

Chemically Active Wetting — •Susanne Liese[1], Xueping Zhao[2], Christoph Weber[1], and Frank Jülicher[3] — [1]Faculty of Mathematics, Natural Sciences, and Materials Engineering: Institute of Physics, University of Augsburg, Augsburg, Germany — [2]Department of Mathematical Sciences, University of Nottingham Ningbo China — [3]Max Planck Institute for the Physics of Complex Systems, Dresden, Germany

Wetting of liquid droplets on passive surfaces is ubiquitous in our daily lives, and the governing physical laws are well-understood. When surfaces become active, however, the governing laws of wetting remain elusive. Here we derive the non-equilibrium thermodynamic theory for active wetting, where the surface is active due to a binding process that is maintained away from equilibrium. We show that active binding fundamentally changes the wetting behavior, leading to steady, non-equilibrium states with droplet shapes reminiscent of a pancake or a mushroom. The origin of such anomalous shapes can be explained by mapping to electrostatics, where pairs of binding sinks and sources correspond to electrostatic dipoles along the triple line. This is an example of a more general analogy, where localized chemical activity gives rise to a multipole field of the chemical potential. The underlying physics is relevant for cells, where droplet-forming proteins can bind to membranes accompanied by the turnover of biological fuels.

O 102.10 Fri 12:30 H 0110

Intracellular wetting between biomembranes and liquid-like condensates — •Lukas Hauer[1], Katharina Sprobeck[1], Amir Houshang Bahrami[2,3], and Roland L. Knorr[1,4] — [1]Humboldt Univeristät zu Berlin — [2]Bilkent University, Ankara — [3]MPI-DS, Göttingen — [4]University of Tokyo

Wetting has been recently identified as physiologically important in fundamental cellular processes: phase-separated condensates (e.g., proteins and RNA) form liquid droplets in cells and interact with membranes, e.g., during autophagy in eukaryotic cells or protein storage in plant vacuoles. Upon contact, the droplets can exert wetting forces on the membrane that deforms. This creates a competition of mechanical forces of the membrane elasticity and the droplet capillarity, giving rise to elastocapillary phenomena. In this talk, I will present a minimal model system comprising giant lamellar vesicles (GUVs) filled with aqueous phase-separating polymers(PEG/Dextran). We create liquid-liquid interfaces inside GUVs by osmotic quenches, yielding deformed vesicles with excess membrane area. The excess membrane accumulates at the liquid-liquid interface and assumes differing morphologies, ranging from micro-tubules to sheets, to stomatocytes. We find that the morphology transition depends on the liquid-liquid surface tension. Our results will help to explain resembling in vivo observations during the morphogenesis of protein storage vacuoles in plants.

O 103: Focus Session: Molecular Nanostructures on Surfaces: On-Surface Synthesis and Single-Molecule Manipulation IV

Time: Friday 10:30–13:00 Location: HE 101

Topical Talk O 103.1 Fri 10:30 HE 101

Toward nanofabrication with molecular building blocks — •Christian Wagner — Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, 52425 Jülich, Germany

The control of single-molecule junctions with a scanning probe microscope not only directly benefits molecular electronics and molecular machines, but is also a stepping stone to the more ambitious goal of (supra)molecular nanofabrication. In my talk I will describe key aspects of controlled molecular manipulation. Perhaps the biggest challenge is the lack of information about the atomic configuration of a tip-molecule-surface junction during manipulation. We have developed strategies to monitor this configuration in real time during manipulation or to circumvent the problem using concepts such as reinforcement learning. Precise knowledge of the molecule-surface interactions arising from van der Waals forces and local chemical bonding is central to this endeavor, and I will present our efforts to probe these interactions via molecular manipulation. Appropriate models are another key component in understanding and controlling the manipulation process. I will outline our modeling strategies, ranging from machine-learned models trained on DFT data to models based entirely on experimental observations. Finally, I will discuss scanning quantum dot microscopy, an application enabled by single-molecule manipulation, and report surface potential measurements on atomic chains.

O 103.2 Fri 11:00 HE 101

Faster Single-Molecule Manipulations by STM Tunneling Current Analysis with Neural Networks — •Michael Obermayr, Bernhard Ramsauer, and Oliver T. Hofmann — Institute of Solid State Physics, NAWI Graz, Graz University of Technology, Petersgasse 16, 8010 Graz, Austria

Scanning tunneling microscopy (STM) allows precise manipulation of single atoms and molecules on surfaces. Recent AI-driven advancements in manipulating arbitrary molecules [1] open the door to automatic assembly of artificial nanostructures. A significant time portion is required for the imaging before and after each manipulation step, which often acts as the speed bottle neck. In this work we train a neural network on the tunneling current to predict the shift in position and orientation of an object, limiting the need for constant imaging.

The challenge in training a neural network lies in obtaining a suitable dataset containing all necessary inputs and outputs. Here we start with a measurement series of single-molecule manipulations paired with the respective precedent and subsequent images of the surface. The action outcome is extracted from these images with machine vision. Thus, a suitable training dataset for neural networks can be generated systematically, while ensuring broad applicability towards arbitrary molecules.

The fully trained network allows to track the manipulated molecule despite skipping a significant fraction of the imaging steps, which leads to an accelerated manipulation process.

[1] B. Ramsauer et al., J. Phys. Chem. A, 127, 2041 (2023)

O 103.3 Fri 11:15 HE 101

Tip-induced and thermally activated rotation of bowl-shaped bridged-triphenylamine derivatives on Au(111) — •Sajjan Mohammad[1], Milan Kivala[2], Meike Stöhr[1], and Sabine Maier[1] — [1]Department of Physics, Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany — [2]Institute of Organic Chemistry, University of Heidelberg, Germany

We studied the adsorption and rotation under external excitation of bowl-shaped bridged-triphenylamine derivatives on Au(111) using low-temperature scanning probe microscopy. The primarily observed three-lobed structure suggests that individual bridged-triphenylamine molecules adsorb in a bowl-up conformation, with the central atom anchoring to the Au(111) surface and the three benzene rings suspended above it. The onset and mechanism of rotation were examined through STM tip manipulation experiments and thermal activation. Our results demonstrate that the central atom of a bridged triphenylamine acts as an effective axle, ensuring sufficient anchoring to prevent diffusion and facilitating low rotational barriers for single molecule rotors on a metal surface.

O 103.4 Fri 11:30 HE 101

Thermal with electronic excitation for unidirectional rotation of molecule on surface — •Kwan Ho Au-Yeung[1,2], Suchetana Sarkar[1], Christian Joachim[3], and Francesca Moresco[1] — [1]Center for Advancing Electronics Dresden, TU Dresden, 01062 Dresden, Germany — [2]Physikalisches Institut, Karlsruhe Institute of Technology, 76131Karlsruhe, Germany — [3]GNS & MANA Satellite, CEMES, CNRS, 29 rue J. Marvig, 31055 Toulouse, France

Exploring the limits of the microscopic reversibility principle, we investigated the interplay between thermal and electron tunneling excitations for the unidirectional rotation of a molecule-rotor on the Au(111) surface. We identified a range of moderate voltages and temperatures where heating the surface enhances the unidirectional rotational rate of a chemisorbed DMNI-P rotor. When manipulated by voltage pulses, the rotational processes are predominantly governed by inelastic tunneling effects. Conversely, at increased temperatures, the rotation adopts a stochastic character. At each electron transfer event during tunneling, the quantum mixing of ground and excited electronic states brings part of the thermal energy in the excited electronic states of the rotor.

O 103.5 Fri 11:45 HE 101

Tuning the planarity of a thiantherene-based molecule on Au(111) — Kwan Ho Au-Yeung[1], Suchetana Sarkar[1], Sattwick Haldar[2], •Pranjit Das[1], Dmitry A. Ryndyk[3,4], Andreas Schneemann[2], and Francesca Moresco[1] — [1]Center for Advancing Electronics Dresden, TU Dresden, 01062 Dresden, Germany — [2]Inorganic Chemistry I, TU Dresden, 01069 Dresden, Germany — [3]Institute for Materials Science, TU Dresden, 01062 Dresden, Germany — [4]Theoretical Chemistry, TU Dresden, 01062 Dresden, Germany

We conducted a comprehensive study on the adsorption geometries of a non-planar aromatic thianthrene-based molecule on the Au(111) surface by scanning tunneling microscopy. The molecules demonstrate two self-assembled structures depending on the orientation of the C-S-C bonds: close-packed islands where they are non-planar and quasi one-dimensional chains where they are planar. Using the STM tip to isolate a molecule from the island, it is observed that the non-planar molecules transform into the planar configuration.

O 103.6 Fri 12:00 HE 101

Flexible Superlubricity Unveiled in Sidewinding Motion of Individual Polymeric Chain — •J.G. Vilhena[1,2], Rémy Pawlak[2], Thilo Glatzel[2], Giacomo Prampolini[3], and Ernst Meyer[2] — [1]University of Basel, Switzerland — [2]Universidad Autónoma de Madrid, Spain — [3]Consiglio Nazionale delle Ricerche (ICCOM-CNR), Italy

A combination of low temperature atomic force microcopy and molecular dynamic simulations is used to demonstrate that soft designer molecules realize a sidewinding motion when dragged over a gold surface. Exploiting their longitudinal flexibility, pyrenylene chains are indeed able to lower diffusion energy barriers via on-surface directional locking and molecular strain. The resulting ultralow friction reaches values on the order of tens of pN reported so far only for rigid chains sliding on an incommensurate surface. Therefore, we demonstrate how molecular flexibility can be harnessed to realize complex nanomotion while retaining a superlubric character. This is in contrast with the paradigm guiding the design of most superlubric nanocontacts (mismatched rigid contacting surfaces).

O 103.7 Fri 12:15 HE 101

Distance-Dependence of Orbital Density Mapping Using a CO-Functionalized STM Tip — •Fabian Paschke, Leonard-Alexander Lieske, Florian Albrecht, and Leo Gross — IBM Research Europe - Zurich, 8803 Rueschlikon, Switzerland

Knowledge over the tip apex in scanning probe techniques is indispensable to reliably identify the structure and electronic properties of surfaces and adsorbed molecules [1]. In particular, CO-terminated tips are widely used to achieve atomic resolution in atomic force microscopy [2]. In scanning tunneling microscopy (STM) the defined states of a CO tip apex enable high-resolution mapping of ionic resonances, and resulting orbital density images reflect a mix of s- and p-wave symmetries of the CO tip states [3-5].

In this work we study the appearance of frontier molecular orbitals of pentacene on a bilayer of NaCl grown on Cu(111) as a function of the tip-sample distance. STM constant-height imaging reveals a transition from *p*- to *s*-wave dominated tunneling. The findings provide an additional control knob to identify molecular electronic and spin states, which often requires STM imaging of orbital densities and careful assignment to calculated molecular orbitals [6].

[1] P. Hapala et al., Phys. Rev. B 90, 085421 (2014). [2] L. Gross et al., Science 325, 1110 (2009). [3] L. Gross et al., Phys. Rev. Lett. 107, 086101 (2011). [4] N. Pavlicek et al., Phys. Rev. Lett. 110, 136101 (2013). [5] A. Gustafsson et al., Phys. Rev. B 93, 115434 (2016). [6] S. Mishra et al., ACS Nano 16, 3264 (2022).

O 103.8 Fri 12:30 HE 101

Visualizing hydrated protons and their interconversion in monolayer water on metal surfaces — •Ye Tian[1], Jiani Hong[1], Duanyun Cao[1], Sifan You[1], Jing Guo[2], Ji Chen[1], En-Ge Wang[1], and Ying Jiang[1] — [1]International Center for Quantum Materials, Peking University, 100871 China — [2]College of Chemistry, Beijing Normal University, 100875 China

Hydrated protons are ubiquitous in solutions and involved in a variety of physical, chemical, biological, and energy-related processes. However, imaging and identifying the configuration of Zundel and Eigen cations and hydrated protons within the H bonding network of water remain a great challenge because of the high similarity between the hydronium (H3O+) and the water molecule (H2O). Recently, we performed the coadsorption of deuterium (D) atoms and D2O molecules on different metal substrates (Au, Cu, Pt, Ru), where the ionized D+ and D2O molecules can self-assemble into a two-dimensional (2D) hydronium-water overlayer. We have substantially improved the resolution and sensitivity of the qPlus-based atomic force microscopy such that the configuration of Zundel and Eigen cations solvated in the 2D water network could be directly visualized with an atomic-level resolution. Furthermore, we conducted the tip manipulation experiments to explore the dynamics of hydronium ions at the interface and found a new proton transfer pathway, accompanied with a simultaneous proton transfer from the water layer to the substrate, which is beyond the previously known elementary steps of hydrogen evolution reaction (HER).

O 103.9 Fri 12:45 HE 101

Exploring in-plane interactions beside an adsorbed molecule with lateral force microscopy — Shinjae Nam[1], Elisabeth Riegel[1], Lukas Hörmann[2,3], Oliver T. Hofmann[2], Oliver Gretz[1], •Alfred J. Weymouth[1], and Franz J. Giessibl[1] — [1]Universität Regensburg, Deutschland — [2]TU Graz, Österreich — [3]University of Warwick, UK

Atomic force microscopy (AFM) with a CO-tip can directly image the internal structure of a planar molecule. However, H-atoms usually cannot be directly observed due to their small size. At the same time, H-atoms are highly important, since they can direct on-surface chemical reactions. Measuring in-plane interactions at the sides of PTCDA molecules with lateral force microscopy allowed us to directly identify hydrogen atoms via their repulsive signature. Additional features in the force data could not be explained by H-bonding of the CO tip with the PTCDA sides. Instead, they are caused by electrostatic interaction of the large dipole of the metal apex. DFT calculations allowed us to estimate the strength of the dipole at the metal tip apex. To confirm that this dipole generally affects measurements on weakly-polarized systems, we investigated the a single CO molecule. We determined the radially-symmetric atomic interaction to be valid over a large solid angle of 5.4 sr, corresponding to 82°. Therefore in both the PTCDA and CO systems, the underlying interaction preventing direct observations of H-bonding and causing a collapse of the radially-symmetric model is the dipole at the metal apex, which plays a significant role when approaching closer than standard imaging heights.

