

## Molecular Physics Division Fachverband Molekülphysik (MO)

Jochen Küpper  
Center for Free-Electron Laser Science  
Deutsches Elektronen-Synchrotron DESY  
Hamburg  
and  
Department of Physics  
Universität Hamburg  
jochen.kuepper@cfel.de

### Overview of Invited Talks and Sessions

(Lecture halls HS 3042, HS 3044, and HS 1015; Poster Tent C)

#### Invited Talks

MO 1.1	Mon	11:00–11:30	HS 3044	<b>Imaging ultrafast molecular dissociation dynamics; from conventional to surprising paths</b> — ●HEIDE IBRAHIM
MO 15.1	Wed	14:30–15:00	HS 3042	<b>Metal Cluster opportunities</b> — ●GEREON NIEDNER-SCHATTEBURG
MO 19.1	Thu	11:00–11:30	HS 3044	<b>Controlling the internal quantum states of chiral molecules</b> — JUHYEON LEE, ELAHE ABDIHA, BORIS SARTAKOV, GERARD MEIJER, ●SANDRA EIBENBERGER-ARIAS

#### Invited Talks of the joint Symposium SAMOP Dissertation Prize 2024 (SYAD)

See SYAD for the full program of the symposium.

SYAD 1.1	Mon	14:30–15:00	Paulusaal	<b>Quantum steering of a Szilárd engine</b> — ●KONSTANTIN BEYER
SYAD 1.2	Mon	15:00–15:30	Paulusaal	<b>Does a disordered Heisenberg quantum spin system thermalize?</b> — ●TITUS FRANZ
SYAD 1.3	Mon	15:30–16:00	Paulusaal	<b>Quantum optical few-mode models for lossy resonators</b> — ●DOMINIK LENTRODT
SYAD 1.4	Mon	16:00–16:30	Paulusaal	<b>Non-Hermitian topology and directional amplification</b> — ●CLARA WANJURA

#### Invited Talks of the joint Symposium Coulomb Explosion Imaging (SYCE)

See SYCE for the full program of the symposium.

SYCE 1.1	Tue	11:00–11:30	Paulusaal	<b>Dissociation of halogenated organic molecules induced by soft X-rays – pathways and early stages</b> — ●EDWIN KUKK
SYCE 1.2	Tue	11:30–12:00	Paulusaal	<b>X-ray induced Coulomb explosion imaging with channel-selectivity</b> — ●REBECCA BOLL
SYCE 1.3	Tue	12:00–12:30	Paulusaal	<b>Time-resolved Coulomb Explosion Imaging using X-ray Free-Electron Lasers</b> — ●TILL JAHNKE
SYCE 1.4	Tue	12:30–13:00	Paulusaal	<b>Dynamics and control of microsolvated biomolecules studied by Coulomb explosion imaging</b> — ●SEBASTIAN TRIPPEL, JOCHEN KÜPPER

#### Prize Talks of the joint Awards Symposium (SYAS)

See SYAS for the full program of the symposium.

SYAS 1.1	Tue	15:00–15:30	Paulusaal	<b>Quantum Simulations with Atoms, Molecules and Photons</b> — ●IMMANUEL BLOCH
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SYAS 1.2	Tue	15:30–16:00	Paulussaal	<b>Spectroscopy of molecules with large amplitude motions: a journey from molecular structure to astrophysics.</b> — ●ISABELLE KLEINER
SYAS 1.3	Tue	16:00–16:30	Paulussaal	<b>Quantum x-ray nuclear optics: progress and prospects</b> — ●OLGA KOCHAROVSKAYA
SYAS 1.4	Tue	16:30–17:00	Paulussaal	<b>3D printed complex microoptics: fundamentals and first benchmark applications</b> — ●HARALD GIESSEN

### Invited Talks of the joint Symposium Controlled Molecular Collisions (SYCC)

See SYCC for the full program of the symposium.

SYCC 1.1	Wed	11:00–11:30	Paulussaal	<b>Dynamics of CO<sub>2</sub> activation by transition metal ions - The importance of intersystem crossing</b> — ●JENNIFER MEYER
SYCC 1.2	Wed	11:30–12:00	Paulussaal	<b>Angular momentum of small molecules: quasiparticles and topology</b> — ●MIKHAIL LEMESHKO
SYCC 1.3	Wed	12:00–12:30	Paulussaal	<b>Manoeuvring chemical reactions one degree of freedom at a time</b> — ●JUTTA TOSCANO
SYCC 1.4	Wed	12:30–13:00	Paulussaal	<b>Cold and controlled collisions using tamed molecular beams</b> — ●SEBASTIAAN VAN DE MEERAKKER

### Invited Talks of the joint Symposium Size Selected Metal Cluster Spectroscopies (SYMC)

See SYMC for the full program of the symposium.

SYMC 1.1	Thu	11:00–11:30	Paulussaal	<b>Infrared spectroscopic studies of molecular activation at metal clusters</b> — ●STUART MACKENZIE
SYMC 1.2	Thu	11:30–12:00	Paulussaal	<b>Dynamic metal-metal cooperation in chemical reactions</b> — ●JANA ROITHOVÁ
SYMC 1.3	Thu	12:00–12:30	Paulussaal	<b>A closer look at the electronic structure of simple metal clusters</b> — ●BERND VON ISSENDORFF
SYMC 1.4	Thu	12:30–13:00	Paulussaal	<b>IR action spectroscopy of metal clusters, complexes and diatomics with free electron lasers</b> — ●ANDRÉ FIELICKE

### Invited Talks of the joint Symposium Ultrafast Quantum Nano-Optics (SYQO)

See SYQO for the full program of the symposium.

SYQO 1.1	Fri	11:00–11:30	Paulussaal	<b>Coherent and incoherent dynamics of colloidal plexcitonic nanohybrids</b> — ●ELISABETTA COLLINI
SYQO 1.2	Fri	11:30–12:00	Paulussaal	<b>Dissipative Many-Body Dynamics in Atomic Subwavelength Arrays in Free Space</b> — ●STEFAN OSTERMANN
SYQO 1.3	Fri	12:00–12:30	Paulussaal	<b>Quantum dot sources: efficiency, entanglement, and correlations.</b> — ●ANA PREDOJEVIĆ
SYQO 1.4	Fri	12:30–12:45	Paulussaal	<b>Compact chirped fiber Bragg gratings for single-photon generation from quantum dots</b> — ●VIKAS REMESH, RIA KRÄMER, RENÉ SCHWARZ, FLORIAN KAPPE, YUSUF KARLI, THOMAS BRACHT, SAIMON COVRE DA SILVA, ARMANDO RASTELLI, DORIS REITER, STEFAN NOLTE, GREGOR WEIHS
SYQO 1.5	Fri	12:45–13:00	Paulussaal	<b>Observing Ultrafast Coherent Dynamics following Selective Excitation of a Single Quantum Dot</b> — ●DARIUS HASHEMI KALIBAR, PHILIPP HENZLER, RON TENNE, ALFRED LEITENSTORFER

### Sessions

MO 1.1–1.7	Mon	11:00–13:00	HS 3044	<b>Coulomb-explosion Imaging (joint session MO/A)</b>
MO 2.1–2.8	Mon	11:00–13:00	HS 1098	<b>Attosecond Physics I (joint session A/MO)</b>
MO 3.1–3.8	Mon	17:00–19:00	HS 1015	<b>Novel Spectroscopies</b>
MO 4.1–4.6	Mon	17:00–18:30	HS 3044	<b>Strong-field Ionization and Imaging (joint session MO/A)</b>
MO 5.1–5.7	Tue	11:00–13:00	HS 1010	<b>Interaction with Strong or Short Laser Pulses I (joint session A/MO)</b>

MO 6.1–6.8	Tue	11:00–13:00	HS 3044	<b>Ultracold Molecules and Precision Spectroscopy (joint session MO/Q)</b>
MO 7.1–7.18	Tue	17:00–19:00	Tent C	<b>Poster: Spectroscopy</b>
MO 8.1–8.6	Tue	17:00–19:00	Tent C	<b>Poster: Collisions</b>
MO 9.1–9.7	Wed	11:00–13:00	HS 1010	<b>Attosecond Physics II / Interaction with VUV and X-ray light (joint session A/MO)</b>
MO 10.1–10.6	Wed	11:00–13:00	HS 1015	<b>Ultracold Molecules (joint session Q/MO)</b>
MO 11.1–11.7	Wed	11:00–12:45	HS 3044	<b>X-ray Spectroscopy</b>
MO 12	Wed	13:00–14:00	HS 3044	<b>Members' Assembly</b>
MO 13.1–13.8	Wed	14:30–16:30	HS 1010	<b>Interaction with Strong or Short Laser Pulses II (joint session A/MO)</b>
MO 14.1–14.8	Wed	14:30–16:30	HS 1015	<b>Atomic Clusters (joint session A/MO)</b>
MO 15.1–15.7	Wed	14:30–16:30	HS 3042	<b>Spectroscopy of Metal Clusters</b>
MO 16.1–16.8	Wed	14:30–16:30	HS 3044	<b>Ultrafast Dynamics I</b>
MO 17.1–17.12	Wed	17:00–19:00	Tent C	<b>Poster: Cold Molecules</b>
MO 18.1–18.12	Wed	17:00–19:00	Tent C	<b>Poster: Cluster</b>
MO 19.1–19.6	Thu	11:00–12:45	HS 3044	<b>Chirality</b>
MO 20.1–20.9	Thu	14:30–16:45	HS 3042	<b>Theoretical Molecular Physics</b>
MO 21.1–21.9	Thu	14:30–16:45	HS 3044	<b>Ultrafast Dynamics II</b>
MO 22.1–22.9	Thu	17:00–19:00	Tent C	<b>Poster: Molecules in Strong Fields</b>
MO 23.1–23.9	Thu	17:00–19:00	Tent C	<b>Poster: Chirality</b>
MO 24.1–24.8	Thu	17:00–19:00	Tent C	<b>Poster: Experimental Techniques</b>
MO 25.1–25.7	Fri	11:00–12:45	HS 3044	<b>Novel Experimental Approaches</b>
MO 26.1–26.6	Fri	14:30–16:00	HS 3042	<b>Cluster</b>
MO 27.1–27.8	Fri	14:30–16:30	HS 3044	<b>Ultrafast Dynamics III and High-harmonic Generation (joint session MO/A)</b>

### Members' Assembly of the Molecular Physics Division

Wednesday 13:00–14:00 HS 3044

## MO 1: Coulomb-explosion Imaging (joint session MO/A)

Time: Monday 11:00–13:00

Location: HS 3044

## Invited Talk

MO 1.1 Mon 11:00 HS 3044

**Imaging ultrafast molecular dissociation dynamics; from conventional to surprising paths** — ●HEIDE IBRAHIM — Advanced Laser Light Source (ALLS) @ Institut National de la Recherche Scientifique (INRS-EMT), Varennes, QC, Canada

Coulomb explosion imaging (CEI) is a powerful tool to track a broad variety of molecular dynamics; even if they occur in a non-concerted manner and require single-molecule detection sensitivity. Upon photoexcitation of a molecule it will break apart. We can see fragments following direct, conventional dissociation paths, as well as fragments deviating from this minimum energy path. The latter are called roaming fragments and explore the potential energy landscape in a statistical manner. At the user facility ALLS we use CEI in combination with high repetition rate laser systems. Dissociating and roaming fragments in formaldehyde are directly captured using CEI, a hard-to-grasp statistically occurring signal. Individual pathways are distinguished based on state-of-the-art theory analysis.

MO 1.2 Mon 11:30 HS 3044

**Dynamics of H<sub>2</sub>-roaming processes, H<sub>3</sub><sup>+</sup> formation in ethanol and aminoethanol initiated by two-photon double-ionization**

— ●AARON NGAI<sup>1</sup>, SEBASTIAN HARTWEG<sup>1</sup>, JAKOB ASMUSSEN<sup>2</sup>, BJÖRN BASTIAN<sup>3</sup>, LTAIEF BEN LTAIEF<sup>2</sup>, MATTEO BONANOMI<sup>4,5</sup>, CARLO CALLEGARI<sup>6</sup>, MICHELE DI FRAIA<sup>6</sup>, KATRIN DULITZ<sup>7</sup>, RAIMUND FEIFEL<sup>8</sup>, SARANG GANESHAMANDIRAM<sup>1</sup>, SIVARAMA KRISHNAN<sup>9</sup>, AARON LAFORGE<sup>10</sup>, LANDMESSER FRIEDEMANN<sup>1</sup>, MICHELBACH MORITZ<sup>1</sup>, PAL NITISH<sup>6</sup>, PLEKAN OKSANA<sup>6</sup>, RENDLER NICNICOLAS<sup>1</sup>, RICHTER FABIAN<sup>1</sup>, SCOGNAMIGLIO AUDREY<sup>1</sup>, SIXT TOBIAS<sup>1</sup>, SQUIBB RICHARD<sup>8</sup>, SUNDARALINGAM AKGASH<sup>2</sup>, STIENKEMEIER FRANK<sup>1</sup>, and MUDRICH MARCEL<sup>2</sup> — <sup>1</sup>Physikalisches Institut, Albert-Ludwigs-Universität Freiburg, Freiburg, Germany — <sup>2</sup>Department of Physics and Astronomy, Aarhus University, Aarhus, Denmark — <sup>3</sup>Wilhelm Ostwald Institute for Physical and Theoretical Chemistry, University of Leipzig, Leipzig, Germany — <sup>4</sup>Dipartimento di Fisica Politecnico, Milano, Italy — <sup>5</sup>Istituto di Fotonica e Nanotecnologie (CNR-IFN) Milano, Italy — <sup>6</sup>Elettra - Sincrotrone Trieste S.C.p.A., Basovizza, Trieste, Italy — <sup>7</sup>Institut für Ionenphysik und Angewandte Physik, Universität Innsbruck, Innsbruck, Austria — <sup>8</sup>Department of Physics, University of Gothenburg, Göteborg, Sweden — <sup>9</sup>Department of Physics, Indian Institute of Technology Madras, Chennai, India — <sup>10</sup>Department of Physics, University of Connecticut, Storrs, Connecticut, US

The trihydrogen cation (H<sub>3</sub><sup>+</sup>) is the simplest and one of the most abundant triatomic cations in the universe. It plays a crucial role in interstellar gas-phase chemistry as it facilitates molecule-forming chemical reactions. Dynamics in simple alcohols that lead to H<sub>3</sub><sup>+</sup> formation typically involve the unusual so-called "roaming"-mechanism of a neutral H<sub>2</sub> moiety. In comparison to previous experiments using strong-field ionization by infrared (IR) pulses [1], we produce dicationic ethanol and 2-aminoethanol molecules using two-photon double-ionization with extreme ultraviolet (XUV) light, and probe the dynamics of H<sub>3</sub><sup>+</sup> formation with a visible (VIS) pulse in a time-resolved pump-probe scheme. We compare results between measurements with XUV photons either below or above the double-ionization threshold, including the lifetimes of intermediate states.

[1] Ekanayake, N. *et al. Nat. Commun.* **9**, 5186 (2018)

MO 1.3 Mon 11:45 HS 3044

**New endstation for controlled molecule experiments and ultrafast dynamics of OCS** — ●WUWEI JIN<sup>1,2</sup>, IVO VINKLÁREK<sup>1</sup>, SEBASTIAN TRIPPEL<sup>1,3</sup>, HUBERTUS BROMBERGER<sup>1</sup>, SERGEY RYABCHUK<sup>1</sup>, ERIK MÄNSSON<sup>1</sup>, ANDREA TRABATTONI<sup>1</sup>, VINCENT WANIE<sup>1</sup>, FRANCESCA CALEGARI<sup>1</sup>, and JOCHEN KÜPPER<sup>1,2,3</sup> — <sup>1</sup>Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg — <sup>2</sup>Department of Physics, Universität Hamburg — <sup>3</sup>Center for Ultrafast Imaging, Universität Hamburg

Imaging ultrafast photochemical reactions with atomic-spatial and femtosecond-temporal resolution is one of the ultimate goals of physical chemistry and the molecular sciences [1]. We present details on our newly established transportable endstation for controlled molecules (eCOMO) and discuss our ultrafast (sub 10 fs) time-resolved study of the photodissociation dynamics of carbonyl sulfide (OCS) after UV-

photoexcitation at  $\lambda = 267$  nm. OCS was purified and separated from the helium seed gas using the electrostatic deflector [2]. The UV-induced dynamics was probed through strong field ionization using a velocity map imaging spectrometer in combination with a Timepix3 camera [3].

[1] J Onvlee, S Trippel, and J Küpper, *Nat. Commun.* **7462**, 13 (2022)

[2] YP Chang, D Horke, S Trippel, and J Küpper, *Int. Rev. Phys. Chem.* **557**, 34 (2015)

[3] H Bromberger, *et int.* (9 authors), S Trippel, B Erk, and J Küpper, *J. Phys. B.* **144001**, 55 (2022)

MO 1.4 Mon 12:00 HS 3044

**Complete imaging of the reaction pathways of ionized water dimer** — ●LUIA BLUM<sup>1,2</sup>, IVO S. VINKLÁREK<sup>1</sup>, HUBERTUS BROMBERGER<sup>1</sup>, SEBASTIAN TRIPPEL<sup>1</sup>, and JOCHEN KÜPPER<sup>1,2,3</sup>

— <sup>1</sup>Center for Free-Electron Laser Science CFEL, Deutsches Elektronen-Synchrotron DESY, Hamburg — <sup>2</sup>Department of Physics, Universität Hamburg — <sup>3</sup>Center for Ultrafast Imaging, Universität Hamburg

We applied a pure ensemble (92 %) of water dimer (H<sub>2</sub>O)<sub>2</sub>, spatially separated by electrostatic deflection, and subsequently ionized by strong-field ionization, to investigate the ion-radical chemistry of water clusters [1]. The direct observation of fragmentation channels of (H<sub>2</sub>O)<sub>2</sub><sup>+</sup> and (H<sub>2</sub>O)<sub>2</sub><sup>+2</sup> by multi-mass imaging reveals several yet unknown ion-radical pathways and provides a comprehensive picture of (H<sub>2</sub>O)<sub>2</sub><sup>+/+2</sup>, including experimental branching ratios. Furthermore, the ion yields for the Coulomb explosion channels of (H<sub>2</sub>O)<sub>2</sub><sup>+2</sup> indicate electron-recoil-impact ionization as the dominant process, opening the discussion about avenues to control electron recoil and multiple-ionization processes in supramolecular complexes. The study of the (H<sub>2</sub>O)<sub>2</sub><sup>+/+2</sup> ionization fragmentation process is highly relevant to ion-radical heterogeneous chemistry occurring on ice mantles in the Earth's atmosphere and in interstellar space [2].

[1] Vinklársek, I. S., Bromberger, H., Vadassery N., Jin W., Küpper, J., Trippel, S., *submitted*; arXiv:2308.08006 [physics].

[2] Vogt, E., Kjaergaard, H. G., *Annu. Rev. Phys. Chem.*, **73**, 209-231 (2022).

MO 1.5 Mon 12:15 HS 3044

**Understanding fragmentation dynamics of difluorodiiodomethane** — ●NIDIN VADASSERY<sup>1,3</sup>, IVO VINKLÁREK<sup>1</sup>, SEBASTIAN TRIPPEL<sup>1,2</sup>, and JOCHEN KÜPPER<sup>1,2,3</sup>

— <sup>1</sup>Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany — <sup>2</sup>Department of Physics, Universität Hamburg — <sup>3</sup>Department of Chemistry, Universität Hamburg

Unimolecular photo-fragmentation is prevalent in the many chemical reactions that affect the environment, like ozone depletion, synthesis of oxidative hydrocarbons, formation of aerosol particles, *etc.* [1]. The photo-dissociation of man-made and naturally occurring polyhalohydrocarbons is among the major causes which contribute to such climate-impacting reactions. Difluorodiiodomethane (CF<sub>2</sub>I<sub>2</sub>) one such example of polyhalohydrocarbon shows unconventional dynamics near dissociative energies [2]. Here, we present our experimental result of exploring the dissociation dynamics of CF<sub>2</sub>I<sub>2</sub> using near-infrared laser pulses. A pure sample of CF<sub>2</sub>I<sub>2</sub> was produced using the deflector in the eCOMO endstation [3]. We show capability of the end-station to reveal metastable states and unravel the complex quantum-state-specific dynamics during photo-fragmentation.

[1] J. C. G. Martin, *et al., J. Am. Chem. Soc.* **144**, 9240 (2022).

[2] P. Z. El-Khoury, *et al., J. Chem. Phys.* **132**, 124501 (2010).

[3] I. S. Vinklársek, *et int.* (3 authors), J. Küpper, S. Trippel, arXiv:2308.08006 [physics] (2023).

MO 1.6 Mon 12:30 HS 3044

**Ultrafast photofragmentation studies of CF<sub>3</sub>I-I<sup>-</sup> using mass-selected ion-molecule cluster beam apparatus** — ●XIAOJUN WANG<sup>1,4</sup>, MAHMUDUL HASAN<sup>1</sup>, LIN FAN<sup>1</sup>, YIBO WANG<sup>1</sup>, HUI LI<sup>2</sup>, DANIEL SLAUGHTER<sup>3</sup>, and MARTIN CENTURION<sup>1</sup>

— <sup>1</sup>Department of Physics and Astronomy, University of Nebraska-Lincoln, Lincoln, Nebraska 68588, USA — <sup>2</sup>Department of Chemistry, Nebraska Center for Materials and Nanoscience, and Center for Integrated Biomolecular Communication, University of Nebraska-Lincoln, Lincoln, Nebraska

68588, USA — <sup>3</sup>Chemical Sciences Division, Lawrence Berkeley National Laboratory, 1 Cyclotron Rd., Berkeley, California 94720, USA — <sup>4</sup>Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany.

We describe an apparatus for investigating the excited-state dissociation dynamics of mass-selected ion-molecule clusters by mass-resolving and detecting photofragment-ions and neutrals, in coincidence, using an ultrafast laser operating at high repetition rates. The apparatus performance is tested by measuring the photofragments:  $I^-$ ,  $CF_3I^-$  and neutrals from photoexcitation of the ion-molecule cluster  $CF_3I^-$  using femtosecond UV laser pulses with a wavelength of 266 nm. The experimental results are compared with our ground state and excited state electronic structure calculations as well as the existing results and calculations, with particular attention to the generation mechanism of the anion fragments and dissociation channels of the ion-molecule cluster  $CF_3I^-$  in the charge-transfer excited state.

Reference: Rev. Sci. Instrum. 94, 095111 (2023)

MO 1.7 Mon 12:45 HS 3044

**Coulomb explosion imaging of ultrafast photochemistry in molecular photoswitches** — KIERAN CHEUNG<sup>1</sup>, CLAUD PETER

SCHULZ<sup>2</sup>, ARNAUD ROUZÉE<sup>2</sup>, TILL JAHNKE<sup>3</sup>, DANIEL ROLLES<sup>4</sup>, GIUSEPPE SANSONE<sup>5</sup>, MICHAEL MEYER<sup>3</sup>, MARK BROUARD<sup>1</sup>, TERRY MULLINS<sup>1</sup>, and •KASRA AMINI<sup>2</sup> — <sup>1</sup>Chemistry Research Laboratory, Department of Chemistry, University of Oxford, Oxford OX1 3TA, UK — <sup>2</sup>Max-Born-Institut, Max-Born-Str. 2A, 12489 Berlin, Germany — <sup>3</sup>European XFEL, Schenefeld, Germany — <sup>4</sup>J.R. Macdonald Laboratory, Department of Physics, Kansas State University, Manhattan, KS, USA — <sup>5</sup>Physikalisches Institut, Universität Freiburg, D-79106 Freiburg, Germany

Here, we present an X-ray Coulomb explosion imaging (CEI) study into the photofragmentation and photochemistry of trans-4,4-difluoroazobenzene (DFAB) measured with the COLTRIMS Reaction Microscope at the SQS station of European XFEL. We first provide a systematic study of X-ray fragmentation in DFAB with covariance analysis. We then present pump-probe X-ray CEI measurements of DFAB excited to its first excited state under different visible pump excitation conditions. We discuss the limited ability of trans-DFAB to undergo trans-to-cis isomerization after initial population of its  $S_1$  state, and reveal the onset of a dissociative ionization photodissociation process.

## MO 2: Attosecond Physics I (joint session A/MO)

Time: Monday 11:00–13:00

Location: HS 1098

MO 2.1 Mon 11:00 HS 1098

**Ultrafast photoelectron spectroscopy with odd and even high-order harmonics** — •MARVIN SCHMOLL<sup>1</sup>, BARBARA MERZUK<sup>1</sup>, SAMUEL DISCHER<sup>1</sup>, DOMINIK ERTEL<sup>1</sup>, IOANNIS MAKOS<sup>1</sup>, CLAUD D. SCHRÖTER<sup>2</sup>, THOMAS PFEIFER<sup>2</sup>, ROBERT MOSHAMMER<sup>2</sup>, LUCA POLETTI<sup>3</sup>, FABIO FRASSETTO<sup>3</sup>, and GIUSEPPE SANSONE<sup>1</sup> — <sup>1</sup>Universität Freiburg, Physikalisches Institut, Freiburg, Germany — <sup>2</sup>Max-Planck-Institute for Nuclear Physics, Heidelberg, Germany — <sup>3</sup>CNR-Institute of Photonics and Nanotechnologies, Padova, Italy

High-order harmonic generation (HHG) in noble gases produces odd harmonics of the driving field. Adding a weaker second harmonic one can break the underlying symmetry and achieve both odd and even high-order harmonics.

We present an implementation of a collinear setup for such two-color HHG similar to what was first presented in ref. [1], which allows to adjust the relative phase between the fundamental and second harmonic component. Being implemented in combination with a collinear beamline for XUV-IR interferometry [2] we can perform high stability ultrafast photoelectron spectroscopy using these high order harmonics.

Our first results using Argon as a target gas show the viability of the method by demonstrating delay-dependent oscillations in the photoelectron yield for specific energies. These exhibit a period equal to that of the fundamental driving field as opposed to twice that period, which is known from experiments with odd orders only.

[1] N. Dudovich et al., Nature Phys. 2, 781 (2006)

[2] D. Ertel et al., Rev. Sci. Instrum. 94, 073001 (2023)

MO 2.2 Mon 11:15 HS 1098

**Extreme ultraviolet wave packet interferometry using table-top high harmonic generation** — •SARANG DEV GANESHAMANDIRAM, FABIAN RICHTER, IANINA KOSSE, RONAK SHAH, MARIO NIEBUHR, GIUSEPPE SANSONE, FRANK STIENKEMEIER, and LUKAS BRUDER — Institute of Physics, University of Freiburg, Hermann-Herder-Str. 3, 79104 Freiburg, Germany

Quantum interference techniques such as wave packet interferometry (WPI) in the extreme ultraviolet (XUV) domain set the basis for establishing advanced nonlinear spectroscopy methods in this wavelength regime [1]. These methods are however very difficult to implement at short wavelengths due to the required high phase stability and sensitivity. We are exploring methods based on acousto-optical phase modulation (PM) to solve these problems. First results from applications in seeded FELs and table-top high-harmonic generation (HHG) are promising [2,3]. Here, we will present an interferometer setup specifically designed for application with table-top HHG and discuss current challenges.

[1] S. Mukamel, et al., Multidimensional Attosecond Resonant X-Ray Spectroscopy of Molecules: Lessons from the Optical Regime, Annu. Rev. Phys. Chem. 64, 101 (2013).

[2] A. Wituschek, et al., Tracking attosecond electronic coherences using phase-manipulated extreme ultraviolet pulses, Nat Commun 11, 883 (2020).

[3] A. Wituschek et al., Phase cycling of extreme ultraviolet pulse sequences generated in rare gases, New J. Phys. 22, 092001 (2020).

MO 2.3 Mon 11:30 HS 1098

**Controlling Photoabsorption Interferometrically with Intense Laser Pulses from Microscopic to Macroscopic Gases** — •YU HE<sup>1</sup>, SHUYUAN HU<sup>1</sup>, GERGANA D. BORISOVA<sup>1</sup>, YIZHU ZHANG<sup>1,3</sup>, MARC REBHOLZ<sup>1</sup>, METTE B. GAARDE<sup>2</sup>, CHRISTIAN OTT<sup>1</sup>, and THOMAS PFEIFER<sup>1</sup> — <sup>1</sup>Max-Planck-Institut für Kernphysik, Heidelberg, Germany — <sup>2</sup>Louisiana State University, Baton Rouge, USA — <sup>3</sup>Tianjin University, Tianjin, China

Photoabsorption results from the interference between the incident field and the newly generated one radiated by the induced dipole oscillation. This dipole-emitted field can be controlled by the interplay of pulse propagation and intense laser pulses, giving rise to different absorption lineshapes. By temporally confining this new field through emptying the population of the excited state after its excitation, we achieve a local enhancement of absorbance in transient absorption spectroscopy [1,2]. In addition, in tandem with theory, we experimentally demonstrate the transition of absorption profiles from natural Lorentzian to Fano-like, which then become broader with further emergence of spectral bifurcations, finally turning back to near-Lorentzian lines in optically dense helium [3]. The integrated interferometric scenario in ultrafast absorption spectroscopy provides insights into the behavior of ensembles of dipole emitters and their temporal control. Refs: [1] He et al., Phys. Rev. Lett. 129 273201 (2022). [2] He et al., manuscript submitted (2023). [3] He et al., manuscript in preparation.

MO 2.4 Mon 11:45 HS 1098

**Time- and Frequency-resolved Characterization of Collective Nuclear Dynamics** — •LUKAS WOLFF and JÖRG EVERS — Max-Planck-Institut für Kernphysik Heidelberg, Germany

Mössbauer nuclei have become an important tool for high precision tests and spectroscopy owing to their extremely narrow linewidths and long coherence times. In recent years, ensembles of nuclei embedded in suitably engineered waveguide structures allowed for the observation of cooperative phenomena such as superradiant decay and collective level shifts. This constituted the field of nuclear quantum optics of collective nuclear excitations. A direct and unambiguous characterization of such level schemes in the time or frequency domain alone is challenging and, thus, new data acquisition and evaluation techniques are of great importance to access the underlying collective dynamics [1]. To this end, we study the time- and frequency-resolved collective behaviour of nuclear ensembles upon x-ray pulses with different temporal and spectral shape to extract signatures for collective and nonlinear dynamics of Mössbauer resonances [2]. We expect our

results to help guide future experiments investigating such dynamics using suitably-shaped x-ray pulses and pulse sequences that can be created using time-domain control of nuclear resonances.

- [1] L. Wolff and J. Evers, *Phys. Rev. Res.* **5**, 013071 (2023)  
 [2] L. Wolff and J. Evers, *Phys. Rev. A* **108**, 043714 (2023)

MO 2.5 Mon 12:00 HS 1098

**Designing a Topological Thin-Film X-Ray Cavity** — ●HANNS ZIMMERMANN<sup>1,2</sup> and ADRIANA PÁLFFY<sup>1</sup> — <sup>1</sup>Julius-Maximilians-Universität Würzburg — <sup>2</sup>Universität der Bundeswehr München

A promising platform for the quantum control of high-frequency photons are thin-film cavities, with one or several embedded layers of resonant nuclei such as <sup>57</sup>Fe with a Mössbauer transition at 14.4 keV. At grazing incidence, incoming x-rays couple evanescently to the cavity. In turn, the cavity field drives the nuclear transitions. The resulting nuclear response is well described by a recently-developed quantum optical model based on the electromagnetic Green's function [1,2].

Here, we investigate theoretically a thin-film cavity design with multiple embedded <sup>57</sup>Fe layers, such that its inter-layer couplings are mostly restricted to the nearest neighbouring layers by intercalating additional layers with high electron densities. Via the geometrical properties of these domains and control of the evanescent field pattern, we implement alternating coupling strengths between the resonant layers. We show that this leads to an x-ray photonic realization of the non-hermitian Su-Schrieffer-Heeger model and investigate how for certain configurations localized nuclear excitations emerge at the edges of the cavity.

- [1] X. Kong, et al. *Phys. Rev. A* **102**, 033710 (2020)  
 [2] P. Andrejić and A. Pálffy, *Phys. Rev. A* **104**, 033702 (2021)

MO 2.6 Mon 12:15 HS 1098

**Single-shot electron spectroscopy of highly transient matter** — ●SARA SAVIO<sup>1</sup>, LARS FUNKE<sup>1</sup>, NICLAS WIELAND<sup>1,3</sup>, LASSE WÜELFING<sup>1</sup>, MARKUS ILCHEN<sup>2,3</sup>, and WOLFRAM HELML<sup>1</sup> — <sup>1</sup>Fakultät Physik, Technische Universität Dortmund, Maria-Goeppert-Mayer-Straße 2, 44227 Dortmund, Germany — <sup>2</sup>Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany — <sup>3</sup>University of Hamburg, Middle Way 177, 20148 Hamburg, Germany

Single-shot electron spectroscopy can be used as a tool to investigate photo-ionization processes and the various subsequent relaxation dynamics, ie how the inner shell vacancies are redistributed and filled in atoms and molecules. This work investigates the generation of double-core holes (DCH) in neon atoms with very short lifetimes using the help of intense and tightly focused X-ray pulses at European XFEL at the attosecond frontier. An electron-time-of-flight (e-TOF) spectrometer equipped with a multi-electrostatic lens system followed by a microchannel plate (MCP) based detector is used to specifically collect DCH Auger electrons in single-shot spectroscopy. The wavelength tun-

ability and high X-ray intensity at European XFEL together with this spectroscopic technique enable the study of highly transient systems. Examining the electronic structure of a core-excited system before relaxation can allow for gaining essential insights into ultrafast processes and nonlinear photoabsorption under extreme intensities thus opening a new field of spectroscopy of transient matter.

MO 2.7 Mon 12:30 HS 1098

**Interatomic Coulombic Decay from Auger final states in aqueous solution** — ●ANDREAS HANS<sup>1</sup>, DANA BLOSS<sup>1</sup>, RÉMI DUPUY<sup>2</sup>, FLORIAN TRINTER<sup>3</sup>, UWE HERGENHAHN<sup>3</sup>, OLLE BJÖRNEHOLM<sup>4</sup>, and ARNO EHRESMANN<sup>1</sup> — <sup>1</sup>Universität Kassel und CINSaT, Kassel, Germany — <sup>2</sup>Sorbonne Université, Paris, France — <sup>3</sup>Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — <sup>4</sup>Uppsala University, Uppsala, Sweden

Interatomic Coulombic decay of resonant Auger final states (RA-ICD) has been discovered about a decade ago. Due to the site-selective character of the resonant excitation and the typically emitted slow electrons, RA-ICD has been envisioned to enhance the efficiency of radiation therapies. So far, the mechanism had only been observed experimentally in prototypical van der Waals dimers. Here, we present the transfer of the idea to the liquid phase. To this end we investigate the decay of  $2p \rightarrow 3d$  resonantly excited solvated  $\text{Ca}^{2+}$  ions. We show, that using multi-electron coincidence spectroscopy increases the contrast for slow electrons drastically and that RA-ICD can be readily observed in the liquid phase.