O 104: Focus Session: Ultrafast Processes in Organic Semiconductors and Perovskites III (joint session O/CPP)

Time: Friday 10:30–11:45 Location: MA 004

O 104.1 Fri 10:30 MA 004

Unveiling ultrafast vibronic dynamics in organic chromophores from first principles — •Michele Guerrini[1], Jannis Krumland[1,2], and Caterina Cocchi[1,2] — [1]Physics Department and Center for Nanoscale Dynamics(CeNaD), Carl von Ossietzky Universität Oldenburg, Germany — [2]Physics Department and IRIS Adlershof, Humboldt-Universität zu Berlin, Germany

Identifying and quantifying the interplay between electron and nuclear dynamics in the excited state of organic molecules is of paramount importance to unveil and interpret their complex photophysics upon ultrafast excitation. In this work, we investigate from first principles the ultrafast vibronic dynamics of p-coumaric acid chromophore [1]. We start off the simulations in excited states corresponding to bright or dark transitions. By monitoring the kinetic energy spectral density [2] and the population dynamics, we analyze the characteristics of the electron-nuclear motion and its evolution over a time window of 300 fs. We observe that anharmonic and non-adiabatic effects are particularly pronounced in excitations with charge-transfer character and lead to instabilities with the eventual distortion of the system. On the other hand, the dynamics initialized from bright excited states with delocalized character remains electronically and structurally stable, and the resulting nuclear motion is markedly harmonic.

[1] Theor. Chem. Acc. 142, 110 (2023)

[2] J. Phys. Chem. A, 125, 9619 (2021)

O 104.2 Fri 10:45 MA 004

Ultrafast Charge-Transfer in Spiro-Bridged Triphenylamine Derivatives Investigated by Broadband Transient Absorption Spectroscopy — •Danyellen Galindo[1], Angelina Jocic[2], Anna Weidlich Weidlich[3], Frank Rominger[2], Thomas Oeser[2], Jonathan Zerhoch[1], Felix Deschler[1], Andreas Drew[3], Milan Kivala[2], and Tiago Buckup[1] — [1]PCI, Universität Heidelberg, Heidelberg — [2]OCI, Universität Heidelberg, Heidelberg — [3]IWR, Universität Heidelberg, Heidelberg

Triphenylamine compounds (TPA) are studied in order to understand their photochemical processes such as charge transfer (CT). This investigation is motivated by their ability in stabilizing positive charges through a delocalize π-system, thereby facilitating hole transport. This work aimed to study the optical properties of Spiro-bridged Fluorene N-heterotriangulenes (FTN-H) and FTN-(CN)6. Both compounds, dissolved in dichloromethane (DCM), underwent transient absorption experiments with 300 nm excitation. Initial findings suggest that the solvent does not play a significant role in the CT process. The non-subsituted compound upon excitation simultaneously displayed S1 electronic band for the molecule core and fluorene side group, succeeded by a 2 ns inter-system crossing (ISC) transition, thus forming a triplet state T*1 that decays to S0 with time constant of 16 *s. FTN-(CN)6 exhibited overall comparable dynamics, but also displayed an ultrafast CT between S1 and T*1 states. Such CT, which can be observed in various donor-acceptor materials, hints at potential applications in solar energy conversion and storage systems.

O 104.3 Fri 11:00 MA 004

The effect of the acceptor strength on intramolecular charge

separation in quadrupolar dye — •Somayeh Souri[1], Katrin WInte[1], Daniel Luenemann[1], Fulu Zheng[2], Mohamed Madjet[2], Teresa Kraus[3], Elena Mena-Osteritz[3], Peter Baeuerle[3], Sergei Tretiak[4], Antonietta De Sio[1], and Christoph Lienau[1] — [1]Oldenburg University, Germany — [2]Bremen University, Germany — [3]Ulm University, Germany — [4]Los Alamos National Laboratory, USA

Quadrupolar dyes, consisting of two terminal acceptors (A) linked to a central donor (D) are of high relevance for applications in nonlinear optics and photovoltaics. They recently demonstrated exceptional nonadiabatic quantum dynamics. In A-D-A thin films, we have uncovered the existence of intermolecular conical intersections, funneling energy into a lower-lying electronic state within less than 50 fs [1]. This raises the question how the strength of the acceptor group affects the quantum dynamics. Chemical intuition suggests that an increase in acceptor strength may accelerate charge transfer and increase its yield. We synthesize three A-D-A molecules with different acceptor strength and study them in polar solvents using ultrafast spectroscopy with 10fs time resolution. Even though we can distinguish between vibronic coupling-induced charge separation on a sub-100 fs scale and slower solvatochromism, we observe only weak effects of the acceptor group on the dynamics. The results suggest that vibronic coupling-induced excited state symmetry breaking plays a major role in quantum dynamics. [1] A. De Sio et al., Nature Nano 16, 63 (2021).

O 104.4 Fri 11:15 MA 004

Probing Site-Specific Photocatalytic Activity of Triazin-Based Graphitic Carbon Nitride via Time-Resolved and Resonant X-Ray Photoelectron Spectroscopy. — •Matz Nissen[1], Charlotte Ruhmlieb[1], Ivan Baev[1], Sebastian Denke[1], Lukas Wenthaus[3], Dmytro Kutnyakhov[3], Nils Wind[2,3], Federico Pressacco[3], Jens Buck[2], Steffen Palutke[4], Marion Kuhlmann[3], Günter Brenner[3], Kai Rossnagel[2,3], Alf Mews[1], and Michael Martins[1] — [1]Universität Hamburg, D — [2]Christian-Albrechts-Universität zu Kiel, D — [3]DESY, Hamburg, D — [4]European XFEL, Hamburg, D

Triazin-Based Graphitic Carbon Nitride (TGCN) has shown potential as a metal-free catalyst for photocatalytic water splitting, utilizing soley a visible light source and water. To gain further insight into the morphology of TGCN and the specific location where water binds during the catalytic reaction, we conducted resonant and time-resolved photoelectron spectroscopy of the carbon and nitrogen sites in a clean and a water environment. These experiments were done at beamline P04 of PETRAIII and beamline PG2 of FLASH respectively. Altering the sample environment shows differences in the resonant maps of the N K-edge. Additionally, time resolved measurements showed an excitation in the nitrogen spectra, specifically in the ring structure of the graphitic system. These results aim to help to improve the catalytic performance of the sample.

O 104.5 Fri 11:30 MA 004

Ultrafast dynamics in polymeric carbon nitride thin films probed by time-resolved extreme-ultraviolet photoemission spectroscopy — Nataliia Kuzkova[1,2], Igor Yu. Kiyan[1], Iain Wilkinson[1], and •Christoph Merschjann[1] — [1]Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Hahn-Meitner-Platz 1, 14109 Berlin, Germany — [2]Advanced Research Center for Nanolithography, Science Park 106, 1098 XG Amsterdam, The Netherlands

Polymeric carbon nitrides (PCN) have been extensively studied for various possible applications, mainly in the field of photocatalysis, but also as potential organic semiconductors, luminescent materials, etc. Especially photocatalytic applications are affected by the apparently low charge-separation and transport efficiency of PCN. It is therefore of interest to investigate the early-time dynamics of photoexcited charge carriers, specifically at the surface of the photocatalyst. To this end, we applied femtosecond time-resolved pump-probe extreme-ultraviolet photoemission spectroscopy (TRPES) to differently synthesized PCN thin films. Excitation at 400 nm sequentially populates a pair of short-lived transient species, which subsequently produce two different long-lived excited states on a sub-picosecond time scale. These could be assigned to singlet-exciton and charge-transfer states with a high initial spatial correlation, respectively. The results also show the potential of TRPES as a tool for in-situ investigations of early-time dynamics in photocatalytic processes.

[1] Kuzkova et al., *Phys. Chem. Chem. Phys.*, **25**, 27094 (2023)

O 105: 2D Materials VII: Heterostructures (joint session O/TT)

Time: Friday 10:30–12:45 Location: MA 005

O 105.1 Fri 10:30 MA 005

Defect-free two-dimensional core-shell heterostructures: MoS_2-TaS_2/Au(111) — Kai Mehlich[1,2], Mahdi Ghorbani-Asl[3], Daniel Sahm[4], Thais Chagas[4], Daniel Weber[1], Catherine Grover[1], Daniela Dombrowski[1,2], Arkady Krasheninnikov[3], and •Carsten Busse[1,2] — [1]Department Physik, Universität Siegen, Germany — [2]Institut für Materialphysik, Universität Münster, Germany — [3]Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf, Germany — [4]Department Bauingenieurwesen, Universität Siegen, Germany

We prepared two-dimensional core-shell heterostructures of the monolayer transition metal dichalcogenides (TMDCs) MoS_2 and TaS_2 by reactive molecular beam epitaxy (MBE) on chemically inert and weakly interacting Au(111). The heterostructures are in a size regime where quantum confinement can be expected. Despite large lattice mismatch a seamless interconnection of the two materials has been achieved, confirming successful encapsulation of the semiconducting core by a metallic shell. The resulting strain is analyzed on the atomic scale using scanning tunneling microscopy (STM) and compared to continuum models as well as simulations unsing empirical potentials.

O 105.2 Fri 10:45 MA 005

Growth and nanomanipulation of ultrathin Bismuthene nanoflakes on h-BN monolayers — •Antonio J. Martínez-Galera[1,2,3] and José M. Gómez-Rodríguez[2,3,4] — [1]Departamento de Física de Materiales, Universidad Autónoma de Madrid, E-28049 Madrid, Spain — [2]Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid, E-28049 Madrid, Spain — [3]Instituto de Ciencia de Materiales Nicolás Cabrera, Universidad Autónoma de Madrid, E-28049 Madrid, Spain — [4]Departamento de Física de la Materia Condensada, Universidad Autónoma de Madrid, E-28049 Madrid, Spain

Bismuthene, a young member of the family of 2D Materials, exhibits unique electronic properties when reduced to a single layer [1,2]. This study explores the growth of thin bismuth nanoflakes on h-BN monolayers under ultrahigh vacuum conditions. High-resolution scanning tunneling microscopy (STM) images unveil a stacking arrangement of Bi atoms within the nanoflakes, similar to the structure of Bi(110) planes in bulk material. Precise control of nanoflakes thickness, down to the lower limit of a bilayer, has been achieved by adjusting the deposited Bi amount on the h-BN surfaces. In addition to the structural characterization, well-controlled nanomanipulation experiments by using the STM tip have been conducted with these nanoflakes.

References:

[1] F. Reis et al. Science 357, 287-290 (2017).

[2] S. Zhang et al. Chem. Soc. Rev. 47, 982-1021 (2018).

O 105.3 Fri 11:00 MA 005

Optical properties of van der Waals TMD heterostructures from first-principles — •Riccardo Reho[1,2] and Zeila Zanolli[1,2] — [1]Chemistry Dept., Debye Institute for Nanomaterials Science, Utrecht University, Utrecht, The Netherlands — [2]ETSF

Van der Waals (VdW) heterostructures of two-dimensional transition metal dichalcogenides (TMDs) provide a unique platform to investigate rich phenomena stemming from the intricate interactions of charge, spin, and moiré superlattice with many-body effects. Controlling and predicting their optical properties with precision remains challenging.

Employing the ab initio GW-BSE method, we will offer an in-depth microscopic description of the optical properties of TMDs heterostructures. Beyond conventional electronic and absorption analyses, we also delve into the Photoluminescence spectra, leveraging a recent implementation we introduced in the Yambo code. We discuss on the roles of various degrees of freedom, such as mechanical strain and twist alignment. Our findings underscore the importance of structural properties, especially geometrical relaxation and computational subtleties, in ensuring accurate predictions of the band structure and absorption spectra for MoS_2/WS_2 and $MoSe_2$/WSe_2 heterostructures. Notably, we report a pronounced excitonic shift attributed to twisting and strain, shedding light on their profound impact on optical properties.

O 105.4 Fri 11:15 MA 005

Complexity of excitons at the TMD-Graphene interface — •Amir Kleiner, Daniel Hernangómez Pérez, and Sivan Refaely-Abramson — Weizmann Institute of Science, Rehovot, Israel

The complex optical characteristics of heterostructures composed of layered 2D materials are of great importance and interest. Specifically, the interaction between light and matter at the interface of layered transition metal dichalcogenides (TMDs) and graphene draw significant interest, facilitating the understanding of related energy-transfer mechanisms and their structural roots. We use state-of-the-art first principles calculations to study the dependence of the excitonic composition and absorption properties of the representative WS_2 - Graphene heterostructures on the structural details of these structures. Examining the cases of 0° and 30° degrees of interlayer twist angle, we find that the induced Brillouin zone mismatch, and concomitantly the energy level alignment between the graphene Dirac cone and the TMD bands, dictate the excitonic properties resulting in significant variation between the two systems. In particular, these set the charge-separation of nature of low-energy interlayer excitons and the state hybridization of optically active intralayer excitonic peaks. We use our results to numerically quantify the graphene-induced homogeneous broadening as a function of heterostructure alignment. Our findings provide guidelines for optical excitations based on the composition of the heterostructure, indicating a direct connection between the stacking of the layers, the separation of charges within the excitons, and the broadening of optical features.