MO 2.8 Mon 12:45 HS 1098

**Attoclock, what can or has actually been measured?** — ●OSSAMA KULLIE — <sup>1</sup>Theoretical Physics, Institute of Physics, University of Kassel

Attoclock is designed to measure the delay time required for a particle to tunnel, or undergo field-ionization, from an atom interacting with a strong laser field. However, some authors claim that the duration the attoclock measures is not a good proxy for tunneling time. In previous works, we showed a model that describes the tunnel- or field-ionization of the attoclock experiment for He- [1] and H-atom [2], in the adiabatic and nonadiabatic field calibrations [3]. In the present talk, we show that it is possible to interpret the attoclock measurement in such a way that real-valued tunnel-time or the delay time due to the barrier region or the classically forbidden region can be determined. Furthermore, we show that in the limit of weak measurement the attoclock provides the interaction time inside the barrier, which is usually measured by the Larmro clock. The limit of thick barrier, the interaction time and the superluminal tunneling are discussed, [1] A. S. Landsman et al, *Optica* **1**, 343 (2014), U. S. Sainadh et al, *Nature* **586**, 75 (2019). [2] C. Hofmann et al. *J. Mod. Opt.* **66**, 1052 (2019). [3] O. Kullie, *Phys. Rev. A* **92**, 052118 (2015), O. Kullie *J. Phys. Commun.* **2**, 065001 (2018), O. Kullie and I. A. Ivanov, arXiv:2005.09938v6.

## MO 3: Novel Spectroscopies

Time: Monday 17:00–19:00

Location: HS 1015

MO 3.1 Mon 17:00 HS 1015

**Two-Dimensional IR Spectroscopy of Bifunctional Vibrational Probes** — ●CLAUDIA GRÄVE, STEFAN FLESCH, LUIS IGNACIO DOMENIANNI, JÖRG LINDNER, and PETER VÖHRINGER — Universität Bonn, Wegelerstr. 12, 53115 Bonn, Germany

The nitrile group is a superb vibrational probe for the dynamics of biomolecular systems.<sup>[1]</sup> Unfortunately, it exhibits a relatively small transition dipole moment, which causes sensitivity issues in IR-spectroscopic studies.

Here, we study the vibrational dynamics of small organic nitriles featuring an azide residue, which can later be utilized for protein incorporation. The two nitriles studied here are 4-azidobenzonitrile ( $\text{N}_3\text{-C}_6\text{H}_4\text{-CN}$ ) and 3-(4-azidophenyl)propionitrile ( $\text{N}_3\text{-C}_6\text{H}_4\text{-CC-CN}$ ).

We report on the linear Fourier-transform infrared spectra, as well as on the femtosecond pump-probe and 2DIR spectroscopy. The linear spectra in the azide and nitrile stretching region are highly complex due to Fermi resonances involving the  $\text{N}_3$ -fundamental. Furthermore, the 2DIR spectra reveal the detailed vibrational energy flows between the azide and the nitrile groups separated by the phenyl ring. Finally, coherent oscillations resulting from impulsive excitation of the Fermi

multiplets were observed by means of pump-probe measurements. Our results suggest that propionitrile is a superior vibrational probe as compared to the bare CN.

### Literature:

- [1] H. Kim and M. Cho, *Chem. Rev.* **2013**, *113*, 5817-5847.

MO 3.2 Mon 17:15 HS 1015

**Nonlinear 2D spectroscopy of single molecules** — ●SIMON DURST, SANCHAYEETA JANA, and MARKUS LIPPITZ — Universität Bayreuth

Fluorescence-detected 2D electronic spectroscopy (F-2DES) allows the measurement of ultrafast electron dynamics in complex systems while disentangling this spectral information from energetically similar phenomena, such as molecular vibration. Measuring the 2D spectra of single molecules instead of molecular ensembles should provide an even clearer picture of the underlying physics and give insight into the statistical distribution of optical properties and the environment of the molecules.

To measure these spectra we use four collinear, phase-modulated femtosecond pulses, generated by a four-arm Mach-Zehnder Interfer-

ometer to excite single molecules with a confocal microscope. Their emitted fluorescence is modulated at the mixing frequencies of the individual pulses, so we can use phase-sensitive lock-in detection to detect the nonlinear signal and separate it from linear effects. This measurement technique promises high spectral, temporal and spatial resolution.

In this talk, we present our setup and method to measure the 2D spectra of single dibenzoterrylene (DBT) molecules, which are immobilised in a PMMA matrix. We show results from these measurements and compare them with the ensemble data.

MO 3.3 Mon 17:30 HS 1015

**Coherent multidimensional spectroscopy of PTCDA monomers on argon clusters** — •YILIN LI, ARNE MORLOK, ULRICH BANGERT, FRIEDEMANN LANDMESSER, FRANK STIENKEMEIER, and LUKAS BRUDER — Institute of Physics, University of Freiburg, Germany

The interaction and dynamics between single molecules and the environment is of great interest yet challenging to study. In our approach, we dope rare gas clusters with single molecules in the gas phase [1]. This provides us an ensemble of doped nanoparticles, each particle well isolated from other environmental influences. We study these systems with coherent multidimensional electronic spectroscopy, a versatile technique providing further insights into intra- and intermolecular couplings on ultrashort time scales [2,3]. Recently we started analysing 2D beating maps to obtain information about the electronic and vibrational coherences, which are otherwise covered by line broadening mechanisms, together with pump-probe measurements to study the decoherence and the population decay of the system. First results of PTCDA monomers on argon clusters will be presented.

[1] M. Bohlen et al., *J. Chem. Phys.* 156, 034305 (2022)

[2] L. Bruder et al., *Nat. Commun.* 9, 4823 (2018)

[3] U. Bangert, F. Stienkemeier, L. Bruder, *Nat. Commun.* 13, 3350 (2022)

MO 3.4 Mon 17:45 HS 1015

**Next generation fs transient spectroscopy based on 1030 nm pump** — •FERDINAND BERGMEIER and EBERHARD RIEDLE — Lehrstuhl f. BioMolekulare Optik, Fakultät f. Physik, LMU München

The measurement of transient electronic spectra, vital for unraveling complex photophysical, chemical, and biological processes, is achieved through fs excitation and broadband UV to NIR detection. We present an innovative transient absorption spectrometer based on a newly engineered kHz noncollinear optical parametric amplifier (NOPA) pumped by a modern Yb-based 250 fs industrial-grade pump laser. Coupled with a fully redesigned CaF<sub>2</sub>-based ultrabroad probe pulse, this spectrometer retains the positive aspects of the earlier Ti:Sa-based system. This ensures compactness with minimal optics and adjustments, boasting a warm-up time of under ten minutes.

The probe continuum spans 320 to 950 nm with 1030 nm pumping and to below 280 nm with 515 nm pumping. The pump is tunable from 220 to 950 nm with a sub-20 fs pulse length and no spectral gaps. The instrumental response function is below 40 fs. Single-shot spectral referencing achieves a sensitivity of approximately 20 uOD for a half-hour measurement, near the shot noise limit. The continuum splitting onto the signal and reference arm is achromatic and dispersion-free. A perfectly round probe focus is achieved with a Schief-Spiegler.

With the extreme precision of the setup, we address surface-resolved coherent artifacts that are crucial for investigating processes on the 100 fs time scale. Detailed reference and precise chirp measurements of the probe continuum enable sub-10 fs time-zero correction.

MO 3.5 Mon 18:00 HS 1015

**Effects of Strong Coupling on the Chemiluminescent Reaction of Dioxetane** — •MARKUS KOWALEWSKI and MAHESH GUDEM — Stockholm University, Stockholm, Sweden

Chemiluminescence, seen in phenomena like firefly light emission, involves thermally activated chemical processes. Dioxetane, the smallest cyclic peroxide, exhibits chemiluminescence with a lower quantum yield than firefly dioxetane. Utilizing strong light-matter coupling as an alternative strategy, we investigate its impact on dioxetane's chemiluminescence reaction within an optical cavity. The extended Jaynes-Cummings model is used to incorporate the cavity couplings for electronic and vibrational degrees of freedom. Results reveal that cavity interactions can either accelerate or suppress the formation of excited-state products in the dioxetane decomposition, depending on molecular orientation relative to cavity polarization.

MO 3.6 Mon 18:15 HS 1015

**Exploring the scaling factors for infrared modes of polycyclic aromatic nitrogen heterocycles** — •DOMENIK SCHLEIER<sup>1,2</sup>, JERRY KAMER<sup>1</sup>, JONATHAN MARTENS<sup>3</sup>, GIEL BERDEN<sup>3</sup>, JOS OOMENS<sup>3,4</sup>, and JORDY BOUWMAN<sup>5,6,7</sup> — <sup>1</sup>Leiden Observatory, Leiden, The Netherlands — <sup>2</sup>Uni Paderborn, Paderborn, Germany — <sup>3</sup>FELIX Laboratory, Nijmegen, The Netherlands — <sup>4</sup>University of Amsterdam, Amsterdam, The Netherlands — <sup>5</sup>Laboratory for Atmospheric and Space Physics, Boulder, USA — <sup>6</sup>Department of Chemistry, Boulder, USA — <sup>7</sup>Institute for Modeling Plasma, Atmospheres and Cosmic Dust, Boulder, USA

Infrared (IR) emission bands by interstellar Polycyclic Aromatic Hydrocarbons (PAHs) offer detailed insights into the chemistry and physics of the interstellar medium. It has been suggested that hetero atom substituted species such as Polycyclic Aromatic Nitrogen Heterocycles (PANHs) also contribute to the aromatic IR emission bands. The analysis of the emission bands, and thus the interpretation of the molecular characteristics of the carriers, heavily relies on the use of density functional theory (DFT) calculated IR spectra. However, there are significant challenges in accurately predicting the experimental IR band positions, particularly for PANH emission vibrational modes around 6  $\mu\text{m}$ . In this work, we present gas-phase mid-infrared (mid-IR) spectra of cationic 3-azafluoranthene and its protonated congener to investigate their experimental IR band positions in relation to DFT calculated bands.

MO 3.7 Mon 18:30 HS 1015

**VUV Photoionization spectroscopy of cyano-substituted PAHs** — •MADHUSREE ROY CHOWDHURY<sup>1,2</sup>, GUSTAVO GARCIA<sup>2</sup>, HELGI HRODMARSSON<sup>3</sup>, JEAN-CHRISTOPHE LOISON<sup>4</sup>, and LAURENT NAHON<sup>2</sup> — <sup>1</sup>Institute of Physics and CINSA<sup>T</sup>, University of Kassel, Kassel, 34132, Germany — <sup>2</sup>Synchrotron SOLEIL, L'Orme des Merisiers, Départementale 128, 91190 Saint Aubin, France — <sup>3</sup>LISA Laboratory, Universités Paris Est-Paris Diderot-Paris 7, UMR CNRS 7583, Créteil, France — <sup>4</sup>Université Bordeaux, CNRS, Bordeaux INP, ISM, UMR 5255, Talence F-33400, France

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous in the interstellar medium (ISM), accounting for about a quarter of the total carbon mass of the ISM. The aromatic infrared emission bands (AIBs) are the signatures of the existence of PAHs in the ISM. Although their presence is well acknowledged, the individual detection of PAHs is notoriously difficult. Substituted PAHs being less symmetric are promising candidates, leading to the detection of the two isomers of cyanonaphthalenes (McGuire et.al. *Science* 2021) in TMC-1. Upon absorbing the VUV radiation, PAHs relax via photoionization and photodissociation processes in competition with radiative cooling. The VUV photoionization and fragmentation of 1- and 2-cyanonaphthalenes is studied using a double imaging photoelectron photoion coincidence spectrometer (i<sup>2</sup>PEPICO). The KE distribution of the photoelectrons is useful to model the photoelectric heating for radiation fields while the state-selected fragmentation of the cations shed light on the photostability of the cyano substituted PAHs.

MO 3.8 Mon 18:45 HS 1015

**Photoelectron Photoion Multicoincidence Study of Micro-Solvated Thymine Molecules** — •BRENDAN WOUTERLOO<sup>1</sup>, STEPHAN SCHMITZ<sup>1</sup>, MADHUSREE ROY-CHOWDHURY<sup>2</sup>, GUSTAVO GARCIA-MACIAS<sup>2</sup>, LAURENT NAHON<sup>2</sup>, FRANK STIENKEMEIER<sup>1</sup>, and SEBASTIAN HARTWEG<sup>1</sup> — <sup>1</sup>Institute of Physics, University of Freiburg — <sup>2</sup>Synchrotron SOLEIL, St. Aubin, France

Studying biomolecules, such as amino acids and nucleobases, in the gas phase allows unparalleled detail and fundamental insights into energetics and dynamics at the molecular level. However, in-vivo biomolecular systems exist mostly in the condensed phase, which can affect ionisation energies as well as fragmentation and relaxation pathways. Micro-solvation can be exploited to bridge the gap between the gas and the condensed phases and allows for the application of double imaging photoelectron photoion photoion coincidence (i<sup>2</sup>PEPIPICO) spectroscopy. The ion-ion coincidence detection enables identification of signals arising from non-local auto-ionisation processes of clusters, such as from intermolecular Coulombic decay. These processes are important to the field of radiation chemistry since the production of low energy electrons can trigger reactions which damage biological material. Here, preliminary results of a i<sup>2</sup>PEPIPICO study of small water clusters (H<sub>2</sub>O)<sub>n</sub> (n=1-10) and water-thymine complexes, will be presented.

## MO 4: Strong-field Ionization and Imaging (joint session MO/A)

Time: Monday 17:00–18:30

Location: HS 3044

MO 4.1 Mon 17:00 HS 3044

**Strong-Field Ionization of Nitrous Oxide Molecule by Short Femtosecond Laser Pulses** — ●FERAS AFANEH — Physics Department, The Hashemite University, P.O. Box 150459, Zarqa 13115, Jordan.

The dissociative photoionization of nitrous oxide molecules, an important atmospheric trace gas, induced by circularly and elliptically polarized laser pulses has been studied by photoelectron photoion coincidence (PEPICO) spectroscopy. PEPICO spectra were used to identify different dissociative photoionization channels. It is observed that the ionized  $N_2O$  and its fragments have different correlation trends at different polarization schemes. The relative contributions of different double and triple dissociative ionization channels to the total fragment ion yield are also deduced from the coincident spectra of these channels. The results show that the double dissociative photoionization channels: the denitrogenation ( $N_2O^{2+} \rightarrow N^+ + NO^+$ ) and the deoxygenation ( $N_2O^{2+} \rightarrow O^+ + N_2^+$ ). Furthermore, a considerable contribution of the triple dissociative ionization channels to the total fragment ion yield is also observed. The channels " $N^+ + NO^+$ " and " $O^+ + N_2^+$ " can be explained by dissociation via the  $X^3\Sigma^-$  and  $1^3\Pi$  states of  $N_2O^{2+}$  as the major peaks in the measured kinetic energy release spectra suggested.

MO 4.2 Mon 17:15 HS 3044

**Theoretical semiclassical modelling of Laser Induced Electron Diffraction (LIED)** — ●ÁLVARO FERNÁNDEZ<sup>1,2</sup>, ARMIN ISKE<sup>3</sup>, ANDREY YACHMENEV<sup>1,4</sup>, and JOCHEN KÜPPER<sup>1,2,4</sup> — <sup>1</sup>Deutsches Elektronen-Synchrotron DESY — <sup>2</sup>Department of Physics, Universität Hamburg — <sup>3</sup>Department of Mathematics, Universität Hamburg — <sup>4</sup>Center for Ultrafast Imaging, Universität Hamburg

Experimental techniques for molecular imaging underwent a very fast development in the past decades. From a broad range of novel techniques, laser induced electron diffraction (LIED) [1] stands out because of its high spatiotemporal resolution, high cross section, and lack of structural damage compared to other modern techniques. However, the complexity of this technique causes the necessity of its own theory to understand the results. A general and accurate quantum simulation of the experiment is, to this date, unfeasible and, for this reason, semiclassical models [2] have arisen as useful predicting algorithms.

In this talk, a comprehensive analysis of the LIED experiment using a semiclassical model will be given. With this model, we can obtain efficient simulations of the outcome for flexible configurations of molecular geometries. The quality of the results will depend on several factors such as the choice of ionisation theory or electrostatic potential model. An study of the relevance of these factors in the computation of effective cross section for high energy electrons will be presented during the talk.

[1] Karamatskos, E. T., *et al.*, *J. Chem. Phys.*, **150**, 24 (2019)

[2] Wiese, J., *et al.*, *Phys. Rev. Research*, **3**, 013089, (2021)

MO 4.3 Mon 17:30 HS 3044

**Wavefunction Reconstruction of Excitonic Edge States using Machine Learning** — ●ARITRA MISHRA and ALEXANDER EIFELD — Max Planck Institute for the Physics of Complex Systems, Dresden, Germany

A typical problem in quantum mechanics is to reconstruct the eigenstate wave functions from measured data. In the case of molecular aggregates, the information about the excitonic eigenstates is important to understand the optical and transport properties [1]. It has been shown for a linear and a 2D arrangement of the aggregate molecules that such a reconstruction is possible from the spatially resolved near field absorption spectra [2].