O 105.5 Fri 11:30 MA 005

Electronic Characterization of Magnetic $CrCl_3$ Monolayers on $NbSe_2$ — •Jan Cuperus, Anna Reinhold, Daniel Vanmaekelbergh, and Ingmar Swart — Debye Institute for Nanomaterials Science, Utrecht University, The Netherlands

Electronic devices today suffer from a considerable loss of energy in the form of heat. With the ever increasing scale of information processing, it is essential to develop devices that are more efficient either in power usage or in information processing. In both cases, new materials are required to enable this. Van der Waals heterostructures are a class of materials that are well suited to offer the required properties. By combining materials with different properties in a heterostructure with a high quality interface, a plethora of properties can be obtained. Recently, this strategy was used to obtain topological superconductivity by combining superconducting $NbSe_2$ with ferromagnetic $CrBr_3$ [1].

We have conducted STM experiments on a similar heterostructure, replacing the out-of-plane ferromagnetism of CrBr3 by the in-plane ferromagnetism of $CrCl_3$. We present the MBE growth of $CrCl_3$ on $NbSe_2$ and structural insight into the formed heterostructure by STM measurements. In addition, the electronic structure is investigated using differential conductance measurements, both with and without an externally applied magnetic field. We show that the superconductivity of $NbSe_2$ couples to the in-plane ferromagnet, thereby introducing in-gap states.

[1] S. Kezilebieke, et al. Nature 588, 424-428 (2020).

O 105.6 Fri 11:45 MA 005

Interplay of tunnelling gap and Faraday-like screening in graphene heterostructures — •Tobias Wichmann[1,2,3], Keda Jin[1,2,4], Jose Martinez Castro[1,4], Honey Boban[5], Lukasz Plucinski[5], Tom G. Saunderson[6,7], Yuriy Mokrousov[6,7], Markus Ternes[1,2,4], F. Stefan Tautz[1,2,3], and Felix Lüpke[1,2] — [1]Peter-Grünberg-Institut (PGI-3), Forschungszentrum Jülich, Germany — [2]Jülich Aachen Research Alliance (JARA) - Fundamentals of Future Information Technology, Germany — [3]Institut für Experimentalphysik IV A, RWTH Aachen, Germany — [4]Institut für Experimentalphysik II B, RWTH Aachen, Germany — [5]Peter-Grünberg-Institut (PGI-6), Forschungszentrum Jülich, Germany — [6]Institute of Physics, Johannes Gutenberg University, Mainz, Germany — [7]Peter-Grünberg-Institut (PGI-1) and Institute for Advanced Simulations (IAS), Forschungszentrum Jülich, Germany

Scanning tunneling spectroscopy of graphene shows a gap around the Fermi energy, as tunnelling channels to the graphene Dirac states are suppressed due to their finite momentum in the graphene plane. Until now, applications of this phenomenon have been lacking. We report the interplay of the tunnelling gap and Faraday-like screening in graphene placed on Fe_3GeTe_2 (FGT). By tunnelling through the electronic gap of the graphene into the underlying FGT surface, we directly access the electronic properties of the graphene/FGT van der Waals interface. Studying the magnetoelectric tunability of the heterostructure properties, we find Faraday-like screening of the electric field exerted by the tunnelling probe by the graphene.

O 105.7 Fri 12:00 MA 005

Probing the phase transition to a coherent 2D kondo lattice — •Cosme Gonzalez Ayani[1,2,3], Michele Pisarra[4], Ivan M. Ibarburu[2,3], Manuela Garnica[2], Rodolfo Miranda[2,3,5], Fabian Calleja[2], Fernando Martin[2,3], and Amadeo L. Vazquez de Parga[2,3,5] — [1]Institute of Physics, Zagreb, Croatia — [2]Universidad autonoma madrid, Madrid, Spain — [3]Imdea Nanociencia, Madrid, Spain — [4]Universita della Calabria, Rende, Italy — [5]IFIMAC, Madrid, Spain

Kondo screening occurs when a magnetic impurity is embedded in a metal, below a given temperature, known as the Kondo temperature, a singlet state forms between the spin of the impurity and the spins of the conduction electrons. When the distance between the magnetic impurities is small enough the physics of the system is expected to be modified. The first experimental evidence was obtained in the 1970s in systems containing rare earths. By means of scanning tunneling microscopy (STM) and spectroscopy (STS) at low temperatures we explore a van der Waals heterostructure consisting in a single layer of 1T-TaS2 on a 2H-TaS2 crystal. The 1T-TaS2 layer presents a (13x13)R13.9 charge density wave (CDW) with a localized electron at the center of every unit cell of the CDW. For temperatures below 28 K the spatially resolved STS shows the presence of a Kondo resonance in the Mott-Hubbard gap. For temperatures below 11 K the system develops a quantum coherent state called Kondo lattice, resembling the physics of 3D heavy fermion metals.

O 105.8 Fri 12:15 MA 005

Engineering quantum dot nanoarrays in van der Waals heterostructures — •Keda Jin[1,2], Lennart Klebl[3], Junting Zhao[1,2], Tobias Wichmann[1,5], F. Stefan Tautz[1,5], Felix Lüpke[1], Dante Kennes[4], Jose Martinez-Castro[1,2], and Markus Ternes[1,2] — [1]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, 52425 Jülich, Germany — [2]Institut für Experimentalphysik II B, RWTH Aachen, 52074 Aachen, Germany — [3]I.Institute for Theoretical Physics, Universität Hamburg, 22607 Hamburg, Germany — [4]Institut für Theorie der statistischen Physik, RWTH Aachen, 52074 Aachen — [5]Institut für Experimentalphysik IV A, RWTH Aachen, 52074 Aachen, Germany

Superlattice engineering in graphene has generated interest for their ability to mimic the moiré potential observed in twistronics. While the moiré potential is sensitive to the twist angle, we present a method to periodically modulate the graphene potential by stacking graphene on 1T/2H-$NbSe_2$. The doping effect from the charge density wave of 1T-$NbSe_2$ allows for the formation of quantum dot nanoarrays. Using scanning tunneling microscopy/spectroscopy, we visualized the localized electronic structures to be independent of the twist angle between graphene and $NbSe_2$. Furthermore, we observed a bias-dependent strip pattern, breaking the six-fold rotational symmetry, which indicates an emergence of correlated states in the quantum dot nanoarrays. Our research paves the way for the fabrication of quantum dot nanoarrays in van der Waals heterostructures, extending beyond graphene to other 2D van der Waals heterostructures that are not limited by twist angle.

O 105.9 Fri 12:30 MA 005

Mechanical Characterization of Molecular Sieving Polymers — •Jakob Kreie[1], André Beyer[1], Yonghang Yang[2], Zhikun Zheng[2], and Armin Gölzhäuser[1] — [1]Physics of Supramolecular Systems and Surfaces, Bielefeld University, Germany — [2]School of Chemistry, Sun Yat-sen University, Guangzhou, Guangdong, Republic of China

To enable the practical use of molecular sieves on a larger scale, it is crucial to achieve a certain mechanical stability and elasticity. Our study deals with the analysis of polymer membranes as molecular sieves, these 2D imine-linked covalent organic frameworks are periodic and porous networks. The focus of the research is on their permeability as well as their mechanical properties.

Through the application of an atomic force microscope, we conducted comprehensive evaluations using free-standing nanoindentation and the bulge test. The results revealed a Young's modulus of 55 GPa and a breaking strength of 83 N/m for a membrane thickness of 45 nm. Moreover, our observations demonstrated remarkable elasticity during repeated tests, even after localized damage to the free-standing membrane had occurred.

O 106: Scanning Probe Microscopy: Light Matter Interaction at Atomic Scales IV

Time: Friday 10:30–12:45 Location: MA 041

O 106.1 Fri 10:30 MA 041

Probing Local Structural and Chemical Properties of Borophene via Tip-Enhanced Raman Spectroscopy — Linfei Li and •Nan Jiang — University of Illinois Chicago, Chicago, IL, 60607

Two-dimensional boron (i.e., borophene) has recently been synthesized and computationally predicted to have promising physical properties for a variety of electronic and energy technologies. However, the fundamental chemical properties of borophene that form the foundation of practical applications remain unexplored. Here, we present atomic-level chemical characterization of borophene materials using ultrahigh vacuum tip-enhanced Raman spectroscopy (UHV-TERS). Atomic-level chemical characterization of SL and BL borophene was achieved by TERS with an angstrom-scale vibrational fingerprint observation. Following that, we measure the interfacial interactions of a vertical Van der Waals heterostructure of borophene with tetraphenyldibenzoperiflanthene (DBP) molecules. TERS reveals subtle ripples and compressive strains of the borophene lattice underneath the molecular layer. The induced interfacial strain is demonstrated to extend in borophene by ~1 nm beyond the molecular region. Furthermore, we use our method to probe the local chemical properties of oxidized borophene. The results show that single oxygen adatoms on borophene can be identified and mapped with ~4.8 Å spatial resolution and single bond (B-O) sensitivity.

O 106.2 Fri 10:45 MA 041

Tip-enhanced Raman spectroscopy study on anatase TiO_2 surfaces — •Kyungmin Kim[1,2], Daiki Katsube[3], Takashi Kumagai[4], Martin Wolf[2], Masayuki Abe[1], and Akitoshi Shiotari[2] — [1]Osaka University, Toyonaka, Japan — [2]Fritz Haber Institute, Berlin, Germany — [3]RIKEN, Wako, Japan — [4]Institute for Molecular Science, Okazaki, Japan

Anatase, a crystalline phase of titanium dioxide (TiO_2), is traditionally considered to exhibit higher catalytic activity compared to rutile. Within the various surface orientations of anatase, the (001) surface and its (1x4) reconstructed counterpart have been identified as particularly reactive. The characterization of adsorbates and defects on these surfaces is crucial for comprehending the catalytic properties of the anatase surface. However, achieving this has been technically challenging. In this study, a single-crystal anatase TiO_2 thin film was fabricated on a $SrTiO_3$(100) substrate using the pulse laser deposition (PLD) technique. Following deposition, reflection high-energy electron diffraction (RHEED) measurements were conducted to verify the (1x4) surface reconstruction. The Anatase TiO_2 film exhibited a thickness of approximately 100 nm. Furthermore, we showcase the utility of tip-enhanced Raman spectroscopy (TERS) in conjunction with scanning tunneling microscopy (STM) under cryogenic conditions for vibrational spectroscopy and nanoscale resolution imaging in characterizing adsorbates and defects on the anatase TiO_2 surface.

O 106.3 Fri 11:00 MA 041

Atomic scale chemical contrast in a plasmon-driven reaction: An STM-TERS study on PTCDA families on Si(111) — •Youngwook Park[1], Ikutaro Hamada[2], Adnan Hammud[1], Egbert Willem Meijer[3], Takashi Kumagai[4], Martin Wolf[1], and Akitoshi Shiotari[1] — [1]Fritz Haber Institute of the Max Planck Society, Berlin, Germany — [2]Osaka University, Suita, Japan — [3]Eindhoven University of Technology, Eindhoven, the Netherlands — [4]Institute for Molecular Science, Okazaki, Japan

A localized surface plasmon is a promising tool to drive chemical reactions by enhancing light-molecule interactions. However, controlling reactions at an atomic level remains as a challenge. Here we demonstrate a sharp contrast in reactivity by minimal chemical modification of a single-molecule photoswitch. The switch comprises a plasmonic nanojunction of an Ag tip and a perylene-di-anhydride (PTCDA) molecule on a Si(111)-7x7 reconstructed surface. With a laser focused at the junction, the molecule toggles between the surface and the tip, accompanying bond breaking and forming of the molecule with the surface, as characterized by scanning tunneling microscopy (STM) and tip-enhanced Raman spectroscopy (TERS). Perylene-di-imide (PDI), an imide-analogue of PTCDA, remains silent on the surface, whereas a methyl substitution at the imide, Me2-PDI, recovers the reactivity. Such dramatic shift in reactivity is intriguing in that the modified part of the molecule (O, NH, and N-Me) even does not directly participate in the chemical bonds with the surface. The results suggest the possibility of a rigorous submolecular-level control of plasmon chemistry.

Topical Talk O 106.4 Fri 11:15 MA 041

Characterization of just one atom using synchrotron x-rays — •Volker Rose — Argonne National Laboratory, Lemont, IL, USA

The real-space observation of chemistry using scanning tunneling microscopy (STM) or synchrotron-based x-ray microscopy (XM) continues to have a tremendous impact on our understanding of nanoscale materials. However, although STM provides high spatial resolution, it lacks direct chemical contrast. On the other hand, XM can provide chemical sensitivity, but the spatial resolution is inferior. In order to overcome these limitations, we have developed a technique that combines the chemical contrast of synchrotron x-rays with the locality of STM.

In this presentation, we show that x-rays can be used to characterize the elemental and chemical state of just one atom [1]. Using a specialized tip as a detector, x-ray excited currents generated from an iron and a terbium atom coordinated to organic ligands are detected. The x-ray signal can be sensed only when the tip detector is located directly above the atom in extreme proximity, which confirms atomically localized detection in the tunnelling regime. Our work connects synchrotron x-rays with a quantum tunnelling process and opens a path to future x-ray experiments for the characterization of chemical properties of materials at the ultimate single atom limit.

This work was performed at the Advanced Photon Source and the Center for Nanoscale Materials, a U.S. Department of Energy Office of Science User Facility under Contract No. DE-AC02-06CH11357. [1] Nature 618, 69-73 (2023).

O 106.5 Fri 11:45 MA 041

Excited-state spectroscopy of individual molecules with atomic force microscopy — •Lisanne Sellies, Jakob Eckrich, and Jascha Repp — University of Regensburg, Regensburg, Germany

The combination of scanning probe microscopy with an increasing number of spectroscopic techniques, such as Raman and luminescence, provides unprecedented microscopic understanding. Despite its very direct access to well-defined single-molecule model systems, the unambiguous assignment of observations to specific quantum transitions is not always straightforward [1,2], and typically only a small fraction of all transitions can be studied.

Here, we propose a novel type of single-molecule spectroscopy that enables access to many quantum transitions of different types, including radiative, non-radiative and charge-related transitions. The molecules of interest are deposited on thick insulating films, preventing tunneling of charges to and from the underlying support. By controlled charge exchange between the conductive tip of an atomic force microscope and the molecule, we bring a molecule into different electronic configurations. The detection proceeds via the force acting on the tip. Thereby, quantum transitions can be mapped out in energy, enabling access to the energies of the lower-lying electronic states of individual molecules, including out-of-equilibrium states. We demonstrate our technique by applying it on pentacene and PTCDA, shedding light on the recent STM luminescence experiments on PTCDA [1,2].

References: [1] K. Kimura et al., Nature 570, 210-213 (2019) [2] J. Dolezal et al., ACS Nano 16, 1082-1088 (2022)

O 106.6 Fri 12:00 MA 041

Atomic-scale investigation of hot electron dynamics in a single molecule — •Shaoxiang Sheng[1], Yang Luo[1], Alberto M. Jimenez[1], Andrea Schirato[2], Giuseppe Dalla Valle[2], Guilio Cerullo[2], Klaus Kern[1,3], and Manish Garg[1] — [1]Max Planck Institute for Solid State Research, Stuttgart, Germany — [2]Dipartimento di Fisica - Politecnico di Milano, Milan, Italy — [3]Institut de Physique, Ecole Polytechnique Fédérale de Lausanne, Lausanne, Switzerland

The generation and detection of hot electrons have attracted broad attention due to their substantial potential for applications across a wide range of fields1. Extensive studies have been carried out to understand hot electron dynamics in various nanostructures2. Here, we investigate hot electron dynamics by utilizing pump-probe spectroscopy and measuring anti-Stokes emission in the tunnel junction of a scanning tunneling microscope. Our results reveal that the generation of hot electrons is highly localized and their decay in time is energy dependent. We also observe nonlinear four-wave mixing (FWM) signal when the pump and probe pulses overlap in time. Both the hot electron and FWM signals exhibit high sensitivity to the atomic-scale structures and show a large enhancement along the edges of a graphene nanoribbon owing to higher density of the electronic states. This study opens new perspectives in exploiting hot electron induced catalytic reactions in a single molecule and nonlinear optical response in materials at the atomic scales. References: 1. C. Clavero, Nat. Photonics, 8, 95-103 (2015). 2. A. Schirato, et al., Nanophotonics 12, 1-28 (2023).

O 106.7 Fri 12:15 MA 041

Fluorescent single-molecule STM probe — •Niklas Friedrich[1], Anna Roslawska[2], Xabier Arrieta[3], Michelangelo Romeo[2], Eric Le Moal[4], Fabrice Scheurer[2], Javier Aizpurua[3], Andrei G. Borisov[4], Tomas Neuman[4,5], and Guillaume Schull[2] — [1]CIC nanoGune BRTA, Donostia, Spain — [2]Université de Strasbourg, CNRS, IPCMS, Strasbourg, France — [3]Center for Materials Physics (CSIC-UPV/EHU), DIPC, Donostia, Spain — [4]Université Paris-Saclay, CNRS, Orsay, France — [5]Czech Academy of Sciences, Prague, Czech Republic

We present the functionalization of a plasmonic scanning probe tip with a single fluorescent molecule. The functionalization is performed by mechanically attaching an individual PTCDA molecule to the tip's apex atoms in an upright standing configuration. The electroluminescent character of the singly negatively charged PTCDA$^-$ is preserved, despite the molecule's direct contact with a metal electrode. We characterize the change in luminescence caused by different plasmonic tips, changing cavity geometries and varying bias voltage. Furthermore, we discuss theoretical considerations unraveling the photon emission mechanism.

The presented system showcases the possibility to use single molecules as nanoscale sensors able to retrieve optical properties of samples at the atomic scale.

http://arxiv.org/abs/2311.16805

O 106.8 Fri 12:30 MA 041

Hot luminescence from single-molecule chromophores electrically and mechanically self-decoupled by tripodal scaffolds — •Vibhuti Rai[1,2], Nico Balzer[3], Gabriel Derenbach[1], Christof Holzer[4], Marcel Mayor[3], Wulf Wulfhekel[1], Lukas Gerhard[1], and Michal Valášek[3] — [1]Institute for Quantum Materials and Technologies, Karlsruhe Institute of Technology, Kaiserstraße 12, 76131 Karlsruhe, Germany — [2]Freie Universität Berlin, Department of Physics, Arnimallee 14,14195 Berlin, Germany — [3]Institute of Nanotechnology, Karlsruhe Institute of Technology, Kaiserstraße 12, 76131 Karlsruhe, Germany — [4]Institute of Theoretical Solid State Physics, Karlsruhe Institute of Technology, Kaiserstraße 12, 76131 Karlsruhe, Germany

Control over the electrical contact to an individual molecule is one of the biggest challenges in molecular optoelectronics. Chemical route of anchoring individual chromophores via extended tripodal scaffolds is a promising approach for efficient electrical decoupling from metallic leads [1]. Here, we show that NDI chromophores coupled to a gold substrate via a tripodal scaffold and via a vacuum barrier to the tip of a scanning tunnelling microscope enable spectrally and spatially resolved electroluminescence. We show that specific vibrons of the chromophore which are mechanically decoupled from the substrate give rise to hot-luminescence (HL) bands. Vibrons which are coupled to the substrate do not show HL.

[1] Rai, V. et al. Phys. Rev. Lett. 130, 036201 (2023).

O 107: Nanostructured Surfaces and Thin Films

Time: Friday 10:30–12:15 Location: MA 042

O 107.1 Fri 10:30 MA 042

Morphology of nanostructured surfaces — •Jens Uwe Neurohr[1], Michael Klatt[2], and Karin Jacobs[1] — [1]Experimental Physics and Center for Biophysics, Saarland University, Saarbrücken, Germany — [2]Institut für KI Sicherheit, Deutsches Zentrum für Luft- und Raumfahrt (DLR), Wilhelm-Runge-Straße 10, 89081 Ulm, Germany

Nanorough surfaces are often classified by the root mean square (RMS) roughness. However, a single scalar value can capture only limited geometric information about the complex structured surface. Indeed, there are different families of nanorough surfaces that are clearly distinguished by their geometric features, but they can have the same RMS values. As an important consequence, the RMS fails to predict the adhesive behavior of bacteria on nano-structured surfaces (in fact, even within one family) [1]. A solution to this problem is offered by the so-called Minkowski functionals that provide a complete characterization of „additive shape information“as rigorously proven in integral geometry [4]. These comprehensive shape descriptors have been successfully applied to a broad range of applications [1,2,3]. Here, we will use the Minkowski functionals to distinguish families of nanorough surfaces. This presentation will, moreover, show how they can predict the adhesive behavior of Staphylococcus aureus on Black Si.

[1] Spengler et al., Nanoscale 11:19713 (2019); [2] Schröder-Turk et al., Adv. Mater. 23, 2535 (2011); [3] Wagner, Physik Journal 14, 41 (2016); [4] Schneider and Weil, Stochastic and integral geometry (2008), Vol. 1 Springer

O 107.2 Fri 10:45 MA 042

Geometric tuning of the structural and magnetic properties of magnetic thin films via deposition onto highly ordered arrangements of nanospheres — •Asmaa Qdemat[1], Emmanuel Kentzinger[1], Johan Buitenhuis[2], Sabine Pütter[3], Mai Hussein Hamed[1,4], Connie Bednarski-Meinke[1], Oleg Petracic[1], and Thomas Brückel[1] — [1]Jülich Centre for Neutron Science JCNS-2, Forschungszentrum Jülich GmbH, 52425 Jülich, Germany — [2]Institute for Biological Information Processes, Biomacromolecular Systems and Processes (IBI-4), Forschungszentrum Jülich GmbH, 52425 Jülich, Germany — [3]Jülich Centre for Neutron Science (JCNS) at Heinz Maier-Leibnitz Zentrum (MLZ), Forschungszentrum Jülich GmbH, 85747 Garching Germany — [4]Faculty of Science, Helwan University, 11795 Cairo, Egypt

Using curved surfaces as substrates for thin film deposition induces lateral thickness variation, allowing deposited properties to vary. [Co/Pd]n multilayers with different Co thicknesses were deposited on a flat silicon substrate, and densely packed two-dimensional arrays of silica nanospheres with different radii of curvature were formed by the drop-casting method [1]. Scanning electron microscopy, X-ray reflectivity (XRR), grazing incidence small-angle X-ray scattering, SQUID and neutron reflectivity were used to characterise the obtained nanostructure. Compared to the flat multilayer, the cap multilayer showed a different anisotropy axis direction. A change in coercivity as a function of film thickness and radius of curvature was also observed. [1] A. Qdemat, et.al., RSC Adv., 10, 2020.

O 107.3 Fri 11:00 MA 042

Fabrication of Nanofluidic Channels by Pulsed Laser Irradiation of SiOx-coated Fused Silica — •Nastaran Bakhtiari[1,2] and Jürgen Ihlemann[1] — [1]Institut für Nanophotonik Göttingen e.V., Hans-Adolf-Krebs-Weg 1, 37077 Göttingen, Germany — [2]Theoretical Physics and Center of Interdisciplinary Nanostructure Science and Technology, FB10, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany.

Considerably, nanofluidic channels have recently garnered attention. Existing methods for fabricating nanochannels are complex, costly, and time-intensive. In this study, we efficiently created transparent nanofluidic channels on fused silica substrates (SiO2) using a simple two-step process. Employing single-pulse excimer laser irradiation with a rear configuration treated a UV-absorbing silicon suboxide (SiOx) film through the transparent SiO2 substrate. A polydimethylsiloxane (PDMS) superstrate applied before laser exposure served as a confinement for controlled structure formation. Under optimal laser fluence, the thin SiOx film buckled, leading to the formation of channels with a width ranging from 10 to 20 μm and a height of 800 to 1200 nm, exhibiting a bell-like cross-sections following the so-called Euler buckling mode. Wider channels displayed varicose or telephone cord morphologies. Subsequent high-temperature annealing oxidized SiOx, resulting in transparent SiO2 channels. These nanochannels show promise in effectively transporting fluids of various viscosities, demonstrated through capillary action and in line with the Lucas-Washburn equation.

O 107.4 Fri 11:15 MA 042

Phase-Resolved Sum-Frequency Generation Microscopy of Lipid Rafts in Model Membranes — •Alexander Fellows, Ben John, Martin Wolf, and Martin Thämer — Fritz-Haber-Institut der MPG, Berlin, Germany

Since the lipid raft model was developed, it became clear that the lateral assembly and packing of phospholipids in cell membranes is a critical parameter controlling their physiological function. However, whilst studies of model systems have observed and characterised many structural aspects of such condensed lipid domains, their exact structure down to the molecular level remains unknown.

Here, we utilise our new advancement in molecular imaging through phase-resolved SFG microscopy to investigate model lipid rafts in lipid monolayers with mixed chirality. Through rotationally-dependent hyperspectral imaging of the lipid domains, we fully determine their 3D structures, showing that their in-plane molecular packing is highly curved and that their mesoscopic structure is part of a hierarchal spiral motif. Furthermore, through enantiotopic substitution measurements, we demonstrate that all aspects of the structural directionality are dependent on the lipid chirality, but importantly also that the formed structures display a clear deviation in mirror symmetry for different chirality mixtures. Such an observation gives an important insight into the enantioselective interactions that dominate many membrane processes.

O 107.5 Fri 11:30 MA 042

Atomistic insights into surface roughening of palladium hydride thin films — Minaam Qamar[1], •Matous Mrovec[1], Apinya Ngoipala[2], Matthias Vandichel[2], and Ralf Drautz[1] — [1]ICAMS, Ruhr University Bochum, Germany — [2]University of Limerick, Ireland

Palladium is one of the primary electrocatalysts for the hydrogen evolution reaction. It has been observed experimentally that surfaces of Pd thin films roughen during hydrogenation, but atomic-scale mechanisms behind these degradation processes are still not clear. We carried out large-scale atomistic simulations of PdH surfaces of Pd substrates using the Atomic Cluster Expansion (ACE). ACE is a data-driven interatomic potential with a formally complete basis that can reach quantum accuracy while remaining highly computationally efficient. We generated a versatile ACE potential for the Pd-H system based on large database of density functional theory calculations and validated it extensively for a broad range of properties and atomic configurations. We will present findings of molecular dynamics and Monte Carlo simulations of Pd/PdH interfaces and discuss the mechanisms responsible for the surface roughening.