Here, we consider the aggregates arranged in two sublattices in a 2D arrangement, each sub lattice having a particular orientation of the molecules as described in [3]. Interestingly, such an arrangement can lead to the formation of topological excitonic edge states. We study the reconstruction of the excitonic wave function of such a system from the near field absorption spectra. The reconstruction is further investigated in the presence of disorder in the Hamiltonian and noise added to the spectra.

[1] X. Gao and A. Eisfeld, *J. Phys. Chem. Lett.* **9**, 6003 (2018)

[2] F. Zheng, X. Gao and A. Eisfeld, *Phys. Rev. Lett.* **123**, 163202

(2019)

[3] J. Yuen-Zhou, S. K. Saikin, N. Y. Yao and A. Aspuru-Guzik, *Nature Materials* **13**, 1026 (2014)

MO 4.4 Mon 17:45 HS 3044

**Molecular self-probing for the visualisation of vibrational wave-packet dynamics and its laser-induced modification** — ●GERGANA D. BORISOVA<sup>1</sup>, PAULA BARBER BELDA<sup>1</sup>, SHUYUAN HU<sup>1</sup>, PAUL BIRK<sup>1</sup>, VEIT STOOSS<sup>1</sup>, MAXIMILIAN HARTMANN<sup>1</sup>, ROBERT MOSHAMMER<sup>1</sup>, ALEJANDRO SAENZ<sup>2</sup>, CHRISTIAN OTT<sup>1</sup>, and THOMAS PFEIFER<sup>1</sup> — <sup>1</sup>Max-Planck-Institut für Kernphysik, 69117 Heidelberg — <sup>2</sup>Institut für Physik, Humboldt-Universität zu Berlin, 12489 Berlin

We present an all-optical pump-control scheme for molecular wave-packet (WP) visualisation and control, where the molecular ground state acts as an intrinsic self-probe of the system, imprinting the evolution of an excited wave packet onto the coherent dipole emission [1]. In a proof-of-principle experiment, coherent extreme ultraviolet (XUV) light creates a vibrational wave packet in the electronically excited  $D^1\Pi_u3\pi\pi$  state of neutral  $H_2$ . Measured XUV absorption spectra of the  $D$ -state vibronic resonances provide access to the WP dynamics after reconstruction of the time-dependent dipole response [2], which probes the vibrating wave packet through the molecular ground state. An intense near-infrared (NIR) pulse, applied shortly after the WP excitation, is used to control the wave-packet evolution and through this its revival. With increasing NIR intensity the WP revival shifts to earlier times. We identify state-specific NIR-induced phase shifts as the origin of the observed time shifts, which can be applied even to complex molecular systems to coherently steer the recovery of vibrational wave packets on electronically excited potential-energy curves at a desired time. [1] arXiv:2301.03908 [2] PRL **121** (2018) 173005

MO 4.5 Mon 18:00 HS 3044

**Ultrafast imaging of rare-gas clusters from their formation to their ionization dynamics** — ●ALESSANDRO COLOMBO for the RareGas Clusters at SwissFEL-Collaboration — ETH Zurich, 8049 Zürich, Switzerland

Coherent Diffraction Imaging (CDI) experiments performed at Free-Electron Lasers (FELs) allow to capture femtosecond snapshots of isolated nanosamples, and are a unique tool for spatially and temporally resolve ultrafast electron dynamics at the nanoscale. Isolated atomic clusters represent the perfect prototypical system for such investigations, thanks to their simple electronic structure and their highly tunable size and shape [1]. We present imaging studies performed at SwissFEL on mixed Ar/Xe nanoclusters produced by supersonic expansion into vacuum. Imaging results at 1 keV photon energy reveal fascinating and unexpected shapes at a spatial resolution of few nanometers, which stimulate further research about the thermodynamics and kinematics of these systems. Additionally, the FEL was tuned to photon energies around 0.67 keV, corresponding to the xenon 3d electronic resonance. Fluctuations of the scattering cross-section of Xe can be identified in the CDI reconstructions even several tens of eV away from the 3d edge. The observed behavior can be interpreted as the footprint of ultrafast ionization dynamics happening within the FEL pulse duration, giving insights into the evolution of high charge states, their optical properties and the contribution of transient electronic resonances.

[1] A. Colombo and D. Rupp. (2023) in *Structural Dynamics with X-ray and Electron Scattering*, Royal Society of Chemistry, *in press*

MO 4.6 Mon 18:15 HS 3044

**High repetition rate ultrafast electron diffraction with direct electron detection** — FERNANDO RODRIGUEZ DIAZ, MARK MERÖ, and ●KASRA AMINI — Max-Born-Institut, Max-Born-Str. 2A, 12489 Berlin, Germany

Ultrafast electron diffraction (UED) is a power tool that can monitor the nuclear dynamics of photo-induced gas-phase reactions in real-time with picometre and <250-fs spatiotemporal resolution. However, the temporal resolution of state-of-the-art gas-phase UED setups, often operating at <1-kHz, is insufficient to time-resolve rapidly evolving photo-induced processes (e.g., <350-fs predicted timescale of photoisomerization which plays a crucial role in vision). The limited temporal resolution is due to the severe space-charge dispersion experienced in electron pulses containing  $10^4$  to  $10^5$  electrons.

Here, we present a new 30-kHz 100-keV UED setup employing direct electron detection that will be capable of performing time-resolved measurements of photochemical reactions in gas-phase molecules with <100-fs temporal resolution, going beyond the current state-of-the-art in keV and MeV gas-phase UED. This is made possible by operating below the severe space-charge dispersion regime using electron pulses

containing very few electrons ( $<10^2$ ) but with sufficient electron flux ( $>10^6$  electrons/s) thanks to the high repetition rate of our system. Latest results from the commissioning of our pump-probe UED instrument is presented with details of the current implementation of radiofrequency-compressed electron pulses and the correction of time-of-arrival jitter issues.

## MO 5: Interaction with Strong or Short Laser Pulses I (joint session A/MO)

Time: Tuesday 11:00–13:00

Location: HS 1010

### Invited Talk

MO 5.1 Tue 11:00 HS 1010

**Strong-field coherent control in the extreme ultraviolet domain** — ●F. RICHTER<sup>1</sup>, U. SAALMANN<sup>2</sup>, M. WOLLENHAUPT<sup>3</sup>, E. ALLARIA<sup>4</sup>, C. CALLEGARI<sup>4</sup>, M. DANAILOV<sup>4</sup>, L. GIANESSI<sup>4</sup>, M. ZANGRANDO<sup>4</sup>, and L. BRUDER<sup>1</sup> — <sup>1</sup>Institute of Physics, University of Freiburg — <sup>2</sup>Max-Planck-Institut für Physik komplexer Systeme, Dresden — <sup>3</sup>Institute of Physics, University of Oldenburg — <sup>4</sup>Elettra - Sincrotrone Trieste S.C.p.A., Trieste, Italy

Coherent control drew a lot of interest in recent years spanning over various fields of research regarding the promising abilities for quantum computing and precision measurements. Coherent control extended to the strong-field regime is particularly promising for the manipulation of matter and the control of photochemical reactions. In this work, we develop a scheme to extend strong-field coherent control to the XUV domain. With intense XUV pulses, we induce Rabi oscillations in atoms, leading to Autler-Townes level splittings in the photoelectron spectra [1]. In the near infrared domain, the feasibility to coherently control the population of the Autler-Townes doublet has been shown, based on chirp manipulation of the laser pulses [2,3]. To establish comparable schemes in the XUV domain, we implement chirp control of the XUV pulses from the free electron laser FERMI. By manipulating the chirp of the XUV pulses in a controlled way, we demonstrate strong-field coherent control of Autler-Townes states in the XUV domain.

[1] S. Nandi et al. *Nature* 608, 488\*493 (2022). [2] M. Wollenhaupt et al., *Appl. Phys. B* 82, 183\*188 (2006). [3] U. Saalman et al., *Phys. Rev. Lett.* 121, 153203 (2018).

MO 5.2 Tue 11:30 HS 1010

**Intra-cavity photoelectron tomography and pulsed standing waves at 100 MHz repetition rate** — ●JAN-HENDRIK OELMANN, TOBIAS HELDT, LENNART GUTH, NICK LACKMANN, LUKAS MATT, THOMAS PFEIFER, and JOSÉ R. CRESPO LÓPEZ-URRUTIA — Max-Planck-Institut für Kernphysik, Heidelberg, Germany

To get access to multiphoton ionization studies at high laser intensities ( $\sim 10^{13}$  W/cm<sup>2</sup>) while maintaining the high 100 MHz repetition rate of the driving frequency comb, we have recently developed a novel polarization-insensitive enhancement cavity with an integrated velocity-map imaging (VMI) spectrometer [1, 2]. Polarization-controlled pulse pairs with a variable time delay allow pump-probe experiments. With this polarization control but in single-pulse operation, we were able to tomographically reconstruct 3D photoelectron angular distributions [3] from xenon MPI at 100 MHz repetition rate, revealing resonant Rydberg states during ionization.

Now, we use counter-propagating pulses colliding at the focus to generate intense femtosecond standing waves in the cavity. We probe the phase of these at the nanometer scale using photoemission from a tungsten nanotip. Colliding pulses offer the dual advantage of enabling Doppler-free excitation schemes and of reducing the interaction volume at the focus.

[1] J.-H. Oelmann *et al.*, *Rev. Sci. Instrum.*, 93(12), 123303 (2022). [2] J. Nauta *et al.*, *Opt. Lett.* 45(8), 2156 (2020). [3] M. Wollenhaupt *et al.*, *Appl. Phys. B* 95(4), 647-651 (2009).

MO 5.3 Tue 11:45 HS 1010

**Reconstruction of Three Dimensional Molecular Density from XFEL Scattering Images using Machine Learning** — ●SIDDHARTHA PODDAR, ULF SAALMANN, and JAN MICHAEL ROST — Max Planck Institute for the Physics of Complex Systems

As the three-dimensional electron density profile recovery technique for a single macro-molecule from a large dataset of coherent diffraction images generated using an X-ray free-electron laser, I have applied an unsupervised machine learning algorithm namely Generative

Adversarial Network (GAN). It learns to mimic the high-dimensional distribution of given images by generating its own 'fake' distribution of images with the help of a deep convolutional neural network called the discriminator which distinguishes samples drawn from the original and fake distributions. To generate samples for this fake distribution of images, GAN creates and constantly modifies a three-dimensional structure. This structure is claimed to be unique and an equivalent version of the target electronic density profile of the molecule.

MO 5.4 Tue 12:00 HS 1010

**Retrieval of the time-dependent bond length in a molecule from photoelectron momentum distributions using deep learning** — ●NIKOLAY SHVETSOV-SHILOVSKIY and MANFRED LEIN — Leibniz Universität Hannover

We apply a convolutional neural network (CNN) to photoelectron momentum distributions produced by strong-field ionization in order to retrieve the time-varying bond length in the dissociating two-dimensional H<sub>2</sub><sup>+</sup> molecule. We consider the pump-probe scheme and treat the motion of the atomic nuclei either classically, semiclassically, or quantum mechanically. In all these cases, the CNN trained on momentum distributions with fixed internuclear distances [1] predicts the time-dependent bond length with a good accuracy. We investigate whether the neural network can also simultaneously retrieve both the internuclear distance and the velocity with which it increases. Therefore, our results show that deep learning can be used not only for static, but also for dynamic molecular imaging.

[1] N. I. Shvetsov-Shilovski and M. Lein, *Phys. Rev. A* 105 L021102 (2022).

MO 5.5 Tue 12:15 HS 1010

**Shaped free electron vortices** — ●DARIUS KÖHNKE, TIM BAYER, and MATTHIAS WOLLENHAUPT — Carl von Ossietzky university Oldenburg, Institute of Physics, Germany

Since their first theoretical proposal [1] and their experimental demonstration [2], free electron vortices have attracted significant attention. Very recently, a novel category of electron spirals, termed "reversible electron spirals" [3], was introduced. Departing from the conventional approach of employing a constant delay between two subpulses, two chirped subpulses were used. Building on this concept, we introduce tailored free electron vortices in multiphoton ionization (MPI) using two subpulses with circular polarization of opposite handedness, modulated by non-trivial spectral phase functions. Through the utilization of different MPI pathways, the quantum system multiplexes the fields of the subpulses, generating multiple complex spectral phases. These spectral phases are encoded in continuum states characterized by different magnetic quantum numbers. The interference of these continuum states gives rise to multiple interferograms of different symmetry that are multiplexed into a single 3D photoelectron momentum distribution. To demultiplex these interferograms and extract the encoded spectral phases, we perform photoelectron tomography and employ Fourier analysis on the measured wave packet. This approach enables the retrieval of spectral information, both from the input laser fields and signatures of the ionization process, embedded within the interferograms. [1] *Phys. Rev. Lett.* 115, 113004 (2015), [2] *Phys. Rev. Lett.* 118, 053003 (2017), [3] *Phys. Rev. A* 106, 043110 (2022)

MO 5.6 Tue 12:30 HS 1010

**Coherent control of 6Li multiphoton ionization by a bichromatic laser field** — ●SILVA MEZINSKA<sup>1</sup>, KLAUS BARTSCHAT<sup>2</sup>, THOMAS PFEIFER<sup>1</sup>, and ALEXANDER DORN<sup>1</sup> — <sup>1</sup>Max-Planck-Institute for Nuclear Physics, Heidelberg, Germany — <sup>2</sup>Drake University, Des Moines, Iowa, USA

This work presents a coherent 6Li multiphoton ionization control by a bichromatic laser field at 780/390 nm. In particular, we demon-

strate a control of the left-right asymmetry of the photoelectron angular distributions with respect to the plane orthogonal to the laser polarization direction with a subwavelength accuracy. In addition, we also consider a delay scan between the two harmonics extending between the second-harmonic pulse advancing the fundamental pulse and vice versa. Here, we study the delay-dependent features of the photoelectron spectra when the two harmonics are temporally overlapping and non-overlapping. All the experimental results are compared with calculations based on the solution of the time-dependent Schrödinger equation in the single-active electron approximation.

MO 5.7 Tue 12:45 HS 1010

**Nonspreading relativistic electron wavepacket in a strong laser field** — ●ANDRE G. CAMPOS, KAREN Z. HATSAGORTSYAN, and CHRISTOPH H. KEITEL — Max Planck Institute for Nuclear Physics

A solution of the Dirac equation in a strong laser field presenting a nonspreading wave packet in the rest frame of the electron is derived. It consists of a generalization of the self-accelerating free electron wave packet [Kaminer et al. Nature Phys. 11, 261 (2015)] to the case with the background of a strong laser field. Built upon the notion of nonspreading for an extended relativistic wavepacket, the concept of Born rigidity for accelerated motion in relativity is the key ingredient of the solution. At its core, the solution comes from the connection between the self-accelerated free electron wave packet and the eigenstate of a Dirac electron in a constant and homogeneous gravitational field via the equivalence principle. The solution is an essential step towards the realization of the laser-driven relativistic collider [Meuren et al. PRL 114, 143201 (2015)], where the large spreading of a common Gaussian wave packet during the excursion in a strong laser field strongly limits the expectable yields.

## MO 6: Ultracold Molecules and Precision Spectroscopy (joint session MO/Q)

Time: Tuesday 11:00–13:00

Location: HS 3044

MO 6.1 Tue 11:00 HS 3044

**Laser cooling of Barium Monofluoride** — ●SEBASTIAN ALEJANDRO MORALES RAMIREZ<sup>1</sup>, MARIAN ROCKENHÄUSER<sup>1</sup>, FELIX KOGEL<sup>1</sup>, PHILLIP GROSS<sup>1</sup>, TATSAM GARG<sup>1</sup>, and TIM LANGEN<sup>1,2</sup> — <sup>1</sup>Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57, 70569, Stuttgart, Germany — <sup>2</sup>Atominstytut, TU Wien, Stadionallee 2, 1020 Vienna, Austria

Laser cooling of molecules has made remarkable progress over the last years, and a wide variety of molecular species from diatomics to polyatomics can now be routinely cooled. Recently, significant efforts have been made to add barium monofluoride (BaF) to the list of laser-coolable species, as this molecule shows great promise for various precision measurement applications and cold chemistry. Here, we report on the first experimental realization of Sisyphus cooling of such BaF molecules. Our progress is enabled by high resolution absorption spectroscopy of BaF's intricate level structure and a detailed modelling of the resulting cooling forces. In order further understand also the collisional properties of BaF, we perform simultaneous absorption spectroscopy of BaF and calcium monofluoride (CaF) molecules. This gives valuable insights into the thermalisation processes occurring inside a cryogenic buffer gas cell.

MO 6.2 Tue 11:15 HS 3044

**Towards a MOT of AlF molecules** — ●SID WRIGHT — Fritz-Haber-Institut der Max Planck Gesellschaft, Berlin

Aluminium monofluoride (AlF) is a promising candidate for laser cooling and trapping. The primary laser cooling transition at 227.5 nm is extremely strong and highly vibrationally diagonal, making it feasible to slow a molecular beam from 200 m/s to rest in around 1 cm. This offers the potential to greatly increase the number and density of molecules available for ultracold experiments.

In this talk, I will present the latest progress towards a magneto-optical trap (MOT) of AlF molecules, focusing on the first laser slowing results, and our development of a slow, continuous molecular beam source.