O 107.6 Fri 11:45 MA 042

A plasma process to enhance electrode performance for large scale hydrogen production — •Timo Wagner[1], Nicolas Wöhrl[1], Vineetha Vinayakumar[2], Christian Marcks[3], Anna Mechler[3], Doris Segets[2], and Axel Lorke[1] — [1]Faculty of Physics and CENIDE, University Duisburg-Essen, Germany — [2]Particle Science and Technology (IVG-PST) and CENIDE, University of Duisburg-Essen — [3]AVT.ERT, RWTH Aachen University

In the context of H2Giga, a flagship project by the German Federal Ministry of Education and Research (BMBF) focused on hydrogen, we devised a plasma treatment for industry-relevant material systems. A very optimized substrate for electrolysis is Nickel Foam. Nickel foams offer a substantial specific surface area, and are widely used in academic research. Though, their higher cost and handling challenges make them less ideal for industrial use compared to bulk nickel plates. With this plasma process we could improve the electrochemical performance of Nickel plate electrodes notably, while also significantly improving the mechanical stability of the electrodes, thus increasing electrode lifetime. During this process, the surface of the sample undergoes a significant restructuring towards a hierarchical porous morphology. Nitrification can be observed by different methods, like XPS, XRD and EDX. This hierarchical structuring can also be observed on different substrate materials, e.g. Copper. Furthermore, we could successfully scale the plasma process from the lab scale of 1cm2 sample area to an industry testing scale of 100cm2.

O 107.7 Fri 12:00 MA 042

Anode surface engineering via ultrathin alumina membrane for dendrite-free sodium metal batteries — •Jiajia Qiu, Changfan Xu, Yu Duan, Huaping Zhao, and Yong Lei — Fachgebiet Angewandte Nanophysik, Institut für Physik & IMN MacroNano, Technische Universität Ilmenau, 98693 Ilmenau, Germany

Considerable growth in portable electronics and electric vehicles has stimulated the urgent demand for new battery systems. Owing to the high theoretical specific capacities (1166 mAh g-1) and low redox potentials (-2.71 V vs. a normal hydrogen electrode) of Na metals, Na metal batteries have attracted high research attention. But its large volume change and dendrite growth lead to low Coulombic efficiency, limited cyclability, and even a safety risk for high-energy-density Na metal batteries. Owing to chemical and electrochemical stability, Al2O3 coatings enable reduced Na dendrite growth, resulting in performance improvement. Simultaneously, a highly ordered ultrathin alumina membrane (UTAM) can change the traditional nucleation and growth modes from the origin, which can provide a porous 3D host to accommodate the large volume change of the Na anode. This novel nanostructure surface engineering via UTAM may bring in new opportunities for next-generation dendrite-free Na metal batteries.

O 108: Electronic Structure Theory II

Time: Friday 10:30–12:30 Location: MA 043

O 108.1 Fri 10:30 MA 043

Downfolding approaches to electron-phonon coupling — •Jan Berges[1], Nina Girotto[2], Arne Schobert[3], Erik van Loon[4], Michael Sentef[1], Sergey Brener[3], Mariana Rossi[5], Tim Wehling[3], Samuel Poncé[6,7], and Nicola Marzari[8,1] — [1]University of Bremen, Germany — [2]Institute of Physics, Zagreb, Croatia — [3]University of Hamburg, Germany — [4]Lund University, Sweden — [5]MPI for the Structure and Dynamics of Matter, Hamburg, Germany — [6]ETSF, UCLouvain, Belgium — [7]WEL Research Institute, Wavre, Belgium — [8]EPFL, Lausanne, Switzerland

Despite the success of density-functional perturbation theory (DFPT), the simulation of electron-phonon interactions in materials remains challenging, especially for large systems or in presence of strong correlations. We address this problem by downfolding it to effective low-energy models, using localized representations in the basis of Wannier functions. Based on data from DFPT, this allows us to obtain phonon frequencies at high precision and low computational cost via corrections to the static phonon self-energy [Phys. Rev. X **13**, 041009 (2023)]. A generalization beyond the harmonic approximation is possible, providing free energies, forces, and force constants for structurally distorted systems on supercells, based on DFPT data for the primitive cell of the undistorted system alone. This is an ideal platform for the study of charge-density waves (CDWs) and polarons. We apply it to perform molecular-dynamics simulations [arXiv:2303.07261] and to calculate phonon dispersions [Nano Lett., in press (2023), arXiv:2307.13791] for two-dimensional transition-metal dichalcogenides in the CDW phase.

O 108.2 Fri 10:45 MA 043

All-electron Periodic *GW* Method with Numeric Atom-centered Orbitals: Systematic Benchmark and Application to Temperature-dependent Band Structure — •Min-Ye Zhang[1,2], Uthpala Herath[3], Volker Blum[3], Xinguo Ren[2], Hong Jiang[4], and Matthias Scheffler[1] — [1]The NOMAD Laboratory at the FHI of the Max-Planck-Gesellschaft and IRIS-Adlershof of the Humboldt-Universität zu Berlin — [2]Institute of Phyiscs, Chinese Academy of Sciences, China — [3]Department of Mechanical Engineering and Materials Science, Duke University, U.S. — [4]College of Chemistry and Molecular Engineering, Peking University, China

The many-body G_0W_0 method is the state-of-the-art for theoretically analyzing the electronic structure of materials. The all-electron approach was recently implemented in FHI-aims for periodic systems. This method employs numeric atom-centered orbitals (NAOs) and localized resolution-of-identity approximation.[1] A thorough benchmark is still needed. Here we benchmark the precision in terms of standard NAOs and auxiliary basis functions for 50 gapped systems. We also measure the strong scaling with different system sizes for efficiency benchmark. By combining G_0W_0 with molecular dynamics, we showcase the possibility to study the temperature dependence of the band structure with electron-electron and electron-vibration self-energy considered jointly. This approach enables the study of strongly anharmonic materials where the phonon picture and electron-phonon coupling concepts break down.

[1] X. Ren *et al.*, *Phys. Rev. Materials* **5**, 013807 (2021).

O 108.3 Fri 11:00 MA 043

A Multimer Embedding Approach for Molecular Crystals up to Anharmonic Vibrational Properties — •Johannes Hoja, Alexander List, and Adrian Daniel Boese — University of Graz, Graz, Austria

Accurate modeling of molecular crystals is crucial for drug design and crystal engineering. However, periodic density functional calculations using hybrid functionals are often prohibitively expensive for practically relevant molecular crystals. One way of circumventing expensive periodic calculations is the usage of a subtractive embedding scheme, where the periodic calculation is only performed with a lower-cost method and then monomer energies and multimer interaction energies are replaced by those of the high-level method. Herein, we present such a multimer embedding approach for energies, structures, and vibrational properties of molecular crystals containing up to trimer interactions [1]. We evaluate this approach for the X23 benchmark set by approximating periodic PBE0+MBD by embedding multimers into less expensive PBE+MBD calculations. We show that trimer interactions are crucial for approximating lattice energies within 1 kJ/mol and cell volumes within 1 %. Harmonic vibrational free energies can already be approximated within 1 kJ/mol at the monomer or dimer level. Finally, we also utilize this embedding approach to incorporate anharmonic effects via second-order vibrational perturbation theory (VPT2) calculations of monomers and dimers.

[1] J. Hoja, A. List, A.D. Boese, *J. Chem. Theory Comput.*, in press, DOI: 10.1021/acs.jctc.3c01082.

O 108.4 Fri 11:15 MA 043

Interpreting ultrafast electron transfer on surfaces with first-principles electronic couplings — •Simiam Ghan[1], Elias Diesen[1], Christian Kunkel[1], Karsten Reuter[1], and Harald Oberhofer[2] — [1]Fritz-Haber Institute of the Max-Planck Society, Berlin — [2]Department of Physics, University of Bayreuth

The electronic coupling between adsorbates and surfaces is invoked in models of e.g. charge transfer, chemisorption, and quantum impurities (Kondo physics). While the coupling matrix element H_{ad} is usually approximated, we demonstrate here that it can be calculated directly from first principles Density Functional Theory. This is achieved through a projection of the Kohn-Sham Hamiltonian upon a basis of diabatic states localized on either the adsorbate or surface systems. An appropriate integration of couplings over the Brillouin zone allows for calculation of the Newns-Anderson chemisorption function, a coupling-weighted density of states which gives the line broadening of an adsorbate frontier state upon adsorption. This broadening corresponds to the experimentally-observed lifetime of an electron in the state, which we confirm for core-excited $\mathrm{Ar}^*(2p_{3/2}^{-1}4s)$ atoms on a number of transition metal surfaces. The use of explicit coupling matrix elements is found to be highly advantageous, elucidating the ultrafast electron transfer process in terms of coupling to the various bands of the surface[1]. The suitability of the scheme for treating fully chemisorbed systems is discussed.

[1] S. Ghan *et al. J. Chem. Phys.* **158**, 234103 (2023).

O 108.5 Fri 11:30 MA 043

Efficient Force Implementation for Implicit Solvation in All-Electron Full-Potential DFT — Daniel Waldschmidt[1], •Jakob Filser[2], and Harald Oberhofer[1,3] — [1]Technical University of Munich — [2]Fritz-Haber-Institut der MPG, Berlin — [3]University of Bayreuth

Implicit solvation remains a popular method to incorporate solvation effects into density functional theory (DFT). Rather than explicitly including solvent molecules in the *ab initio* calculation, this family of methods treats the solvent as a structureless dielectric medium, in which the solute resides in a cavity. This drastically reduces the size of the computationally expensive *ab initio* system while also removing the need to sample over different solvent configurations.

We have previously developed a multipole expansion implicit solvation model with multiple subcavities (MPE-*nc*) and implemented it in the DFT package FHI-aims. It solves the electrostatic problem to good accuracy at high computational efficiency. In a DFT context, we found it to not be the bottleneck of the calculation. Here, we discuss the derivation and implementation of analytical forces for MPE-*nc*, as well as their computational efficiency. This recent development lifts a rather severe limitation for the practical applicability, now making common modeling tasks such as geometry optimization and molecular dynamics accessible. We further address some minor adaptations to the model which are necessary to ensure the continuity of the potential energy surface and its agreement with the analytic forces, both prerequisites for most applications involving forces.

O 108.6 Fri 11:45 MA 043

A comprehensive exploration of structural and electronic properties of Molybdenum clusters — •Yao Wei, Alejandro Santana Bonilla, and Lev Kantorovich — Department of Physics, King's College London, London, WC2R 2LS , United Kingdom

Molybdenum clusters, characterised by their unique structure and intriguing catalytic properties, have gained significant attention in recent years. In several existing studies density functional theory (DFT) methods have been used to find the lowest energy Mo clusters and explore their electronic and magnetic structure. In all cases, with the exception of a single recent study, where a genetic algorithm was employed, initial geometries of the clusters, prior to geometry optimisation, were chosen using heuristic approaches based on symmetry considerations and known structures. DFT calculations were performed using different types of pseudopotentials, from hard to soft, and different types of basis sets. However, no comprehensive study has yet been done in which a DFT method with the best control on its precision would be complemented by a reliable global minimum search method to find the lowest energy Mo clusters. In this work, we employ a combination of a plane wave-based DFT method and ab initio random structure searching (AIRSS) technique to find the lowest energy clusters of up to 10 Mo atoms. In each case, the search has been performed for clusters with different spin multiplicities, which enabled us to explore their magnetic structure. Free energies of the Mo clusters, within the quasi-harmonic approximation, are also calculated and discussed. Similar methodology based on a stochastic exploration of the coordinate phase space is then presented for studying adsorption of Mo clusters on graphene and investigating their catalytic properties towards the CO dissociation reaction.

O 108.7 Fri 12:00 MA 043

Benchmark Auxiliary-Field Quantum Monte Carlo Method — •Zoran Sukurma[1], Martin Schlipf[2], Moritz Humer[1], Amir Taheridehkordi[1], and Georg Kresse[1,2] — [1]Kolingasse 14-16, University of Vienna — [2]Berggasse 21, VASP GmbH

Phaseless Auxiliary-Field Quantum Monte Carlo (ph-AFQMC) is a promising method to approximate the ground state properties of various quantum systems, including molecules, solids, and lattice models.

We present a scalable Fortran implementation of ph-AFQMC and demonstrate its excellent performance and favorable computational scaling. We apply the method to the HEAT set, the benzene molecule, water clusters, and a 2x2x2 diamond supercell. We observe a mean absolute deviation of the total energy of 1.15kcal/mol for the HEAT set. For water clusters, we find that ph-AFQMC yields binding energies that differ from CCSD(T) by typically less than 0.5kcal/mol. Excellent agreement with CCSD(T) is observed for diamond. In addition, we investigate modifications of the phaseless approximation that can help to reduce overcorrelation problems common to ph-AFQMC. Using the benzene molecule as an example, we demonstrate that the modified algorithm reproduces the same energy as the original algorithm with 400 Slater determinants.

Our results underscore the potential of ph-AFQMC to accurately predict ground-state properties in various systems.

Z. Sukurma, M. Schlipf, M. Humer, A. Taheridehkordi, and G. Kresse, J. Chem. Theory Comput. 19, 4921 (2023)

O 108.8 Fri 12:15 MA 043

Phaseless auxiliary field quantum Monte Carlo with projector-augmented wave method for solids — Amir Taheridehkordi[1], Martin Schlipf[2], Zoran Sukurma[1], •Moritz Humer[1], and Georg Kresse[1,2] — [1]University of Vienna, Faculty of Physics and Center for Computational Materials Physics, Austria — [2]VASP Software GmbH, Vienna, Austria

In this talk, we present our phaseless auxiliary field quantum Monte Carlo (ph-AFQMC) implementation using the plane-wave based projector augmented wave method. We showcase the accuracy and the feasibility of its application to solids. The AFQMC groundstate wavefunction is represented as an ensemble of Slater determinants placed in fluctuating auxiliary fields. This is achieved by transforming the two-body component of the many-body Hamiltonian into an effective one-body potential through the Hubbard Stratonovich transformation. Our implementation is numerically verified by consistent correlation energies from the primitive-cell sampling and the corresponding supercell calculations. We compare the equation of state of diamond and the correlation energies for a range of prototypical solid materials to conventional deterministic quantum-chemistry methods. A down-sampling technique along with natural orbitals accelerates the convergence with respect to the number of orbitals and crystal momentum points. Additionally, we demonstrate that our implementation is competitive in terms of accuracy and computational cost for dense crystal momentum point meshes compared to a well-established coupled-cluster singles-doubles with perturbative triples approach.