MO 6.3 Tue 11:30 HS 3044

**Low-energy collisions between two indistinguishable tritium-bearing hydrogen molecules: HT+HT and DT+DT** — ●RENAT SULTANOV — Odessa College, Department of Mathematics — 201 W. University Blvd., Odessa, TX 79764 USA

Elastic and rotational energy transfer collisions between two tritium-containing hydrogen molecules are computed at low- and very low energies, down to ultra-cold temperatures:  $T \simeq 10^{-8}$  K. A pure quantum-mechanical approach is applied. A high-quality global six-dimensional potential energy surface (PES) has been appropriately modified and used in these calculations. In the case of the symmetrical  $H_2+H_2$  or  $D_2+D_2$  collisions one can use the original  $H_4$  PES as it is, i.e. without transformations. However, in the case of the non-symmetrical (or symmetry-broken)  $HD+H_2/D_2$ ,  $HT+HT$ ,  $DT+DT$  scattering systems one should also apply the original  $H_4$  potential (PES), but propagation (solution) of the Schrödinger equation runs (in this case) over the corrected Jacobi vector [1,2].

1. R. A. Sultanov, D. Guster, S. K. Adhikari, Phys. Rev. A 85,

052702 (2012).

2. R. A. Sultanov, D. Guster, S. K. Adhikari, J. Phys. B 49 (2016) 015203.

MO 6.4 Tue 11:45 HS 3044

**First laser spectroscopy of a rovibrational transition in the molecular hydrogen ion  $H_2^+$**  — ●MAGNUS ROMAN SCHENKEL, SOROOSH ALIGHANBARI, and STEPHAN SCHILLER — Institut für Experimentalphysik, Heinrich-Heine-Universität Düsseldorf, 40225 Düsseldorf, Germany

The molecular hydrogen ion  $H_2^+$  is the simplest molecule and has been the subject of innumerable theoretical studies, culminating in highly precise predictions of its level energies [1]. Comparisons of these predictions and measured transition frequencies would offer excellent opportunities in fundamental physics that go beyond the results achieved with the related  $HD^+$  [2]: a direct determination of the proton-electron mass ratio. In this work we report the first vibrational laser spectroscopy of para- $H_2^+$  between low-lying rovibrational levels [3]. We observed a first overtone electric quadrupole (E2) transition at 2.4  $\mu\text{m}$  and determined its spin-averaged frequency with  $1.2 \times 10^{-8}$  fractional uncertainty, finding agreement with theory. By using  $HD^+$  as a test molecule, we also show that E2 spectroscopy is possible with  $1 \times 10^{-12}$  uncertainty. This demonstrates that determining  $m_p/m_e$  spectroscopically with competitive accuracy is a realistic prospect.

This work has received funding from DFG and NRW via grants INST-208/774-1 FUGG, INST-208/796-1 FUGG and from the ERC (grant No. 786306, \*PREMOL\*).

[1] V. I. Korobov and J.-P. Karr, Phys. Rev. A 104, 032806 (2021).

[2] S. Alighanbari et al., Nat. Phys. 19, 1263 (2023).

[3] M. R. Schenkel et al., Nat. Phys., to appear (2023).

MO 6.5 Tue 12:00 HS 3044

**Frequency metrology system for spectroscopy of molecular hydrogen ions in ALPHATRAP** — ●V. VOGT<sup>1</sup>, I.V. KORTUNOV<sup>1</sup>, K. SINGH<sup>2</sup>, A. KULANGARA THOTTUNGAL GEORGE<sup>2</sup>, B. TU<sup>2,3</sup>, C.M. KÖNIG<sup>2</sup>, F. RAAB<sup>2</sup>, J. MORGNER<sup>2</sup>, T. SAILER<sup>2</sup>, V. HAHN<sup>2</sup>, F. HEISSE<sup>2</sup>, M. BOHMAN<sup>2</sup>, K. BLAUM<sup>2</sup>, S. STURM<sup>2</sup>, and S. SCHILLER<sup>1</sup> — <sup>1</sup>Institut für Experimentalphysik, Univ. Düsseldorf, 40225 Düsseldorf — <sup>2</sup>Max-Planck-Institut für Kernphysik, 69117 Heidelberg — <sup>3</sup>Institute of Modern Physics, Fudan University, Shanghai 200433

At MPIK, an experiment is in preparation aiming at ultra-high precision vibrational spectroscopy of single molecules  $H_2^+$  and  $HD^+$  in the Penning-trap apparatus ALPHATRAP. We require laser light at 1.1  $\mu\text{m}$  and 5.48  $\mu\text{m}$ , respectively, with linewidth 10 Hz, instability below 1 Hz, and absolute frequency measurement capability with uncertainty below  $10^{-13}$ . In addition the laser light must be available 24/7, tunable and switchable under computer control so as to implement appropriate molecule interrogation schemes. We have developed a laser system, similar to [1,2], consisting of spectroscopy laser, reference cavity, transfer laser, frequency comb, hydrogen maser and GNSS receiver at the U. Düsseldorf and transferred it to MPIK, where it has been put into operation again and refined. To transport the spectroscopy light to the Penning-trap, optical fibers with path length cancellation will be implemented. We report the current performance of the system and discuss whether it satisfies the requirements of the experiment.

- [1] I. V. Kortunov et al., Nat. Phys. 17, 569 (2021)  
 [2] S. Alighanbari et al., Nat. Phys. 19, 1263 (2023)

MO 6.6 Tue 12:15 HS 3044

**Photodissociation spectrum of a single trapped CaOH<sup>+</sup>** — ●ZHENLIN WU, STEFAN WALSER, BRANDON FUREY, MARIANO ISAZA-MONSALVE, ELYAS MATTIVI, RENÉ NARDI, and PHILIPP SCHINDLER — Institut für Experimentalphysik, Universität Innsbruck, Innsbruck, Austria

Molecular ions can be sympathetically cooled and crystallized in atomic ion crystals confined in radio-frequency traps, which are ideal for molecular spectroscopy on the single molecule scale. Their application in quantum technologies and the exploration of fundamental physics have also been proposed and demonstrated. Most experiments investigating the internal structure of trapped molecular ions rely on dissociation-based state detection methods and quantum logic spectroscopy via co-trapped atomic qubit ions. In our setup, we aim to study triatomic CaOH<sup>+</sup> molecular ions generated in trapped Ca<sup>+</sup> ion experiments in the presence of water vapor. As the first step towards quantum logic spectroscopy of a single trapped polyatomic ion, we investigate the single-photon and two-photon photodissociation process of CaOH<sup>+</sup> which excites the molecule to its unbound first electronic excited state. We report the photodissociation cross section spectrum of CaOH<sup>+</sup> obtained from measurement of a single CaOH<sup>+</sup> located in an ion chain. This result can be the basis of dissociation-based spectroscopy for studying the rovibrational structure of CaOH<sup>+</sup>. In addition, the reported spectrum can be useful in large-scale trapped Ca<sup>+</sup> quantum experiments for recycling Ca<sup>+</sup> ions when they form undesired CaOH<sup>+</sup> ions via background gas collisions.

MO 6.7 Tue 12:30 HS 3044

**Collisional shift and broadening of Rydberg states in thermal nitric oxide** — ●ALEXANDER TRACHTMANN<sup>1</sup>, FABIAN MUNKES<sup>1</sup>, PATRICK KASPAR<sup>1</sup>, FLORIAN ANSCHÜTZ<sup>1</sup>, PHILIPP HENGEL<sup>2</sup>, YANNICK SCHELLANDER<sup>3</sup>, PATRICK SCHALBERGER<sup>3</sup>, NOR-

BERT FRUEHAUF<sup>3</sup>, JENS ANDERS<sup>2</sup>, ROBERT LÖW<sup>1</sup>, TILMAN PFAU<sup>1</sup>, and HARALD KÜBLER<sup>1</sup> — <sup>15</sup>. Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart — <sup>2</sup>Institut für Intelligente Sensorik und Theoretische Elektrotechnik, Universität Stuttgart, Pfaffenwaldring 47, 70569 Stuttgart — <sup>3</sup>Institut für Großflächige Mikroelektronik, Universität Stuttgart, Allmandring 3b, 70569 Stuttgart

We report on the collisional shift and line broadening of Rydberg states in nitric oxide (NO) with increasing density of a background gas at room temperature [1]. As a background gas we either use NO itself or nitrogen (N<sub>2</sub>). The precision spectroscopy is achieved by a sub-Doppler three-photon excitation scheme with a subsequent readout of the Rydberg states realized by the amplification of a current generated by free charges due to collisions. [1] arXiv:2310.18256

MO 6.8 Tue 12:45 HS 3044

**Highly-resolved Stark effect measurements of Rydberg states in thermal nitric oxide** — ●FABIAN MUNKES<sup>1</sup>, ALEXANDER TRACHTMANN<sup>1</sup>, MATTHEW RAYMENT<sup>2</sup>, FLORIAN ANSCHÜTZ<sup>1</sup>, ETTORE EDER<sup>1</sup>, YANNICK SCHELLANDER<sup>3</sup>, PHILIPP HENGEL<sup>4</sup>, PATRICK SCHALBERGER<sup>3</sup>, NORBERT FRUEHAUF<sup>3</sup>, JENS ANDERS<sup>4</sup>, ROBERT LÖW<sup>1</sup>, TILMAN PFAU<sup>1</sup>, STEPHEN HOGAN<sup>2</sup>, and HARALD KÜBLER<sup>1</sup> — <sup>15</sup>. Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart — <sup>2</sup>Department of Physics and Astronomy, University College London, Gower Street, London WC1E 6BT, UK — <sup>3</sup>Institut für Großflächige Mikroelektronik, Universität Stuttgart, Allmandring 3b, 70569 Stuttgart — <sup>4</sup>Institut für Intelligente Sensorik und Theoretische Elektrotechnik, Universität Stuttgart, Pfaffenwaldring 47, 70569 Stuttgart

We demonstrate Stark effect measurements at room temperature of high-lying Rydberg states in nitric oxide. These states are generated using a three-photon continuous-wave excitation scheme. The readout is based on the detection of charged particles created by collisional ionization of Rydberg molecules. A theoretical discussion of the obtained experimental results is given.

## MO 7: Poster: Spectroscopy

Time: Tuesday 17:00–19:00

Location: Tent C

MO 7.1 Tue 17:00 Tent C

**In Situ Hyperpolarized Benchtop NMR for Biomolecular Analysis at Natural Isotopic Abundance** — ●JINGYAN XU<sup>1,2,3</sup>, RAPHAEL KIRCHER<sup>1,2,3</sup>, and DANILA BARSKIY<sup>1,2,3</sup> — <sup>1</sup>Johannes Gutenberg University, Mainz, Germany — <sup>2</sup>Helmholtz-Institut, Mainz, Germany — <sup>3</sup>Helmholtzzentrum für Schwerionenforschung, Darmstadt, Germany

Nuclear Magnetic Resonance (NMR) is a key noninvasive tool in fields ranging from research and industry to medicine. Benchtop NMR spectrometers have recently emerged as practical alternatives to traditional high-field NMR systems, especially for on-the-spot analysis and process monitoring. A notable challenge with these systems is the detection of low-abundance heteronuclei like <sup>13</sup>C or <sup>15</sup>N, hindered by low spin polarization resulting from weak interactions with the magnetic field. Our study introduces a novel hyperpolarization technique to overcome this limitation by integrating Signal Amplification by Reversible Exchange (SABRE) with a Spin-Lock Induced Crossing (SLIC) pulse sequence. Applied to various molecules, this method achieves up to 12% polarization for <sup>15</sup>N and 0.4% for <sup>13</sup>C, without needing sample transfer. Furthermore, our method allows for continuous hyperpolarization, paving the way for advanced applications in rapid 2D spectroscopy and relaxometry. This development offers a cost-effective, efficient means of detecting diluted chemicals using benchtop NMR, signaling a significant leap forward in diverse industrial and research applications.

MO 7.2 Tue 17:00 Tent C

**An experimental setup to study the influence of hydration on small charged molecular systems by rotationally resolved vibrational spectroscopy** — ●ERIC ENDRES<sup>1</sup>, CHRISTIAN SPRENGER<sup>1</sup>, FRANZISKA DAHLMANN<sup>2</sup>, and ROLAND WESTER<sup>1</sup> — <sup>1</sup>Institut f. Ionenphysik und Angewandte Physik, Universität Innsbruck, Technikerstraße 25/3, A-6020 Innsbruck, Austria — <sup>2</sup>KTH Royal Institute of Technology, Stockholm, Sweden

Hydration with individual water molecules significantly influences the

structure and, consequently, the function of biomolecules. This contribution introduces an experimental setup designed to examine the influence of hydration on the structure of small biomolecules using rotationally resolved pre-dissociation spectroscopy.

Ions are generated using a custom-built Nano-ESI system housed within a controlled environment, regulating e.g. humidity and temperature, enabling the control of the hydration level. A double skimmer setup gently transfer the ions into vacuum, avoiding breaking apart the loosely bound water molecules. Through an octupole guide and a quadrupole guide the water clusters are let into a cryogenic 16-pole wire ion trap. Trap temperatures below 3K can be achieved, enabled binding of up to four helium atoms on protonated glycine ions. The confined ions are irradiated by a narrow-band laser system with an output linewidth in the IR below 0.1 cm<sup>-1</sup>

The low temperatures in combination with the small linewidth of the laser system potentially lead to rotationally resolved vibrational spectroscopy. Here the current status will be reported.

MO 7.3 Tue 17:00 Tent C

**Towards photodetachment spectroscopic studies of deprotonated naphthalene.** — ●MICHAEL HAUCK, SRUTHI PURUSHU MELATH, CHRISTINE LOCHMANN, ROBERT WILD, KATRIN ERATH-DULITZ, and ROLAND WESTER — Institut für Ionenphysik und Angewandte Physik, Universität Innsbruck, Technikerstraße 25/3, 6020 Innsbruck, Austria

Polycyclic aromatic hydrocarbon (PAH) molecules are important in the study of interstellar chemistry. The discovery of benzonitrile in the molecular cloud TMC-1 hints at the existence of larger aromatic molecules which might explain still unassigned infrared bands [1,2]. We are interested in the spectroscopic study of deprotonated naphthalene (C<sub>10</sub>H<sub>7</sub><sup>-</sup>). Photoelectron spectroscopic studies of this molecule were performed, which found the electron affinities of two possible isomers [3].

Here we present a 2D tomography photodetachment scan of depro-

tonated naphthalene in our cryogenic 16 pole trap, which we measure in order to extract the value of the absolute photodetachment cross section. We further plan to examine the behaviour of the cross section near threshold and measure the electron affinity of the molecule.

- [1] J. Gao, G. Berden and J. Oomens, *Ap. J.* 787, 170 (2014)
- [2] B. A. McGuire et al., *Science* 359, 202 (2018)
- [3] M. L. Weichman, J. B. Kim and D. M. Neumark, *J. Phys. Chem. A* 119, 6140 (2015)

MO 7.4 Tue 17:00 Tent C

**Combination of action-based interferometric measurements with depletion spectroscopy** — ●LEONIE WERNER, ULRICH BANGERT, YILIN LI, ARNE MORLOK, LUKAS BRUDER, and FRANK STIENKEMEIER — University of Freiburg, Institute of Physics, Hermann-Herder-Straße 3, 79104 Freiburg im Breisgau, Germany

Isolation spectroscopy in ultracold helium nanodroplets enables the investigation of single and multiple embedded organic molecules in a cold, weakly perturbing environment [1]. In our lab, we combine this method with interferometric techniques, such as wave packet interferometry (WPI) and two-dimensional electronic spectroscopy (2DES) [2, 3]. This allows us to achieve high temporal and energy resolution at the same time. Standard detection techniques are laser-induced fluorescence or photoionization. Yet, these methods have problems to capture non-radiative photochemical processes or pose a challenge in overcoming the high ionization potentials of organic molecules. To solve these issues, we are exploring beam depletion detection methods [1]. First tests will be presented.

- [1] J. P. Toennies and A. F. Vilesov, *Angew. Chem. Int. Ed.* 43, 2622 (2004).
- [2] L. Bruder et al., *Nat. Commun.* 9, 4823 (2018).
- [3] L. Bruder et al., *J. Phys. B* 52, 183501 (2019).

MO 7.5 Tue 17:00 Tent C

**Investigating the Photodissociation Dynamics of CH<sub>2</sub>Br with VMI** — ●LILITH WOHLFART, CHRISTIAN MATTHAEI, and INGO FISCHER — Julius-Maximilians-Universität, 97074 Würzburg, Germany

Bromomethyl belongs to the class of organic halogen radicals. Therefore, it can potentially influence the atmosphere by reacting with the ozone layer and causing its depletion similar to HCFCs. The photoionization of bromomethyl was already investigated by several groups, including Steinbauer and coworkers. They determined the ionization energy and structure with VUV synchrotron radiation and investigated the dissociative photoionization. To obtain further insights into the dissociation of bromomethyl, we analyzed the fragments of the radical using velocity map imaging (VMI).

CH<sub>2</sub>Br-NO<sub>2</sub> was used as a precursor for the halogenated methyl radical, because the weaker C-NO<sub>2</sub> bond can be cleaved through pyrolysis. Subsequently, laser light in the UV region was deployed to dissociate the formed CH<sub>2</sub>Br radical. The major dissociation pathway gave the methylene and bromine fragments which were detected with SPI and REMPI respectively. With velocity map ion imaging, the translational kinetic energy distribution of the photofragments was determined. The recorded images of the bromine and methylene photofragments showed an anisotropic distribution, implying a direct dissociation.

MO 7.6 Tue 17:00 Tent C

**Photodissociation dynamics of the CHCl<sub>2</sub> radical** — ●JONAS FACKELMAYER and INGO FISCHER — Julius-Maximilians-Universität Würzburg, 97074 Würzburg, Germany

Monitoring the atmospheric abundances of the ozone-depleting CFCs has revealed a global increase in emission of these banned substances.<sup>[1]</sup> Meanwhile the dimensions of the ozone-hole have been reported to be at a all time high in 2023.<sup>[2]</sup> This makes the photodissociation of these compounds of great importance since it often results in the release of highly reactive halogen radicals. While the dissociation dynamics of molecular halocarbons have been studied in detail in the past, less is known about their open shell counterparts.

The photofragmentation of the open shell CHCl<sub>2</sub>, generated by pyrolysis from the bromide and iodide precursors CHCl<sub>2</sub>Br/CHCl<sub>2</sub>I, was investigated in a free jet utilising time-of-flight mass-spectrometry and velocity map imaging. Photodissociation was achieved by a pulsed dye laser in the range of 230 - 250 nm mainly producing CHCl and Cl fragments, while ionisation was provided by either a second dye laser (REMPI) or a frequency multiplied solid state laser at 118 nm (SPI). Insights into the involved dissociation mechanisms are discussed.

- [1] L. M. Western et al., *Nat. Geosci.* 2023, 16, 309\*313.

- [2] European Space Agency, *Ozone hole goes large again*, 2023.

MO 7.7 Tue 17:00 Tent C

**Time-resolved photoelectron and photoion spectroscopy of phenanthridine - an experimental and computational study** — ●KATHARINA THEIL<sup>1</sup>, JONAS FACKELMAYER<sup>1</sup>, LIONEL POISSON<sup>2</sup>, LOU BARREAU<sup>2</sup>, and INGO FISCHER<sup>1</sup> — <sup>1</sup>Institute of Physical and Theoretical Chemistry, University of Würzburg, Am Hubland, D-97074 Würzburg — <sup>2</sup>Institut des Sciences Moléculaires d'Orsay (ISMO) UMR 8214, Rue André Rivière, Bâtiment 520, Université Paris-Saclay, F-91405 Orsay Cedex, France

Understanding the fundamental photophysical processes in molecules is essential for deciphering their photochemistry, given that molecules rarely undergo reactions directly from their initially excited electronic states. Recently, the focus has shifted towards investigating polycyclic aromatic nitrogen heterocycles (PANHs) within the extensive studies of polycyclic aromatic hydrocarbons (PAHs) as potential carriers of 'unidentified infrared bands' and diffuse interstellar bands.<sup>[1]</sup> Here we investigate the ultrafast excited-state dynamics of phenanthridine, a prototypical PANH, employing femtosecond time-resolved pump-probe spectroscopy conducted in the gas phase. The real-time monitoring of these dynamics is facilitated through time-resolved photoionization and photoelectron imaging. The experimental results are accompanied by theoretical calculations.

- [1] D. McNaughton et al., *Phys. Chem. Chem. Phys.*, 2007, 9, 591-595.

MO 7.8 Tue 17:00 Tent C

**Ultrafast UV-Vis spectroscopy on a series of novel Fe(III) photosensitizers with linked organic chromophores** — ●MIGUEL ANDRE ARGÜELLO CORDERO<sup>1</sup>, LENNART SCHMITZ<sup>2</sup>, MATTHIAS BAUER<sup>2</sup>, and STEFAN LOCHBRUNNER<sup>1</sup> — <sup>1</sup>University of Rostock, Germany — <sup>2</sup>University of Paderborn, Germany

In recent years, the exploration of photocatalytic methodologies for solar fuel production has garnered attention due to their promise as a sustainable energy source. Central to these approaches are molecular photosensitizers (PS) with a metal component for efficient light absorption. However, the reliance on precious metals in conventional PS has prompted a fervent exploration into alternatives employing cost-efficient metals, like iron. Fe(III)-based PS exhibit a distinctive bichromophoric nature, stemming from the existence of ligand-to-metal charge transfer transitions (LMCT). This unique attribute facilitates the capture of the entire visible spectrum, resulting in the population of enduring LMCT states. The strategic modification of the ligand structure further allows for the modulation of energy levels and excited state lifetimes. In this study, we introduce a series of novel Fe(III) PS featuring organic chromophores attached to their ligand backbones. By extending the chromophores, we uncover intriguing ultrafast electronic dynamics following optical excitation. Their investigation is carried out through fs-UV-Vis transient absorption spectroscopy. This work presents our findings on pump-probe spectroscopy applied to the Fe(III) PS, providing a comprehensive discussion on the observed results in the context of electronic relaxation pathways.

MO 7.9 Tue 17:00 Tent C

**Time-Resolved Spectroscopic Studies on the Net Heterolysis of Homopolar Selenium-Carbon Bonds** — ●DANIEL JAN GREINDA, ANNA FRANZISKA TIEFEL, CARINA ALLACHER, ELIAS HARRER, ROGER JAN KUTTA, JULIA REHBEIN, ALEXANDER BREDER, and PATRICK NUERNBERGER — Universität Regensburg, 93040 Regensburg

When thinking about chemical bonds, one considers polarity as the major factor determining if a bond cleavage occurs homolytically or heterolytically, as bonds with a negligible dipole moment exclusively undergo homolysis, whereas heterolysis requires a sufficiently high dipole moment or some kind of external bond activation [1]. We demonstrate that by combining photochemistry [2] with the radical chemistry of organoselenium compounds [3] and the properties of the solvent hexafluoroisopropanol [3, 4], net heterolysis of the homopolar selenium-carbon bond can be achieved and utilized in a subsequent S<sub>N</sub>1-type substitution. The mechanism of this reaction is disclosed using transient absorption spectroscopy with streak-camera detection [5] and other advanced spectroscopic techniques, as well as theoretical and synthetic investigations.

- [1] H. Brueckner, *Reaktionsmechanismen* Springer Spektrum (2008).
- [2] B. Koenig et al., *Eur. J. Org. Chem.*, 15, 1979-1981 (2017).
- [3] I. Colomer et al., *Nat. Rev. Chem.*, 1, 0088 (2017).
- [4] S. Park et al., *Angew. Chem. Int. Ed.*, 61, e202208611 (2022).

[5] R. J. Kutta *et al.*, *Appl. Phys. B*, **111**, 203-216 (2013).

MO 7.10 Tue 17:00 Tent C

**Ultrafast Time-Resolved NIR-Spectroscopy of Metal Complexes** — ●NINA BRAUER<sup>1</sup>, MIGUEL ANDRE ARGÜELLO CORDERO<sup>1</sup>, SAMIRA DABELSTEIN<sup>1</sup>, JAKOB STEUBE<sup>2</sup>, LENNART SCHMITZ<sup>2</sup>, MATTHIAS BAUER<sup>2</sup>, and STEFAN LOCHBRUNNER<sup>1</sup> — <sup>1</sup>University of Rostock, Germany — <sup>2</sup>University of Paderborn, Germany

Light induced charge transfer in metal complexes is a crucial part in many photocatalytic processes and holds great potential for future applications in solar energy harvesting. However, today most of the utilized organo-metallic complexes contain a rare metal center ion, such as ruthenium or iridium. A promising, cost-efficient replacement are iron based complexes, which are the subject of current research to optimize their photoactive properties.

One of the most prominent measurement techniques is transient absorption spectroscopy, during which the sample is photoexcited and probed with a broadband femtosecond laser pulse. Up until now, time-resolved measurements of iron complexes have been limited to the spectral region of visible light. In this work, the supercontinuum from an Yttrium Aluminum Garnet (YAG) crystal is used to probe the transient absorption of Fe(III)-complexes in the near-infrared (NIR). With a probing spectrum ranging from 820 nm up to 1250 nm, the dynamics of the excited state absorption of these complexes are investigated.

MO 7.11 Tue 17:00 Tent C

**Simplified photoelectron photoion covariance spectrometer for challenging UV pump-probe experiments** — ●NICOLAS LADDA, FABIAN WESTMEIER, TONIO ROSEN, SUDHEENDRAN VASUDEVAN, HANGYEOL LEE, SIMON RANECKY, SAGNIK DAS, JAYANTA GHOSH, TILL STEHLING, HENDRIKE BRAUN, JOCHEN MIKOSCH, THOMAS BAUMERT, and ARNE SENFTLEBEN — Institut für Physik, Universität Kassel, Heinrich-Plett-Strasse 40, 34132 Kassel, Germany

Velocity Map Imaging spectroscopy is a powerful method for investigating photoionization processes. The technique can be combined with time-of-flight spectrometry to gain further insight into the underlying processes. The resulting device can be used for photoelectron photoion covariance (PEPICO) measurements. Here we want to present our simple, but geometrically optimized PEPICO spectrometer, which can measure photoelectron momenta and photoion masses with high resolution while minimizing the background signal originating from scattered UV photons. The spectrometer will be used to investigate the dynamics of chiral molecules by studying time-resolved photoelectron circular dichroism [1]. In addition to special viewports made of single-crystal calcium fluoride (CaF<sub>2</sub>) with a broadband AR coating (190 - 900 nm) to minimize reflections and scattering, baffles made of dendritic copper oxide ( $\delta$ -CuO) with high UV absorption are used to capture the residual scattered photons [2].

[1] Lux, C. *et al.* *Angew. Chem. Int. Ed.* **51**, 5001\*5005 (2012)

[2] Clarkin, O. J., Dissertation, Queen's University, (2012)

MO 7.12 Tue 17:00 Tent C

**Resonant double core hole spectroscopy of ultrafast decay dynamics in Fe complexes** — ●JULIUS SCHWARZ<sup>1</sup>, MATZ NISSEN<sup>1</sup>, ALBERTO DE FANIS<sup>2</sup>, ALJOSCHA RÖRIG<sup>2</sup>, KAROLIN BAEV<sup>5</sup>, FLORIAN TRINTER<sup>4</sup>, TIM LAARMANN<sup>1,6</sup>, NILS HUSE<sup>1</sup>, PHILIPPE WERNET<sup>3</sup>, MICHAEL MEYER<sup>2</sup>, THOMAS BAUMANN<sup>2</sup>, SIMON DOLD<sup>2</sup>, TOMMASO MAZZA<sup>2</sup>, YEVHENIY OVCHARENKO<sup>2</sup>, SERGEY USENKO<sup>2</sup>, MARKUS ILCHEN<sup>1,2</sup>, ANDREAS PRZYSTAWIK<sup>1,6</sup>, HAMPUS WIKMARK<sup>3</sup>, and MICHAEL MARTINS<sup>1</sup> — <sup>1</sup>Universität Hamburg, Germany — <sup>2</sup>European XFEL, Hamburg, Germany — <sup>3</sup>Uppsala University, Sweden — <sup>4</sup>Fritz-Haber-Institut, Berlin, Germany — <sup>5</sup>DESY, Hamburg, Germany — <sup>6</sup>The Hamburg Centre for Ultrafast Imaging, Hamburg, Germany

Resonant double core hole (DCH) spectroscopy allows for the observation of ultrafast dynamic processes in small 3d-metal compounds in the gas phase with enhanced sensitivity. Using the intense X-Ray pulses of the European XFEL, electron and ion spectroscopy was used to reveal the signature of iron  $2p^2$  resonant DCH excitation in iron pentacarbonyl and ferrocene. Comparing the experimental results to theoretical calculations reconstructs single core hole (SCH) and DCH photon-matter interactions in the two targets. The DCH Auger-Meitner electron signals offer insight to the electron dynamics during the core hole lifetime and their dependence on the chemical environment. The product ions show evidence for DCH processes in multiply charged iron

cations.

MO 7.13 Tue 17:00 Tent C

**Accurate molecular ab initio calculations in support of photodissociation experiments** — ●GIORGIO VISENTIN<sup>1,2</sup>, BO YING<sup>2,3</sup>, STEPHAN FRITZSCHE<sup>1,2,3</sup>, and GERHARD PAULUS<sup>2,3</sup> — <sup>1</sup>Helmholtz-Institut Jena, Fröbelstieg 3, 07743 Jena, Germany — <sup>2</sup>GSI Helmholtzzentrum für Schwerionenforschung, 64291 Darmstadt, Germany — <sup>3</sup>Friedrich Schiller University, Max-Wien-Platz 1, 07743, Jena, Germany

Novel experimental techniques based on pump-probe femtosecond laser pulses have paved the way to the investigation of ultrafast molecular processes, such as photodissociation. In this process, the molecule is first ionized; then, the collision with the ejected electron induces two competing mechanisms, i.e., dissociation by excitation to a dissociative electronic excited state or dissociation by ionization to a dissociative charge state. Evaluation of these mechanisms is a crucial step in the experimental understanding of the molecular photodissociation dynamics. In this framework, accurate ab initio calculations of the potential energy curves (PECs) of the molecular ions provide a valuable tool in support of the experiment. In this abstract, an accurate relativistic ab initio molecular approach is proposed to model the PECs of Ar<sub>2</sub><sup>+</sup> in the electronic ground and lowest-lying excited states. This approach yields results in reasonable agreement with the available literature data and supports the ion-beam experiments investigating the dissociation pathways of Ar<sub>2</sub><sup>+</sup> molecular ion. Furthermore, the success of the aforementioned theoretical approach prospects the investigation on the photodissociation of heavier diatomics.

MO 7.14 Tue 17:00 Tent C

**Decoherence in molecular systems with structured spectral densities studied with Gaussian wavepacket propagation** — ●SREEJA LOHO CHOUDHURY<sup>1</sup>, RAINER HEGGER<sup>1</sup>, ROCCO MARTINAZZO<sup>2,3</sup>, and IRENE BURGHARDT<sup>1</sup> — <sup>1</sup>Institute of Physical and Theoretical Chemistry, Goethe University Frankfurt, Germany — <sup>2</sup>Department of Chemistry, Università degli Studi di Milano, Italy — <sup>3</sup>Instituto di Scienze e Tecnologie Molecolari, CNR, Milano, Italy

We investigate the time scale of decoherence in complex molecular systems following laser excitation [1]. Vibronic coupling Hamiltonians in conjunction with realistic, structured spectral densities are employed in order to track decoherence on a typical time scale of femtoseconds. Tensor network methods, notably the Gaussian-based Multi-Configuration Time-Dependent Hartree approach [2], are used to obtain accurate decoherence estimates obtained from the time-evolving purity. We focus on a donor-acceptor system that has recently been studied [1], comprising tens of vibrational degrees of freedom. This system is subject to a coherent excitation energy transfer (EET) process and exhibits irreversible decay features despite the finite dimensionality. Numerical decoherence decay is compared with analytical estimates for pure dephasing in spin-boson systems [3-4]. [1] M. Asido *et al.*, *Phys. Chem. Chem. Phys.* **24**, 1795 (2022). [2] P. Eisenbrandt, M. Ruckebauer and I. Burghardt, *J. Chem. Phys.* **149**, 174102 (2018). [3] O. Prezhdo and P. Rossky, *Phys. Rev. Lett.* **81**, 5294 (1998). [4] M. A. Schlosshauer, *Decoherence and the Quantum-To-Classical Transition*, Springer (2007).

MO 7.15 Tue 17:00 Tent C

**Photoelectron spectroscopy study of anthracene anions in gas phase** — ●KEVIN SCHWARZ, AGHIGH JALEHDOOST, and BERND V. ISSENDORFF — Institute of Physics, University of Freiburg, Hermann-Herder-Str. 3, 79104 Freiburg, Germany

Organic semiconductors like anthracene (C<sub>14</sub>H<sub>10</sub>) show interesting properties and keep being of interest across science and technology. They are used, for instance, in organic solar cells. To get a better understanding of those molecules they are investigated in the gas phase by anion photoelectron spectroscopy (PES) to gain knowledge on the different involved electronic and vibrational modes of the molecules, especially on electronic relaxation processes giving information about the dynamics within the molecules. Furthermore, temperature-resolved studies are of high interest due to changes in the vibrational modes accessible to the clusters, so to address those measurements and also to increase the resolution of the spectra in general a radio frequency ion trap will be added into the setup.

MO 7.16 Tue 17:00 Tent C

**Phase-sensitive detection of photons** — LUCAS LUDWIG, ●SANCHAYETA JANA, SIMON DURST, and MARKUS LIPPITZ — Chair

for experimental physics III, Universität Bayreuth, Bayreuth, Germany

In quantum optics, the measured signal is often photon detection events. Modulation and phase-sensitive detection of photons is essential for many applications. Even though phase-sensitive or lock-in detection is a compelling technique invented over 90 years ago, all commercial lock-in detectors use an analog voltage measured by a detector (e.g., a photodiode) as the input signal. Thus, one cannot use a commercial lock-in detector with photon counters for phase-sensitive detection of photons. For this reason, we have developed a method for lock-in detection of photons, which we use for measuring 2D spectra of single molecules.

We employ AOMs to phase-modulate four optical pulses for the fluorescence-detected two-dimensional electronic spectroscopy (F-2DES) experiment. A reference diode detects the interference signal, and the output goes to an FPGA, where three phase lock loops (PLL) lock the phase difference between the pulses and send the trigger pulse to a time tagger. A single photon counting detector detects the fluorescence signal from the molecules after excitation with the four phase-modulated pulses. These photons are also registered by the time tagger, thus enabling phase-sensitive detection of photons.

This work will discuss the construction of PLLs in FPGA and their characteristics.