O 109: Focus Session: Proximity Effects in Epitaxial Graphene II

Time: Friday 10:30–13:00 Location: MA 141

Topical Talk O 109.1 Fri 10:30 MA 141

Heavy fermion quantum matter and topological superconductivity in artificial van der Waals heterostructures — •Jose Lado — Department of Applied Physics, Aalto University, Finland

Van der Waals materials provide a versatile platform for realizing a variety of emergent quantum states, including magnetic, correlated, and superconducting states, among others. Here we show that twisted van der Waals heterostructures provide a natural materials platform for realizing heavy fermion quantum materials [1,2] and topological moire superconductors [3,4]. We will discuss how a full heavy-fermion phase diagram can be realized in twisted graphene multilayers [1]. We will further show how heavy-fermion quantum matter can be experimentally realized in a bilayer made of two-dimensional materials [2]. We experimentally demonstrate [3] the emergence of a moire Yu-Shiba-Rusinov electronic structure stemming from the twist between the two van der Waals materials leading to a topological superconducting state. We will furthermore establish [4] a strategy to engineer highly tunable topological superconductivity in twisted graphene bilayers by exploiting a combination of moire patterns and proximity effects to 2D materials. Our results show that moire physics provides a powerful strategy to engineer ultra-clean heavy-fermion materials and artificial topological superconductors using van der Waals materials.

[1] Phys. Rev. Lett. 127, 026401 (2021), [2] Nature 599, 582*586 (2021), [3] Nano Lett. 2022, 22, 1, 328-333 (2022), [4] arXiv:2307.04605 (2023)

O 109.2 Fri 11:00 MA 141

Charge density wave in highly-doped single-layer graphene — Vivien Enenkel[1], Philipp Rosenzweig[2], Hrag Karakachian[2], Fabian Paschke[1], Ulrich Starke[2], and •Mikhail Fonin[1] — [1]Fachbereich Physik, Universität Konstanz, 78457 Konstanz, Germany — [2]Max-Planck-Institut für Festkörperforschung, Heisenbergstraße 1, 70569 Stuttgart, Germany

Recently, an emergence of electron correlation effects, such as superconductivity or charge density waves (CDW), in highly doped graphene monolayer has been predicted [1]. In our work, we perform Yb intercalation underneath zero-layer graphene on SiC to induce heavy electron doping in graphene. As reported previously, under Yd intercalation a van Hove singularity in graphene is pushed to the Fermi level, giving rise to an extended VHS, effectively pinning an almost non-dispersive flat band at $\mathrm{E_F}$ [2]. We use low-temperature scanning tunneling microscopy to discriminate between different structures of Yb-intercalated graphene, which correspond to different Yb arrangements at the interface. For three structures, $\mathrm{d}I/\mathrm{d}U$ measurements show a pronounced gap feature centered at $\mathrm{E_F}$. Together with strong changes of STM contrast upon bias voltage variation, the presence of the gap at $\mathrm{E_F}$ points at the formation of a CDW state in the intercalated graphene monolayer. [1] M. L. Kiesel *et al.*, Phys. Rev. B 86, 020507 (2012); [2] P. Rosenzweig *et al.*, Phys. Rev. B 100, 035445 (2019).

O 109.3 Fri 11:15 MA 141

Shaping superconducting proximity effect on graphene with nanometric precision — •Stefano Trivini[1], Eva Cortès-del Rio[6], Tim Kokkeler[9], Jon Ortuzar[1], Vladimir Cherkez[3,4], Jean-Yves Veuillen[3,4], Pierre Mallet[3,4], F. Sebastian Bergeret[8,9], Juan Carlos Cuevas[5,6], Ivan Brihuega[6,5,7], and Nacho Pascual[1,2] — [1]CICnanoGUNE 20018 San Sebastián, Spain — [2]Ikerbasque, 48013 Bilbao, Spain — [3]Institut Néel, F-38400 Grenoble, France. — [4]Universitè Grenoble Alpes, F-38042 Grenoble, France. — [5]IFIMAC, E-28049 Madrid, Spain. — [6]UAM, E-28049 Madrid, Spain. — [7]Instituto Nicolás Cabrera E-28049 Madrid, Spain — [8]DIPC 20018 San Sebastian, Spain — [9]CFM-MPC, E-20018 San Sebastián, Spain

Graphene can be turned superconducting by the proximity effect. The role of graphene gate-tunable density of states is crucial for understanding the stabilization of a collective proximitized state. Here, we investigate the proximitized superconductivity in graphene bilayers in contact with Pb islands grown on top. In particular, we compare, using LT-STM, the effect of superconducting Pb on graphene grown on the two faces of a polar SiC(1000) crystal. The effect of surface DOS turns out to be crucial. On the Si-side, we identify a collective superconducting state with a small pseudogap, while in the C-side, superconductivity is strong but local. Furthermore, we developed a method to laterally manipulate the Pb islands on graphene and build controllable Superconductor-Graphene structures.

O 109.4 Fri 11:30 MA 141

Growth optimization and comparison of PASG epitaxial graphene on 4H- and 6H-SiC — •Teresa Tschirner, Yefei Yin, Klaus Pierz, Frank Hohls, and Hans Werner Schumacher — Physikalisch-Technische Bundesanstalt, Bundesallee 100, 38116 Braunschweig, Germany

Fabricating epitaxial graphene by polymer-assisted sublimation growth (PASG) on SiC offers various advantages for application, such as production of large-area ultra-smooth defect- and bilayer-free graphene sheets, as well as reliable reproducibility. With the PASG method, the early formation of a buffer layer stabilizes the SiC surface and prevents step bunching of unfavorably high SiC surface terrace steps. We compare graphene growth on different polytypes of SiC with different parameters (such as miscut, polymer concentration, etc.) and the influence on the electronic properties of graphene. Differences between 4H- and 6H-SiC growth are two and three different terrace types, respectively, having inequivalent surface energies and subsequently different surface decomposition velocities [1]. We observe different step patterns on 6H-SiC of either alternating 0.25 and 0.5 nm high terrace-steps with alternating surface potential or equally distributed 0.75 nm steps. 4H-SiC on the other hand results for optimal growth in equally distributed very low step heights of 0.5 nm and equivalent SiC terraces with equal surface potential. The impact on the electronic transport is investigated by magneto-transport measurements.

[1] D. Momeni et al., Adv. Funct. Mater. 30, 2004695 (2020)

Topical Talk O 109.5 Fri 11:45 MA 141

Superconducting quantum devices in twisted graphene layers — •Klaus Ensslin — Physics Department, ETH Zurich

We realize gate-defined quantum devices on magic-angle twisted bilayer graphene. Different correlated states can be tuned by gate voltages, including superconducting states, band insulators and correlated insulators. This way we have fabricated Josephson junctions [1] as well as a SQUID with tunable symmetry [2]. A novel entirely gate-induced ring geometry [3] was employed to detect the Little-Parks effect [4] where the state of the ring as well as that one of the confining potential inside and outside of the ring became tunable. These experiments pave the way to more complex graphene quantum devices involving superconductivity and possibly Coulomb blockade. These experiments have been done in collaboration with Peter Rickhaus, Fokko de Vries, Elias Portoles, Giulia Zheng, Marta Perego, Alexandra Mestre Tora, Shuichi Iwakiri and Thomas Ihn.

[1] de Vries et al. , Nature Nano 16, 760 (2021) [2] Portolés et al., Nature Nano 17, 1159 (2022) [3] Iwakiri et al., Nano Lett. 22, 6292 (2022) [4] Iwakiri et al., arXiv:2308.07400

O 109.6 Fri 12:15 MA 141

Conductivity of intercalated Pb-monolayer in proximity to graphene — •Markus Gruschwitz, Andreas Cordier, Tim Güldenpfennig, Sergii Sologub, and Christoph Tegenkamp — Institut für Physik, TU Chemnitz, Chemnitz, Germany

Densely-packed monolayer of Pb, intercalated below the buffer layer on 4H-SiC(0001), reveals a long range ordered quasi (10×10) reconstruction, revealing charge neutral graphene and providing further exotic effects, e.g. spin-orbit coupling or a superconducting phase [1]. We investigated this 2D heterostructure in detail by means of in-situ multiprobe-STM as a function of tip geometry, tip spacing and temperature. Thereby, around 3.5 μm wide parallel terraces of intercalated graphene separated by narrow and less conductive barriers are formed, causing a tip-spacing dependent behavior from 1D to 2D and strong lateral anisotropy. We utilize finite element simulations to disentangle the different contributions and finally reveal the conductivity of the 2D heterosystem.

At room temperature the conductivity amounts to 600 kS/m. As the temperature is lowered, the conductivity decreases below 100 K similar to previous results [2]. Assuming combined transport channels in the metallic Pb layer and graphene, the increase in resistance is not in agreement with former T-dependent resistance measurements of monolayer graphene. We rather assume a small gap in the range of 2.5 meV in graphene, e.g. induced by spin-orbit coupling into graphene.

[1] Adv. Mater. Interfaces 10, 2300471 (2023); [2] Materials 14, 7706 (2021)

O 109.7 Fri 12:30 MA 141

Magnetotransport of Bi(110) islands on epitaxial graphene — •Julian Koch[1], Sergii Sologub[1,2], Chitran Ghosal[1], and Christoph Tegenkamp[1] — [1]Institut für Physik, TU Chemnitz, Reichenhainerstr. 70, 09126 Chemnitz — [2]Institute of Physics, NAS of Ukraine, Nauki avenue 46, 03028 Kyiv

Magnetotransport measurements using a 4 T magnet were performed on Bi islands on monolayer graphene/SiC with average thickness up to 3.6 bilayers (BL). They are supported by structural investigations using SPA-LEED and STM, which reveal that Bi predominately grows as needle-like islands with a (110) termination (see also [1]).

The carrier concentration determined from the SdH oscillations remains at 1×10^{13} cm^{-1} independent of the Bi coverage. In contrast, photoemission spectroscopy shows a strong doping of the graphene by Bi [2], indicating that the carrier concentration is highly anisotropic. This is confirmed by a positive, temperature independent contribution to the magnetoresistivity, which indicates that the Bi covered regions are electrically dead zones causing the electrons to scatter at the boundaries. This reduces the mobility from around 2250 cm^2/(Vs) for MLG to 1920 cm^2/(Vs) at 2.4 BL Bi, a decrease of approximately 14%. The weak localization of the clean surface is gradually reduced with increasing Bi coverage. This is caused solely by a decrease of the coherence length of the scattered electrons. The scattering lenghts associated with inter- and intravalley scattering remain unchanged.

[1] Koch et al. *J. Phys.: Condens. Matter* **36**, 065701 (2024)

[2] Gierz et al. *Nano Lett.* **8**, 12, 4603 (2008)

O 109.8 Fri 12:45 MA 141

long-term stable epitaxial graphene-based quantum Hall resistance standard for operating under relaxed conditions — •Yefei Yin[1,2], Mattias Kruskopf[1], Pierre Gournay[3], Benjamin Rolland[3], Martin Götz[1], Eckart Pesel[1], Davood Momeni[1], Frank Hohls[1], Klaus Pierz[1], Hansjörg Scherer[1], Rolf J. Haug[2], and Hans W. Schumacher[1] — [1]Physikalisch-Technische Bundesanstalt, Bundesallee 100, 38116 Braunschweig, Germany — [2]Institut für Festkörperphysik, Leibniz Universität Hannover, 30167, Hannover, Germany — [3]Bureau International des Poids et Mesures (BIPM), Pavillion de Breteuil, 92312 Sevres, France

The epitaxial graphene-based quantum Hall resistance standard (QHRS) has excellent potential for resistance metrology due to its large Landau level gap and strong Fermi level pinning. However, since as-grown epitaxial graphene has an electron density of 10^{13} cm^{-2}, permanent control of the carrier density and maintenance of the Hall resistance quantization are essential for practical graphene-based QHRS. This desirable goal is now achievable through the molecular doping by spin-coating F4-TCNQ dopant stacks on the graphene surface [1]. Our graphene-based QHRS show a Hall resistance quantization with an accuracy of $(2 \pm 2) \times 10^{-9}$ (k=2) under relaxed conditions of B = 4.5 T, I = 232.5 μA, and T= 4.2 K simultaneously. The accurate quantization has not shown any signs of degradation over two years, so far. Furthermore, the graphene-based QHRS has maintained quantization accuracy despite shipping between PTB and BIPM.[1] Y. Yin, et al., Adv. Physics Res. 1, 2200015 (2022)

O 110: Surface Magnetism

Time: Friday 10:30–12:45 Location: MA 144

O 110.1 Fri 10:30 MA 144

Artificially-constructed chains of magnetic adatoms on superconducting β-Bi2Pd — •Jinkyung Kim[1,2], Mireia Tena[3,4], Kyungju Noh[1,2], Piotr Kot[1], Yujeong Bae[1,2], Andreas Heinrich[1,2], Nicolas Lorente[3,4], and Deung-Jang Choi[3,4] — [1]Center for Quantum Nanoscience (QNS), Institute for Basic Science, South Korea — [2]Department of Physics, Ewha Womans University, Seoul 03760, South Korea — [3]Centro de Física de Materiales CFM/MPC (CSIC-UPV/EHU), 20018 Donostia-San Sebastián, Spain — [4]Donostia International Physics Center, 20018 Donostia-San Sebastián, Spain