MO 7.17 Tue 17:00 Tent C

**Cogwheel phase cycling in action-based two-dimensional spectroscopy** — ●STEFAN MÜLLER, AJAY JAYACHANDRAN, and TOBIAS BRIKNER — Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg

Coherent two-dimensional (2D) spectroscopies, which detect an action-based signal instead of a coherent signal, have become increasingly popular in recent years. These signals include photoions, internal or external photocurrents, and fluorescence. To extract the coherent information from the incoherent signal, a procedure such as phase cycling can be employed, which is usually carried out in a “nested” fashion, i.e., each pulse phase is incremented sequentially. Here we adapt a procedure from nuclear magnetic resonance spectroscopy, “cogwheel phase

cycling,” in which all pulse phases are varied simultaneously in increments given by so-called winding numbers [1]. We show how to perform a numerical search for these winding numbers. Using a pulse-shaper-assisted setup for fluorescence-detected 2D spectroscopy, we demonstrate that fourth-order and higher-order signals can be acquired with fewer cogwheel phase-cycling steps compared to nested phase cycling while maintaining the same signal selectivity [2]. We predict considerable time savings for various pulse-shaper-based multidimensional spectroscopies.

- [1] M. H. Levitt et al., *J. Magn. Reson.* **155**, 300–306 (2002).  
[2] A. Jayachandran et al., *J. Phys. Chem. Lett.* **13**, 11710 (2022).

MO 7.18 Tue 17:00 Tent C

**Cross-peak analysis of multiquantum signals with polarization-controlled higher-order transient absorption spectroscopy** — ●KATJA MAYERSHOFER<sup>1</sup>, SIMON BÜTTNER<sup>1</sup>, JULIAN LÜTTIG<sup>2</sup>, PETER A. ROSE<sup>3</sup>, JACOB J. KRICH<sup>3,4</sup>, and TOBIAS BRIKNER<sup>1</sup> — <sup>1</sup>Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg — <sup>2</sup>Department of Physics, University of Michigan, Ann Arbor, MI, USA — <sup>3</sup>Department of Physics, University of Ottawa, Ottawa, Ontario, Canada — <sup>4</sup>Nexus for Quantum Technologies, University of Ottawa, Ottawa, Ontario, Canada

A well-established application of two-dimensional (2D) spectroscopy is the characterization of energy transfer processes via analyzing of 2D cross-peaks. In transient absorption (TA) measurements, by contrast, signals from 2D off-diagonal and on-diagonal contributions overlap on the detection axis. In a recent paper, Zanni’s group [1] presented a new polarization scheme that suppresses diagonal peaks and makes it possible to investigate cross-peak features with TA spectroscopy. We adapted and applied this polarization scheme to the new method of higher-order TA spectroscopy [2] that separates signals of different orders and thus isolates signals stemming from exciton–exciton interactions (EEI). Through the combination of higher-order TA spectroscopy and polarization control we aim to analyze cross-peak features in fifth-order signals originating from EEI in a squaraine heterodimer.

- [1] K. M. Farrell et al., *PNAS* **2022**, *119*, e2117398118.  
[2] P. Malý et al., *Nature* **2023**, *616*, 280.

## MO 8: Poster: Collisions

Time: Tuesday 17:00–19:00

Location: Tent C

MO 8.1 Tue 17:00 Tent C

**Excitation and dissociation of astrophysically relevant molecules by ion impacts** — ●MASATO NAKAMURA — College of Science and Technology, Nihon University, Funabashi, Japan

Collision-induced dissociation of molecules by ion impacts plays an important role in the chemical evolution in molecular clouds. To reveal the mechanism of such processes, the energy transfer and fragmentation of molecules by ion impacts at hyperthermal energies are theoretically studied. The classical trajectory (CT) calculation and the spectator model are applied to estimate the energy transfer from translational to internal degrees of freedom. The model predicts the energy-transfer from the translational to the internal degrees of freedom with less efficiency. The threshold energy for CID of CO molecule by ion impacts is calculated for various projectiles. When the projectile is much lighter than the target, it is found that the spectator model works well. The probability of CID depends strongly on the orientation angle at the moment of the contact. Calculation and analysis are extended to other astrophysically relevant molecules.

MO 8.2 Tue 17:00 Tent C

**Intermolecular Coulombic Decay from organic dimers** — ●DEEPTHY MARIA MOOTHERIL<sup>1</sup>, XUEGUANG REN<sup>2</sup>, THOMAS PFEIFER<sup>1</sup>, and ALEXANDER DORN<sup>1</sup> — <sup>1</sup>Max-Planck-Institute of Nuclear Physics, Heidelberg, 69115, Germany — <sup>2</sup>School of Physics, Xi’an Jiaotong University, Xi’an, 710049, China

Inter-atomic/intermolecular Coulombic decay (ICD) is an important electronic relaxation mechanism after inner-valence ionization of atoms or molecules with weakly bound neighbours. Here we study ICD in organic heterocycle dimers like thiophene dimers and pyridine-water complexes induced by electron collisions (109 eV) using the ( $e, e+2ion$ ) coincidence technique [1]. Collisions with electrons causes ionization of

the inner valence orbital. It is observed that the energy released after relaxation to the inner valence vacancy is transferred to the neighbouring molecule, mainly via ICD, ionizes the outer valence orbital of the neighbor and thus inducing Coulomb explosion of the dimer. Comparison of projectile energy loss spectra with theoretical single ionization spectra shows the ICD mainly proceeds from the C  $2s^{-1}$  inner valence vacancy in thiophene dimers and from the O  $2s^{-1}$  and the N  $2s^{-1}$  inner-valence vacancies in pyridine-water clusters [2].

References:

- [1] X. Ren et al 2018 *Nature Physics* **14**(10) 1062–1066  
[2] A. D. Skitnevskaya et al 2023 *J. Phys. Chem. Lett.* **14**(6) 1418–1426

MO 8.3 Tue 17:00 Tent C

**Dynamics of methane  $CH_4$  activation by tantalum cations  $Ta^+$  in gas phase** — MARCEL META<sup>1</sup>, MAXIMILIAN HUBER<sup>1</sup>, ●MAURICE BIRK<sup>1</sup>, MARTIN WEDELE<sup>1</sup>, MILAN ONČÁK<sup>2</sup>, and JENNIFER MEYER<sup>1</sup> — <sup>1</sup>RPTU Kaiserslautern-Landau, Fachbereich Chemie und Landesforschungszentrum OPTIMAS, Kaiserslautern, Germany — <sup>2</sup>Universität Innsbruck, Institut für Ionenphysik und Angewandte Physik, Innsbruck, Austria

To understand reactions on an atomic-level, we make use of well-defined model systems in the gas phase. We are interested in the rearrangement of atoms during a reaction, i.e. the atomistic dynamics which we investigate by measuring energy and angle differential cross sections [1,2].

The reaction  $Ta^+ + CH_4 \rightarrow TaCH_2^+ + H_2$  in its quintet ground state is endothermic and spin forbidden. However the reaction is still observed at room temperature. An efficient crossing from the quintet surface over to the triplet surface leads to the exothermic formation of  $TaCH_2^+$  [3-6]. The title reaction was investigated by measuring experimental energy and angle differential cross sections via crossed beam velocity map imaging supported by quantum chemical calculations.

[1] J. Meyer, R. Wester, *Annu. Rev. Phys. Chem.* 2017, 68, 333; [2] M. Meta et. al., *J. Phys. Chem. Lett.* 2023, 14, 24, 5524; [3] J. F. Eckhard et. al., *J. Phys. Chem.* 2021, 125, 5289; [4] J. M. Bakker et. al., *J. Mol. Spectrosc.* 2021, 378, 111472; [5] L. G. Parke et. al., *J. Phys. Chem.* 2007, 111, 17773; [6] E. Sicilia et. al., *Phys. Chem. Chem. Phys.* 2017, 19, 16178

MO 8.4 Tue 17:00 Tent C

**Dynamics of the oxygen atom transfer reaction between Ta<sup>+</sup>/Nb<sup>+</sup> and carbon dioxide** — ●MARCEL META<sup>1</sup>, MAXIMILIAN E. HUBER<sup>1</sup>, MAURICE BIRK<sup>1</sup>, TIM MICHAELSEN<sup>2</sup>, ATILAY AYASLI<sup>2</sup>, MILAN ONČÁK<sup>2</sup>, ROLAND WESTER<sup>2</sup>, and JENNIFER MEYER<sup>1</sup> — <sup>1</sup>Fachbereich Chemie und Forschungszentrum OPTIMAS, RPTU Kaiserslautern-Landau, Kaiserslautern, Germany — <sup>2</sup>Institut für Ionophysik und Angewandte Physik, Universität Innsbruck, Innsbruck, Austria

The four atom reaction M<sup>+</sup> + CO<sub>2</sub> presents a benchmark system for CO<sub>2</sub> activation in the gas phase and has been investigated throughout the last decades with different methods and along the periodic table. Recent crossed beam experiments on the reaction dynamics of the oxygen atom transfer (OAT) reaction between Ta<sup>+</sup> + CO<sub>2</sub> → TaO<sup>+</sup> + CO showed dominantly indirect dynamics despite the thermal rates being close to collision rate and the reaction being highly exothermic.[1] Here, we compare the dynamics of the OAT reaction for the tantalum cation Ta<sup>+</sup> and its lighter homologue niobium Nb<sup>+</sup> as a means to alter spin-orbit coupling. The main focus is to gain insight into the nature of the bottle-neck for Ta<sup>+</sup> + CO<sub>2</sub>: If it is a submerged transition state or a crossing point between the quintet and triplet surfaces. Ultimately, a multi-method approach of experiment and theoretical modelling is used to better assign a nature to the bottleneck.

[1] M. Meta, M. E. Huber, T. Michaelsen, A. Ayasli, M. Ončák, R. Wester, J. Meyer, *J. Phys. Chem. Lett.* 24, 5524 (2023)

MO 8.5 Tue 17:00 Tent C

**Setup for improved resolution in ion-molecule crossed beam**

**imaging** — ●JERIN JUDY, DASARATH SWARAJ, TIM MICHAELSEN, FABIO ZAPPA, ROBERT WILD, and ROLAND WESTER — Institut für Ionophysik und Angewandte Physik, Universität Innsbruck, Innsbruck, Austria.

Crossed-beam experiments have proven to be a powerful tool for investigating reaction dynamics in the gas-phase [1]. Our group specializes in the study of ion-molecule reactions in combination with a velocity map imaging (VMI) spectrometer to record energy and angle dependent differential cross-sections [2]. Currently we focus on reactions involving laser ionized hydrogen molecules with argon using a newly commissioned crossed-beam experiment, where our aim is to resolve the different vibrational levels in the molecular products. We will present preliminary results on this system and describe the new experimental setup.

[1] N. Balucani *et al.*, *Int.Rev. Phys. Chem.* 25, 109 (2006).

[2] R. Wester, *Phys. Chem. Chem. Phys.* 16, 396 (2014).

MO 8.6 Tue 17:00 Tent C

**Auger spectrum of the ultra-fast dissociating 2p<sub>3/2</sub><sup>-1</sup>σ\* resonance in HCl in the semi-classical one-center approximation** — ●MATEJA HRAST<sup>1,2</sup> and MATJAZ ŽITNIK<sup>1,3</sup> — <sup>1</sup>Jozef Stefan Institute, Ljubljana, Slovenia — <sup>2</sup>Institute of Science and Technology Austria (ISTA), Klosterneuburg, Austria — <sup>3</sup>Faculty of Mathematics and Physics, University of Ljubljana, Slovenia

We present an ab-initio theoretical L-VV resonant Auger spectrum of the ultrafast dissociating 2p<sub>3/2</sub><sup>-1</sup>σ\* resonance in HCl. The decay rates are derived in one-center approximation and a semi-classical description of dissociation is considered. The calculated profiles of Auger spectral lines resemble those of atomic Auger decay but with the characteristic tails extending towards lower electron kinetic energies. The calculated line asymmetries reflect dissociation dynamics along the potential energy curve of the initial state and its relative position with respect to the potential energy curves of the corresponding final states. For some transitions, the line shape is also strongly affected by the variation of Auger decay rate with the internuclear distance.

## MO 9: Attosecond Physics II / Interaction with VUV and X-ray light (joint session A/MO)

Time: Wednesday 11:00–13:00

Location: HS 1010

### Invited Talk

MO 9.1 Wed 11:00 HS 1010

**Attosecond photoionization dynamics in CO<sub>2</sub> using coincidence spectroscopy** — ●IOANNIS MAKOS<sup>1</sup>, DAVID BUSTO<sup>1,2</sup>, DOMINIK ERTEL<sup>1</sup>, JAKUB BENDA<sup>3</sup>, BARBARA MERZUK<sup>1</sup>, FABIO FRASSETTO<sup>4</sup>, LUCA POLETTTO<sup>4</sup>, CLAUS DIETER SCHRÖTER<sup>5</sup>, THOMAS PFEIFER<sup>5</sup>, ZDENĚK MAŠÍN<sup>3</sup>, SERGUEI PATCHKOVSKII<sup>6</sup>, and GIUSEPPE SANSONE<sup>1</sup> — <sup>1</sup>Albert-Ludwigs-Universität Freiburg, Germany — <sup>2</sup>Lund University, Sweden — <sup>3</sup>Charles University, Prague, Czech Republic — <sup>4</sup>IFN-CNR, Padova, Italy — <sup>5</sup>MPIK, Heidelberg, Germany — <sup>6</sup>MBI, Berlin, Germany

Attosecond photoelectron interferometry is used to investigate molecular dynamics upon photoionization, revealing electron correlation effects and electron-nuclear motions interplay. Combining two-color interferometric methods with photoelectron-photoion coincidence spectroscopy enables angle-resolved studies in the recoil frame, providing insights into molecular potential anisotropy. In our study, we investigate carbon dioxide photoionization dynamics using attosecond coincidence spectroscopy. Absorption of an extreme ultraviolet photon, provided by an attosecond pulse train, leads to a superposition of cationic states, coupled to the photoelectron wave packet. Additional infrared photon absorption or emission forms a two-color photoelectron spectrogram. Our work presents CO<sub>2</sub> photoionization time delays, considering the impact of field-induced coupling of ionization channels. Furthermore, we show time-resolved photoelectron angular distributions in the recoil frame by measuring ejected electrons in coincidence with O<sup>+</sup> dissociation fragments.

MO 9.2 Wed 11:30 HS 1010

**Investigation of Correlated Electronic Dynamics by Nonlinear Attosecond Spectroscopy** — ●SAMUEL KELLERER<sup>1</sup>, IOANNIS MAKOS<sup>1</sup>, DOMINIK SCHOMAS<sup>1</sup>, DAVID BUSTO<sup>2</sup>, DOMINIK ERTEL<sup>1</sup>, ROBERT MOSHAMMER<sup>3</sup>, CLAUS DIETER SCHRÖTER<sup>3</sup>, THOMAS PFEIFER<sup>3</sup>, ARJUN NAYAK<sup>4</sup>, DEBOBRATA RAJAK<sup>4</sup>, NAVEED AHMED<sup>4</sup>, SOURIN MUKHOPADHYAY<sup>4</sup>, TAMÁS CSIZMADIA<sup>4</sup>, BALÁZS NAGYILLÉS<sup>4</sup>,

ZSOLT DIVÉKI<sup>4</sup>, KATALIN VARJÚ<sup>4</sup>, JÖRN ADAMCZEWSKI-MUSCH<sup>5</sup>, FABIO FRASSETTO<sup>6</sup>, LUCA POLETTTO<sup>6</sup>, PARASKEVAS TZALLAS<sup>7</sup>, DIMITRIS CHARALAMBIDIS<sup>7</sup>, and GIUSEPPE SANSONE<sup>1</sup> — <sup>1</sup>Uni Freiburg — <sup>2</sup>Uni Lund — <sup>3</sup>MPIK Heidelberg — <sup>4</sup>ELI ALPS Szeged — <sup>5</sup>GSI Darmstadt — <sup>6</sup>CNR-IFN Padova — <sup>7</sup>IESL-FORTH Hellas

The investigation of ultrafast processes like electronic dynamics in small quantum systems demands for generation and control of laser pulses with durations comparable or even shorter than the timescale of the investigated processes. Combining an attosecond source and a photoelectron/photoion coincidence spectrometer offers the possibility to investigate in detail the photoionization process, returning information on the role played by electronic correlation in multiple ionization of atoms. Despite its conceptual simplicity, the study of the two-photon double-ionization process in helium presents formidable experimental challenges, which we plan to address using the intense attosecond pulses provided by the SYLOS laser system available at ELI ALPS. We will present the attosecond beamline and the photoelectron/photoion apparatus used as an end-station for coincidence spectroscopy as well as first results.

MO 9.3 Wed 11:45 HS 1010

**Extracting relative dipole moments from a laser-driven two-electron wave packet in helium by combining attosecond streaking and transient absorption spectroscopy** — ●SHUYUAN HU, YU HE, GERGANA D. BORISOVA, MAXIMILIAN HARTMANN, PAUL BIRK, CHRISTIAN OTT, and THOMAS PFEIFER — Max-Planck-Institut für Kernphysik, 69117 Heidelberg

The electronic structure of atoms and their interaction with light is reflected in complex-valued transition-matrix elements that have a magnitude and phase. In this work, a state-resolved phase of the time-delay dependent modulation of absorption is used to determine the relative signs of transition dipole matrix elements. This measurement relies on precise absolute calibration of the time-delay information, which is achieved by combining attosecond transient absorption and attosec-

ond streaking spectroscopy to simultaneously measure the resonant photoabsorption spectra of laser-coupled doubly excited states in helium, together with the streaked photoelectron spectra. The streaking measurement reveals the absolute time delay zero and the full temporal profile of the interacting electric fields which is then used for a time-dependent few-level simulation of the relevant states. By comparing the 1-fs time-scale modulations across the  $2s2p$  ( $^1P$ ) and  $sp_{2,3+}$  ( $^1P$ ) states between the time-delay calibrated simulation and measurement, we quantify the signs of the transition dipole matrix elements for the laser-coupled autoionizing states  $2s2p-2p^2$  and  $2p^2-sp_{2,3+}$  to be opposite of each other.