Majorana bound state (MBS) has been studied in various condensed matter systems, such as one-dimensional topological superconductor, to realize topological quantum computation. Cr spin chains on superconducting β-Bi2Pd were suggested as one of the candidates of 1D topological superconductor, due to its combination of large magnetic moments and Rashba spin-orbit couplings of the superconducting substrate as shown in the simulation [1]. Taken advantage of atomic manipulation in STM, we artificially constructed a ferromagnetic Cr chain on a β-Bi2Pd surface. An evolution of in-gap states was carefully traced as we increase the length of the chain by attaching atoms one-by-one, and the results show excellent agreements with the simulations. We further investigate into topological phases of Cr chains based on the experimental discoveries, hinted as a formation of MBS at the edges of the chain. [1] Cristina Mier, et al., Physical Review B 104, 045406 (2021)

O 110.2 Fri 10:45 MA 144

Probing In-Gap States: Unveiling Pathways to Topological Superconductivity — •Deung-Jang Choi[1,2,3], Jinkyung Kim[4], Wonjun Jang[4], Yujeong Bae[4], Andreas Heinrich[4], and Nicolas Lorente[1,2] — [1]Centro de Física de Materiales CFM/MPC (CSIC-UPV/EHU), 20018 Donostia-San Sebastián, Spain — [2]Donostia International Physics Center (DIPC), 20018 Donostia-San Sebastián, Spain. — [3]Ikerbasque, Basque Foundation for Science, 48013 Bilbao, Spain — [4]Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul 03760, Korea

Lately, there has been a growing interest in the introduction of impurity states within the superconducting energy gap. Notably, the pursuit of a novel superconducting state known as topological superconductivity heavily relies on creating nanostructures with magnetic impurities on conventional (s-wave) superconductors that arrange spins in a chiral manner. Our study presents the initial findings resulting from the deliberate manipulation of individual atoms, enabling the construction of a chain of Cr atoms on a Bi2Pd superconductor [1,2,3]. These magnetic impurities, situated on diverse substrates, provide an unique opportunity to investigate various many-body effects and unconventional phenomena within different experimental spin systems. This research contributes to an enhanced comprehension of the underlying parameters governing each distinct system. References: [1] Phys. Rev. B 104 (4), 045406 (2021). [2] Phys. Rev. B 104 (24), 245415 (2021). [3] Phys. Rev. Research 4, L032010 (2022).

O 110.3 Fri 11:00 MA 144

Moiré-induced interface electronic states and magnetic properties of monolayer $FeCl_2$ on Bi(111) — •Shigemi Terakawa, Jingrong Ji, Gabriele Domaine, Emily C. McFarlane, Stuart S. P. Parkin, and Niels B. M. Schröter — Max Planck Institute of Microstructure Physics, Halle, Germany

Two-dimensional van der Waals (vdW) magnets have attracted increasing attention because of their potential use in magnetic vdW heterostructures, leading to various novel quantum phases through their unique properties such as the magnetic proximity effect and the moiré superlattice. $FeCl_2$ is a layered antiferromagnet, where the magnetic moments aligned in the out-of-plane direction are coupled ferromagnetically in a unit layer and antiferromagnetically between adjacent layers. We have successfully grown monolayer $FeCl_2$ film on Bi(111) using molecular beam epitaxy. The high crystallinity of the film was confirmed by low-energy electron diffraction (LEED) and scanning tunneling microscopy (STM). LEED and STM show strong moiré patterns. Angle-resolved photoelectron spectroscopy (ARPES) revealed the insulating band structure of monolayer $FeCl_2$. We found metallic X-shaped crossing bands at the $\overline{K}$ points of the moiré Brillouin zone (BZ). The bands can be understood as the interface states originating from the Bi(111) surface states, which are modified by hole doping and back-folding according to the moiré BZ. The magnetic hysteresis curves of monolayer $FeCl_2$ measured by X-ray magnetic circular dichroism (XMCD) show a higher slope at zero field for in-plane configuration, suggesting a magnetic order with an in-plane easy axis.

O 110.4 Fri 11:15 MA 144

Magnetic dichroism and spin polarization in threshold photoemission — •Frank O. Schumann[1], Jürgen Henk[2], Friederike Elisa Wührl[2], David Huber[2], and Wolf Widdra[2] — [1]Max-Planck Institut für Mikrostrukturphysik, Halle, Germany — [2]Institut für Physik, Martin-Luther-Universität Halle-Wittenberg, Germany

An emitted photoelectron can be analyzed with respect to energy, emission direction and spin polarization. The latter parameter is of particular interest for magnetic materials. The photoemission intensity depends on the polarization state of the light and magnetization direction leading to magnetic dichroism usually expressed as asymmetry. This can be used for magnetic contrast in domain imaging. For a microscopic description of spin polarization and magnetic dichroism it is vital to include both the spin-orbit and exchange interaction on an equal footing. We performed an appropriate one-step photoemission calculation for threshold photoemission from a Fe(100) surface. We found good agreement to the experimental magnetic dichroism data obtained with a photon energy of 5.2 eV. Apart from the intensity we obtain the theoretical spin polarization spectra. These are decomposed into an exchange and spin-orbit part. We find a sizable spin-orbit polarization and observe a resemblance to the asymmetry spectrum. We bring this into relation to analytical work, which demonstrated a connection between magnetic dichroism and the spin-orbit polarization in the limit of vanishing exchange interaction [1].
*[1] J. Henk et al., J. Phys.: Condens. Matter **8**, 47 (1996).

O 110.5 Fri 11:30 MA 144

Multi-orbital interactions and spin polarization in single lanthanide adatoms — •Massine Kelai[1], Stefano Reale[1], Jaehyun Lee[1], Philippe Ohresser[2], Deung-Jang Choi[3], Fabrice Scheurer[4], Aparajita Singha[5], and Fabio Donati[1] — [1]Center for Quantum Nanoscience, Institute for Basic Science, Seoul, Republic of Korea — [2]Synchrotron SOLEIL, L'Orme des Merisiers, France — [3]Donostia International Physics Center, Donostia-San Sebastián, Spain — [4]Institut de Physique et Chimie des Matériaux de Strasbourg, Strasbourg, France — [5]Max Planck Institute for Solid State Research, Stuttgart, Germany

Lanthanide atoms on surfaces show enormous potential for quantum information technologies. The use of a specific atom as an atomic-scale memory unit or qubit depends on the magnetic quantum level structure, which arises from the interaction between the atom and the substrate. Understanding the magnetic and electronic properties requires insight into the $4f$ and $5d6s$ orbitals, as well as their mutual interaction. Here, we investigate Nd atoms on Ag(100), HOPG, and Pb(111) using X-ray absorption and magnetic circular dichroism spectroscopies. The results show electronic transitions from atomic-like to bulk-like configurations with increased coordination on Ag(100) and HOPG, while Pb(111) remains bulk-like even for single atoms, and demonstrate that the Coulomb repulsion drives the electronic transition. We also reveal the existence of a fraction of spin-polarized $5d$ electrons for single atoms. This research enhances the understanding of the magnetism of surface-supported lanthanides.

O 110.6 Fri 11:45 MA 144

Long Range Magnetic Order in a Lanthanide, Fe-Porphyrin Metal-Organic Coordination Networks — •Dasom Choi[1,2], Massine Kelai[1,2], Serim Jeon[1,2], Stefano Reale[1,2], Corina Urdaniz[1,2], Andreas J. Heinrich[1,2], Dominik Lungerich[3,4], Christoph Wolf[1,2], Fabio Donati[1,2], and Luciano Colazzo[1,2] — [1]Center for Quantum Nanoscience, Institute for Basic Science (IBS), Seoul, South Korea — [2]Ewha Womans University, Seoul, South Korea — [3]Center for Nanomedicine, IBS, South Korea — [4]Graduate Program of Nano Biomedical Engineering, Advanced Science Institute, Yonsei University, South Korea

This research provides the electronic and magnetic characterization of a metal-organic networks formed using Iron-Tetrakis-(4-Cyanophenyl) Porphyrin (Fe-TCPP) and Dysprosium (Dy). By carefully depositing specific amounts of Fe-TCPP and Dy on Au(111), coordinated bonding between Dy and cyano-groups led to organized structures, forming extensive 2D islands and showcasing significant long-range ferromagnetic order. We used scanning tunneling microscopy/spectroscopy, x-ray magnetic circular dichroism and density functional theory to explore the structural and magnetic attributes of a metal-organic coordination network based on lanthanides. This method of fabrication lays the groundwork for creating magnetic 2D architectures on surfaces capable of storing and manipulating quantum information. This advancement propels the boundaries of quantum computing and information processing, potentially enabling the development of multiqubit systems.

O 110.7 Fri 12:00 MA 144

Recent Progress in the Investigation of the Magnetic Properties of Individual Lanthanide-Based Dimetallofullerene Single-Molecule Magnets — •Tobias Birk[1], Fabian Paschke[2], Vivien Enenkel[1], Fupin Liu[3], Jan Dreiser[4], Vladyslav Romankov[4], Stanislav M. Avdoshenko[3], Alexey A. Popov[3], and Mikhail Fonin[1] — [1]Department of Physics, University of Konstanz, 78457 Konstanz, Germany — [2]IBM Research Europe, 8803 Rüschlikon, Switzerland — [3]IFW Dresden, 01069 Dresden, Germany — [4]SLS, PSI, 5232 Villigen, Switzerland

Due to their rich and versatile chemical and physical properties, single-molecule magnets (SMMs) are in focus of a diverse and broad research community. Especially lanthanide based SMMs are promising candidates for future electronic devices because of long relaxation times and high blocking temperatures. Here we present a dimetallofullerene SMM with a single-electron lanthanide-lanthanide bond within its magnetic core, which shows chemical robustness and a blocking temperature of about 21 K [1]. We perform scanning tunnelling microscopy (STM) and spectroscopy (STS) measurements in order to investigate its electronic and magnetic properties. By investigating two different lanthanides, we demonstrated access to the magnetic core of the molecule [2]. Based on this, we perform inelastic electron tunnelling spectroscopy (IETS), which reveals symmetric conduction steps and additionally a prominent conductance peak due to a Kondo resonance.

[1] F. Paschke *et al.* Adv. Mater. 2102844 (2021).
[2] F. Paschke *et al.* Small 2105667 (2022)

O 110.8 Fri 12:15 MA 144

Tip-assisted motion of Co and Rh atoms on the antiferromagnet Mn/Re(0001) — •Felix Zahner, Kirsten von Bergmann, Roland Wiesendanger, and André Kubetzka — Institut für Nanostruktur- und Festkörperphysik, Universität Hamburg

The diffusion of adatoms on surfaces is a fundamental process, governing the growth and self-assembly of thin films and nanostructures. Adatom diffusion on non-magnetic surfaces has been investigated in real-space by local probes such as field ion microscopy [1] and scanning tunneling microscopy (STM) [2], whereas diffusion on magnetic surfaces has so far only been addressed theoretically [3].

In this contribution, we have studied Co and Rh atoms on a single fcc-stacked layer of Mn on Re(0001). The ground state of this hexagonal layer is a row-wise antiferromagnetic state [4], which reduces the symmetry and results in a uniaxial system. We use the tip of an STM at $T = 4.2$ K to induce adatom motion and investigate to what extent the magnetic state influences the movement of magnetic Co atoms and non-magnetic Rh atoms.

[1] G. Antczak, G. Ehrlich, Surface Science Reports **62**, 39 (2007).
[2] J. Li, R. Berndt, W.-D. Schneider, Phys.Rev.Lett. **76**, 1889(1996).
[3] S. Dennler, J. Hafner, Phys. Rev B **72**, 214414 (2005).
[4] J. Spethmann, S. Meyer, K. von Bergmann, R. Wiesendanger, S. Heinze, A. Kubetzka, Phys. Rev. Lett. **124**, 227203 (2020).

O 110.9 Fri 12:30 MA 144

Shape and magnetism of $Fe_{3-\delta}O_4$ nanoislands on $SrTiO_3$ — •Steffen Tober[1], Yifan Xu[1], Mohammad Tehrani[2,3], Mai H. Hamed[1], Asmaa Qdemat[1], Nadine Seidel[1], Jan-Christian Schober[2,3], Vedran Vonk[2], Connie Bednarski-Meinke[1], Ulrich Rücker[1], Andreas Stierle[2,3], and Emmanuel Kentzinger[1] — [1]Jülich Centre for Neutron Science (JCNS) and Peter Grünberg Institut (PGI), JARA-FIT, Forschungszentrum Jülich — [2]Deutsches Elektronen-Synchrotron DESY, Centre for X-ray and Nanoscience, Hamburg — [3]Universität Hamburg, Fachbereich Physik

The magnetic properties of $Fe_{3-\delta}O_4$ nanoparticles for medical applications depend on the synthesis route influencing their defect structure, ligand shell and shape [1]. Optimisation of the magnetisation requires disentangling preparation-specific influences from intrinsic size effects. $Fe_{3-\delta}O_4$ nanoislands on $SrTiO_3$ are introduced as a ligand-free model system with defined structure and orientation to study the magnetic properties of iron oxide nanostructures. Characterisation by diffraction, grazing incidence small angle X-ray scattering, neutron reflectivity and SQUID indicates the formation of crystalline, uniformly shaped nanoislands with a distinct magnetic shape anisotropy [2]. Our results form the basis for ongoing *in situ* studies probing interdependencies between composition, shape, defect structure and magnetisation of nanostructured iron oxides.

[1] D. Zákutná et al., PRX 10, 031019 (2020),[2] A. Devishvili et al., Rev. Sci. Instrum. 84, 025112 (2013).

O 111: Heterogeneous Catalysis II

Time: Friday 10:30–12:30 Location: TC 006

O 111.1 Fri 10:30 TC 006

Fast Screening of Metal Alloys for CO_2 Activation by Ab initio Calculations and Local AI Rules — •Herzain I. Rivera-Arrieta, Matthias Scheffler, and Lucas Foppa — The NOMAD Laboratory at the FHI of the Max-Planck-Gesellschaft and IRIS-Adlershof of the Humboldt-Universität zu Berlin

Metal alloy catalysts can weaken the bonds in CO_2, enabling reactions to convert the molecule into valuable products such as methanol. However, the immense compositional and configurational space of alloys hinders the direct high-throughput screening of novel materials. Herein, through DFT-mBEEF calculations, we modeled the CO_2 interaction with surfaces of Single-Atom Alloys, i.e., systems with one atomic dopant embedded in a host metal [1]. Then, out of 24 offered candidate properties, we used the Subgroup Discovery (SGD) approach [2] to unveil rules connecting key electronic and geometric properties of the adsorption sites in SAA with the CO_2 activation process. These rules, which constrain the values of the key properties, allow us to focus on the regions in materials space where the promising catalysts may be located. Besides applying the rules for screening new SAA, we also tested them to search promising Dual-Atom Alloys (DAA), i.e., structures with two dopant atoms [3]. Further calculations confirmed that the selected SAA and DAA can indeed activate CO_2.

[1] R. T. Hannagan, *et al.*, *Chem. Rev.*, **120**, 12044 (2020).
[2] S. Wrobel, *1st Europ. Symp. on Princ. of Data Min. and Knowl. Discov.*, **19**, 78 (1997).
[3] D. Behrendt, *et al.*, *J. Am. Chem. Soc.*, **145**, 4730 (2023).

O 111.2 Fri 10:45 TC 006

Ni-In Synergy in CO_2 Hydrogenation to Methanol Explained by Microkinetic Models — •Francesco Cannizzaro, Bart Klumpers, Ivo A. W. Filot, and Emiel J. M. Hensen — Laboratory of Inorganic Materials and Catalysis, Department of Chemical Engineering and Chemistry, Eindhoven University of Technology, 5600 MB Eindhoven, The Netherlands

Highly dispersed Ni-In clusters can promote In_2O_3 catalysts for the hydrogenation of CO_2 to CH_3OH, a key reaction in the fight against global warming. However, fundamental understanding of the role of such clusters is lacking, hampering large-scale application of Ni-In_2O_3 catalysts. Herein, we employed density functional theory (DFT) and microkinetic modeling to study the influence of the composition of In_2O_3-supported Ni-In clusters on CO_2 hydrogenation. Lowest-energy Ni_nIn_{8-n} clusters ($n = 0 - 7$) were identified by combining genetic algorithms with an artificial neural network potential trained by DFT. At higher Ni content, the clusters expose more Ni atoms. The mechanistic pathways for CO_2 hydrogenation were computed for In_2O_3-supported Ni_2In_6 and Ni_6In_2 clusters, representing In-rich or Ni-rich clusters.

Microkinetic simulations show that only the Ni_6In_2 cluster catalyzes methanol via hydrogenation of adsorbed CO_2 to formate, the precursor to methanol, by Ni-H species. Methanol formation competes with direct CO_2 dissociation, which becomes the dominant reaction pathway at high temperatures due to low H coverage. The supported Ni_2In_6 cluster exposes In atoms, which do not stabilize surface hydrides, explaining the low methanol activity and predominant CO formation.

O 111.3 Fri 11:00 TC 006

Systematic evaluation of trends in CO_2 reduction on metal–nitrogen–carbon systems from first principles — •Sze-Chun Tsang, Sihang Liu, and Georg Kastlunger — Catalysis Theory Center, Technical Univ. of Denmark, 2800 Kgs. Lyngby, Denmark

The electrochemical CO_2 reduction reaction (e-CO_2RR) can be a crucial aspect of sustainability. Metal–nitrogen–carbon (MNC) single-atom catalysts (SACs) hold promises towards electroreduction, besting transition-metal (TM) surface catalysts in suppressing side reactions and unlocking new energetic degrees-of-freedom. However, adsorbates on MNCs are often poorly modeled; and the oft-assumed and -used analogy between graphene-based (G-) and molecular MNCs has yet to be examined.

Using density-functional theory, we compare the adsorption energetics relevant to the CO_2RR on G-MNCs and molecular MNCs. We will show energetic trends to be largely metal-dependent and consistent across SAC geometries. We also demonstrate that higher-level theories like the exact-exchange functional are crucial towards capturing said energetics. Further, we examine the pairwise correlation, or the lack thereof, between reaction-intermediate formation energies on $3d$-TM-based MNCs. Finally, we discuss several caveats: (1) the sampling of spin states and its energetic ramifications; and (2) the tendencies for protons to potentially destabilize the active site. These results emphasize the importance of and highlight the difficulties in correctly describing MNC systems, illuminating key theoretical insights into this distinguished class of catalysts.

O 111.4 Fri 11:15 TC 006

High-throughput catalyst screening for CO_2 to methanol conversion with machine-learned force-fields — •Ondřej Krejčí, Prajwal Pisal, and Patrick Rinke — Department of Applied Physics, School of Science, Aalto University, Espoo, Finland

The search for new and better catalysts is one of the key research directions in material science, as heterogeneous catalysis is essential in converting CO_2 to fuel in a closed loop carbon cycle. Approximative predictions of activity, like the Sabatier principle, have been very popular for catalytic screening. However, to take the nanostructure of real thermocatalysts into account, we need to scan the adsorption energies (AEs) for a variety of different facets and binding sites.

In this work, we will present our current workflow for obtaining the relevant AEs in CO_2 thermoreduction to methanol. We employ trained machine learning force-field from the Open Catalyst Project [1], to accelerate the search for ideal catalysts. We have calculated the surface stabilities for various facets with all Miller indices $\in \{-2, -1, 0, 1, 2\}$ and picked the most stable cuts for each facet. Subsequently, we have created all possible high symmetry binding sites on those facets and predicted AEs for the reaction key semi-products: *H, *OH, *OCHO and *OCH_3. The AE distributions are further analysed for material*s activity.

[1] L. Chanussot et al., ACS Catal. **11**, 6059-6072 (2021); R. Tran et al., ACS Catal. **13**, 3066-3084 (2023); https://opencatalystproject.org/

O 111.5 Fri 11:30 TC 006

Exploring Unsupervised Learning for Analysis of Adsorption Energy Distributions in CO2 to Methanol Synthesis — •Prajwal Pisal, Ondrej Krejci, and Patrick Rinke — Department of Applied Physics, Aalto University, P.O.Box 11100, FI-00076 AALTO, Finland

Synthesis of methanol from carbon dioxide using heterogeneous catalysts is a reaction of great relevance from an industrial and environmental perspective, underscoring the need for extensive catalytic exploration. Understanding the adsorption energy distributions (AE) of reactants on catalytic surfaces is crucial for evaluating material reactivity, as catalyst reactivity is often linked to AE.

In this work, we leverage our extensive database of AE for more than 100 catalytic materials using the Open Catalyst Project infrastructure [1]. We perform statistical analysis and unsupervised machine learning, like dimensionality reduction and clustering, on the dataset for recognition of features and patterns in the AE distributions. Utilizing these tools, we classify the materials under study and compare with experimental and literature data (e.g. [2]). This analysis provides a deeper understanding of the key properties of the materials that enhance the catalytic activity. The data obtained will further aid us in building predictive models to maximize methanol yield as a function of AE distributions and experimental conditions.

[1] L. Chanussot et al., ACS Catal. 11, 6059-6072 (2021); R. Tran et al., ACS Catal. 13, 3066-3084 (2023)

[2] A. J. Medford et al, J. Catal. 328, 36-42 (2015).

O 111.6 Fri 11:45 TC 006

Theoretical investigation of bifunctional monolayer catalysts for fuel cell applications — •Thies Reetz and Halil Ibrahim Sözen — Carl von Ossietzky Universität Oldenburg

Road transport is estimated to contribute 10 % of all anthropogenic CO2 emissions. The use of polymer electrolyte membrane fuel cells (PEMFC) is a promising way to reduce these emissions. A problem facing the commercialization of PEMFCs are cell reversal events which occur under hydrogen depletion at the anode leading to the carbon oxidation reaction (COR) at carbon based support materials. In this work novel bifunctional anode catalysts are theoretically designed and investigated which can perform both the hydrogen oxidation reaction (HOR) and the oxygen evolution reaction (OER) simultaneously. The latter reaction is competing with the COR. Therefore by promoting the OER the COR is suppressed and the support material corrosion is slowed down.

The focus in this study was on Platinum and Iridium and bifunctional Pt/Ir alloys. Monolayers of these materials on a rutile support material were investigated in terms of structure and bifunctional catalytic performance and were compared to the performance of pure metal surfaces. The investigation showed that the adsorption energy of important species in the OER and HOR process changes drastically for the monolayer materials compared to the pure metal surfaces. Therefore, bifunctional monolayers on a support material are a possible way to tune PEMFC performance.

O 111.7 Fri 12:00 TC 006

Adaptive Designs for the Efficient Exploration of Catalyst Materials or Reaction Condition Spaces — •Frederic Felsen, Christian Kunkel, Karsten Reuter, and Christoph Scheurer — Fritz-Haber-Institut der MPG, Berlin

Changes of the reaction mechanism often lead to qualitative changes in the performance over a small range of reaction conditions or catalyst compositions. The topology of diagrams that summarize this performance over an investigated domain is therefore typically characterized by extended regimes of smooth behavior separated by narrow such transitions in between (e.g. kinetic phase transitions or peaks of Volcano plots). Such discontinuous topology prevents the efficient exploration with traditional design-of-experiment (DoE) approaches that assume a smoothly varying measurement function over the entire investigated domain.

To this end, we here propose an adaptive DoE algorithm that tackles this issue in a data-efficient way. A support vector machine classification learns and iteratively refines the *a priori* unknown positions of the transitions by optimally designing new data points, i.e. the reaction conditions or catalyst compositions at which the next measurements are to be taken. We illustrate the approach for a vast space of possible promoter compositions for the propane dehydrogenation reaction (PDH). Based on characterization data (Raman spectroscopy and thermogravimetric analysis) for each individual catalyst sample, we identify two distinct regimes, differing not only in automatically identified characterization features but also in catalytic performance.

O 111.8 Fri 12:15 TC 006

Accelerating Catalyst Discovery through Efficient Exploration of a Complex Design Space — •Charles W.P. Pare[1], Christian Kunkel[1], Frederic Felsen[1], Frederik Rüther[2], Sina Stocker[1], Robert Baumgarten[2], Esteban Gioria[1], Raoul Naumann d'Alnoncourt[2], Christoph Scheurer[1], Frank Rosowski[2,3], and Karsten Reuter[1] — [1]Fritz-Haber-Institut der MPG, Berlin — [2]BasCat - UniCat BASF JointLab, Berlin — [3]BASF SE, Catalysis Research, Ludwigshafen

Catalyst promoters often form key components of stable and well-performing industrial heterogeneous catalysts. Yet, today's industrial catalysts often only benefit from one or two promoters. This can often be traced back to the laborious empirical research required to identify promising formulations that jointly act to improve catalyst performance. To overcome such hurdles, we implemented an accelerated catalyst discovery approach by globally exploring a large multi-promoter design space using only a limited number of experiments. Its pillars are an efficient Design-of-Experiment (DoE) planning, a fast parallelized testing protocol and an iterative incorporation of experimental feedback. New and competitive promoter chemistries for the non-oxidative propane dehydrogenation to propylene over Pt on alumina were discovered in less than 100 experiments performed within weeks. The results show the potential of iterative DoE strategies for the data-efficient knowledge-generation and optimization in complex, academically and industrially relevant catalytic systems.

O 112: Overview Talk Ib Chorkendorff

Time: Friday 13:15–14:00 Location: HE 101

Invited Talk O 112.1 Fri 13:15 HE 101

A surface science approach to thermal and electrochemical ammonia synthesis — •Ib Chorkendorff — Technical University of Denmark, Kgs. Lyngby, Denmark

Activating of molecular Nitrogen is an extremely important process as it supplies in the form of fertilizer the nitrogen that it is a prerequisite for building all amino acids and nucleic acids essential for life. It would not be possible to sustain earths current population without having access to such activated nitrogen. After brief review of the history of activating nitrogen we shall concentrate on the ammonia synthesis and motivate why an alternative route to the current commercial Haber-Bosch could be attractive in a de-centralised electrified society. We shall here revisit the very first surfaces science reaction showing the thermal ammonia synthesis over single crystal of Iron by G Somorjai, using a new device allowing for detailed surface science analysis and very sensitive synthesis conditions at ambient conditions. This will be extended to new routes of promotion showing how Cobalt can also be made very active. The active site will be identified by a combination of Cobalt single crystals with and without steps combined with reaction over mass-selected nanoparticles made in situ by a cluster source. In the second part we shall turn to electrochemical ammonia synthesis where we now have shown how one can make ammonia at ambient conditions. The Li mediated process will be discussed and we will show how we over the last 5 years have gone from having a process that did not make any ammonia to now being capable of obtaining more than 80% Faradaic efficiency and high current densities.