MO 9.4 Wed 12:00 HS 1010

**Driving the high harmonic process using a multi-pass cell** — ●BENJAMIN STEINER<sup>1</sup>, DOMINIK ERTTEL<sup>1</sup>, DENNIS GROSCHUPF<sup>1</sup>, ANNE-LISE VIOTTI<sup>2</sup>, MARIO NIEBUHR<sup>1</sup>, BARBARA MERZUK<sup>1</sup>, DAVID BUSTO<sup>1,2</sup>, IOANNIS MAKOS<sup>1</sup>, and GIUSEPPE SANSONE<sup>1</sup> — <sup>1</sup>Institute of Physics, University of Freiburg, Freiburg, Germany — <sup>2</sup>Division of Atomic Physics, Lund University, Sweden

The investigation of electronic-correlation driven processes, such as the Auger decay in krypton [1] or single-photon double-ionisation in helium [2], requires photon energies of 100 eV or higher. Using electron-electron coincidence and attosecond pulses in the XUV spectral range obtained by high-order harmonic generation (HHG), these processes can be resolved in time in a pump-probe scheme. The first challenge is to demonstrate an attosecond source operating at high repetition rates (>50kHz) characterised by a cut-off energy well above 100 eV. For this purpose, we developed a temporal pulse compression scheme based on a gas-filled multi-pass cell for high-power throughput driven by a commercially available Yb-based laser system. The achieved pulses lead to high enough peak intensities for driving the HHG process in neon efficiently, maintaining a sufficient photon flux in the desired energy range. The generated attosecond XUV pulses will then be employed in the already existing attosecond coincidence spectrometer in Freiburg [3] for time-resolved investigations of electron dynamics occurring during the above-mentioned processes.

[1] M. Drescher et al, Nature, 419 (2002) [2] C. Ott et al, Nature, 516 (2014) [3] D. Ertel et al, Rev. Sci. Instrum. 94, 073001(2023)

MO 9.5 Wed 12:15 HS 1010

**Polarization dependence of high-order harmonic generation in the direct measurement of optical waveforms** — ●RONAK NARENDRA SHAH<sup>1</sup>, JAHANZEB MUHAMMAD<sup>1</sup>, IANINA KOSSE<sup>1</sup>, SAMUEL BENGTSOON<sup>2</sup>, RICCARDO MORI<sup>1</sup>, MARIO NIEBUHR<sup>1</sup>, FABIO FRASSETTO<sup>3</sup>, LUCA POLETTI<sup>3</sup>, and GIUSEPPE SANSONE<sup>1</sup> — <sup>1</sup>Physikalisches Institut, Albert-Ludwigs Universität Freiburg, Freiburg, 79104, Germany — <sup>2</sup>Department of Physics, Lund University, PO Box 118, SE-221 00 Lund, Sweden — <sup>3</sup>Istituto di Fotonica e Nanotecnologie, CNR, Padova, Italy

We present the polarization effects in an all-optical technique to measure the electric field of a few cycle laser pulse via high harmonic generation (HHG). In our approach, the generation of an isolated attosecond pulse (IAP) and the associated photon yield serves as an ultrashort temporal gate to characterize the electric field of a weak perturbing unknown pulse. Changing the polarization of the unknown laser pulse from parallel to orthogonal polarization with respect to

the pulse generating IAP, we report the modulation in the harmonic yield at twice the laser period. The experimental results are in good agreement with simulations based on the strong-field approximations.

MO 9.6 Wed 12:30 HS 1010

**Towards AI-enhanced online-characterization of ultrashort X-ray free-electron laser pulses** — ●THORSTEN OTTO<sup>1,2,4</sup>, KRISTINA DINGEL<sup>2</sup>, LARS FUNKE<sup>3</sup>, SARA SAVIO<sup>3</sup>, LASSE WÜLFING<sup>3</sup>, BERNHARD SICK<sup>2</sup>, WOLFRAM HELML<sup>3</sup>, and MARKUS ILCHEN<sup>4</sup> — <sup>1</sup>Deutsches Elektronen-Synchrotron DESY, Notkestraße 85, 22607 Hamburg, Germany — <sup>2</sup>Intelligent Embedded Systems, University of Kassel, Wilhelmshöher Allee 73, 34121 Kassel, Germany — <sup>3</sup>Technische Universität Dortmund, Fakultät für Physik, Maria-Göppert-Mayer-Straße, 44227 Dortmund, Germany — <sup>4</sup>Universität Hamburg, Institut für Experimentalphysik, Luruper Chaussee 149 22761 Hamburg

X-ray free-electron lasers provide ultrashort X-ray pulses with durations typically in the order of femtoseconds, but recently even entering the attosecond regime. The technological evolution of XFELs towards well-controllable light sources for precise metrology of ultrafast processes can only be achieved using new diagnostic capabilities for characterizing X-ray pulses at the attosecond frontier. The spectroscopic technique of photoelectron angular streaking has successfully proven how to non-destructively retrieve the exact time-energy structure of XFEL pulses on a single-shot basis. By using deep learning algorithms, we show how this technique can be leveraged from its proof-of-principle stage towards routine diagnostics at XFELs providing precise feedback in real time.

MO 9.7 Wed 12:45 HS 1010

**Angular Streaking at 1030 nm – measurement of gigawatt-power attosecond pulses at European XFEL** — ●LARS FUNKE<sup>1</sup>, SARA SAVIO<sup>1</sup>, LASSE WÜLFING<sup>1</sup>, NICLAS WIELAND<sup>1</sup>, KRISTINA DINGEL<sup>4</sup>, TORSTEN OTTO<sup>2</sup>, RUDA HINDRIKSSON<sup>4</sup>, LUTZ MARDER<sup>4</sup>, CHRISTOPHER PASSOW<sup>2</sup>, REBECCA BOLL<sup>3</sup>, ALBERTO DE FANIS<sup>3</sup>, SIMON DOLD<sup>3</sup>, TOMMASO MAZZA<sup>3</sup>, DIRK RAISER<sup>3</sup>, MICHAEL MEYER<sup>3</sup>, TERENCE MULLINS<sup>3</sup>, MARKUS ILCHEN<sup>5</sup>, and WOLFRAM HELML<sup>1</sup> — <sup>1</sup>Technische Universität Dortmund, Germany — <sup>2</sup>Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany — <sup>3</sup>European XFEL GmbH, Schenefeld, Germany — <sup>4</sup>Universität Kassel, Germany — <sup>5</sup>Universität Hamburg, Germany

Angular Streaking can be used as a method for characterizing ultrashort X-ray pulses by overlapping the pulse with a circularly polarized IR laser pulse in a gaseous target. Photoelectron momenta are shifted in a characteristic way for a given spectro-temporal X-ray pulse structure. Measuring the photoelectron energy spectra with multiple time-of-flight spectrometers allows the reconstruction of pulse structure.

A *Cookiebox*-type photoelectron spectrometer array was set up at the SQS instrument of European XFEL to characterize specially tuned sub-femtosecond soft X-ray FEL pulses.

In the measurement, we found intense attosecond X-ray pulses, with pulse durations on the order of 300 as and a peak power in the hundreds of gigawatts. The lower-than-planned streaking laser wavelength of 1030 nm turned out beneficial for characterizing the ultrashort pulses provided.

## MO 10: Ultracold Molecules (joint session Q/MO)

Time: Wednesday 11:00–13:00

Location: HS 1015

### Invited Talk

MO 10.1 Wed 11:00 HS 1015

**Ultracold interactions between ions and polar molecules** — ●LEON KARPA — Leibniz Universität Hannover, Institut für Quantenoptik, Welfengarten 1, 30167 Hannover, Germany

Ultracold molecules stand out as a promising candidate in a broad spectrum of advanced applications including quantum chemistry, fundamental physics, quantum simulations and information science. Studies of neutral molecular quantum gases and ultracold ion-neutral interactions are two largely complementary interdisciplinary fields that nonetheless share the vision of understanding molecular systems of ever-increasing complexity, and ultimately controlling their properties. In my talk, I will discuss recent advances and challenges in these research domains and how methods from both fields can be used to com-

bine atomic ions with quantum gases of polar molecules. The resulting complex yet precisely controllable system exhibits a hierarchy of tunable attractive and repulsive interactions of different scales, enabling a range of novel experiments and applications. This includes studies of dynamical properties of ultracold polar molecules, ion-molecule collisions in the quantum dominated regime, and the potential formation of ion-molecule many-body bound states.

MO 10.2 Wed 11:30 HS 1015

**Developing a Hybrid Tweezer Array of Rydberg Atoms and Polar Molecules** — ●KAI VOGES, DANIEL HOARE, YUCHEN ZHANG, QINSHU LYU, JONAS RODEWALD, BEN SAUER, and MICHAEL TARBUTT — Centre for Cold Matter, Imperial College London, UK

Hybrid tweezer arrays of atoms and molecules are a novel and versa-

tile platform for quantum science and technology. The combination of Rydberg atoms with their large electric dipole moment and polar molecules with their rich level structure and long state coherence times makes this approach a promising candidate for quantum simulation [1] and computing [2,3].

In this talk, I present our efforts to build a hybrid tweezer array based on ultracold Rb atoms and directly laser-coolable CaF molecules. I discuss the advantages and challenges of using such a hybrid system and present our preparation procedures for the atoms and molecules. Furthermore, I show our efforts in trapping and imaging individual atoms and molecules and present our ideas for loading both species into separate tweezer arrays.

Our approach will make it possible to construct arbitrary patterns of atoms and molecules. Through the dynamic rearrangement of tweezers and the long-range interactions mediated by Rydberg atoms, this hybrid platform will be a compelling candidate for scalable quantum computing.

[1] J. Dobrzyniecki *et al.*, PRA **108**, 052618 (2023)

[2] C. Zhang *et al.*, PRX Quantum **3**, 030340 (2022)

[3] K. Wang *et al.*, PRX Quantum **3**, 030339 (2022)

MO 10.3 Wed 11:45 HS 1015

**Quantum Dynamics of Two Composite Bosons on a One-Dimensional Lattice** — ●CAROLINE STIER, ANDREAS BUCHLEITNER, and GABRIEL DUFOUR — Physikalisches Institut der Albert-Ludwigs-Universität Freiburg

We study how the dynamics of two composite bosons on a one-dimensional lattice are affected by their constituents' quantum statistics as well as their initial state. We formulate an effective Hamiltonian assuming that the two composites – consisting either of two elementary fermions or two elementary bosons – are tightly bound objects. The contact interactions between the elementary constituents are chosen such that the resulting composite particles do not interact when they are located on the same site. However, due to the exchange of identical constituents, the composites experience an effective nearest-neighbor interaction if they are located on adjacent sites. We solve the Schrödinger equation analytically and perform numerical simulations of the dynamics from several initial configurations. In particular, we find that the composites can form a bound state whose group velocity depends strongly on the nature of their constituents.

MO 10.4 Wed 12:00 HS 1015

**Non-abelian invariants in periodically-driven quantum rotors** — ●VOLKER KARLE, AREG GHAZARYAN, and MIKHAIL LEMESHKO — Institute of Science and Technology Austria, Am Campus 1, 3400 Klosterneuburg

This presentation explores the role of topological invariants in the non-equilibrium dynamics of periodically-driven quantum rotors, inspired by experiments on closed-shell diatomic molecules driven by periodic, far-off-resonant laser pulses. This approach uncovers a complex phase space with both localized and delocalized Floquet states. We demon-

strate that the localized states are topological in nature, originating from Dirac cones protected by reflection and time-reversal symmetry. These states can be modified through laser strength adjustments, making them observable in current experiments through molecular alignment and observation of rotational level populations. Notably, in scenarios involving higher-order quantum resonances leading to multiple Floquet bands, the topological charges become non-Abelian. This results in the remarkable finding that the exchange of Dirac cones across different bands is non-commutative, enabling non-Abelian braiding, paving the way for the study of controllable multi-band topological physics in gas-phase experiments with small molecules, as well as for classifying dynamical molecular states by their topological invariants.

MO 10.5 Wed 12:15 HS 1015

**From rotational decay of diatomic molecules to quantum friction** — ●NICOLAS SCHÜLER, OMAR JESÚS FRANCA SANTIAGO, and STEFAN YOSHI BUHMANN — Institute of Physics, University of Kassel, Germany

We study the rotational motion of diatomic molecules in free space and interacting with the quantum electromagnetic field [1]. Using macroscopic quantum electrodynamics [2], we obtain the rotation-dependent decay rates of the molecule. By analyzing the behavior of the resulting rates at zero and finite temperature, we find a connection between the decelerating rotational dynamics and quantum friction.

**Invited Talk**

MO 10.6 Wed 12:30 HS 1015

**Quantum Logic Spectroscopy of the Hydrogen Molecular Ion** — DAVID HOLZAPFEL, FABIAN SCHMID, NICK SCHWEGLER, OLIVER STADLER, MARTIN STADLER, JONATHAN HOME, and ●DANIEL KIENZLER — Otto-Stern-Weg 1, 8093 Zurich, Switzerland

I will present our latest results, implementing pure quantum state preparation, coherent manipulation, and non-destructive state readout of the hydrogen molecular ion  $\text{H}_2^+$ . The hydrogen molecular ion  $\text{H}_2^+$  is the simplest stable molecule, and its structure can be calculated ab-initio to high precision. However, challenging properties such as high reactivity, low mass, and the absence of rovibrational dipole transitions have thus far strongly limited spectroscopic studies of  $\text{H}_2^+$ . We trap a single  $\text{H}_2^+$  molecule together with a single beryllium ion using a cryogenic Paul trap apparatus, achieving trapping lifetimes of 11 h and ground-state cooling of the shared axial motion [1]. With this platform we have recently implemented *Quantum Logic Spectroscopy* of  $\text{H}_2^+$ . We utilize helium buffer-gas cooling to prepare the lowest rovibrational state of ortho- $\text{H}_2^+$  (rotation  $L = 1$ , vibration  $\nu = 0$ ). We combine this with quantum-logic operations between the molecule and the beryllium ion for preparation of single hyperfine states and non-destructive readout, and demonstrate Rabi flopping on several hyperfine transitions. Our results pave the way to high-precision spectroscopy studies of  $\text{H}_2^+$  which will enable tests of theory, metrology of fundamental constants, and an optical molecular clock.

[1] N. Schwegler, D. Holzapfel, M. Stadler, A. Mitjans, I. Sergachev, J. P. Home, and D. Kienzler, Phys. Rev. Lett. **131**, 133003 (2023)

## MO 11: X-ray Spectroscopy

Time: Wednesday 11:00–12:45

Location: HS 3044

MO 11.1 Wed 11:00 HS 3044

**Photon-recoil imaging: Nonlinear X-ray physics in molecules** — ●L. GERMEROOTH<sup>1</sup>, M. AGÅKER<sup>2</sup>, T. BAUMANN<sup>3</sup>, R. BOLL<sup>3</sup>, A. DE FANIS<sup>3</sup>, S. EISEBITT<sup>4</sup>, M. GÉNÉVRIEZ<sup>5</sup>, V. KIMBERG<sup>6</sup>, H. LEE<sup>1</sup>, E. MARIN-BUJEDO<sup>5</sup>, T. MAZZA<sup>3</sup>, M. MEYER<sup>3</sup>, J. MIKOSCH<sup>1</sup>, Y. OVCHARENKO<sup>3</sup>, S. PATCHKOVSKII<sup>4</sup>, D. REISER<sup>3</sup>, J.-E. RUBENSSON<sup>2</sup>, J. SÖDERSTRÖM<sup>2</sup>, P. SCHMIDT<sup>3</sup>, B. SENFTLEBEN<sup>3</sup>, A. SENFTLEBEN<sup>1</sup>, S. USENKO<sup>3</sup>, and U. EICHMANN<sup>4</sup> — <sup>1</sup>Universität Kassel — <sup>2</sup>Uppsala University — <sup>3</sup>XFEL Hamburg — <sup>4</sup>MBI Berlin — <sup>5</sup>UCLouvain — <sup>6</sup>KTH Stockholm

Non-linear Raman spectroscopy was originally developed for narrow-band lasers. It has since become important in the spectroscopy and microscopy of technological and biological processes. Stimulated Raman scattering with optical femtosecond lasers is routinely used to excite coherent vibrational and rotational wavepackets. The advance of ultrabright FELs enabled the extension of non-linear physics to the X-ray domain. Some of us have recently established photon-recoil imaging as a background-free technique to detect stimulated X-ray Raman

scattering (SXRS) [1]. This process is similar to STIRAP, a form of state-to-state coherent control well known in the optical domain. Here we extend the pioneering experiments on Neon atoms to molecules. Near and far-off resonance SXRS in CO molecules populates efficiently an electronically excited long-lived metastable final state. Demonstration of far-off resonance SXRS in molecules opens new possibilities to study site-selective non-linear processes avoiding spontaneous decays. [1] Eichmann *et al.*, Science **369**, 1630 (2020)

MO 11.2 Wed 11:15 HS 3044

**Testing the potential energy curves of the  $\text{H}_2$  B-X system using its Condon diffraction bands** — ●ADRIAN PETER KRONE<sup>1</sup>, PHILIPP SCHMIDT<sup>2</sup>, JOHANNES VIEHMANN<sup>1</sup>, NIKLAS GOLCHERT<sup>1</sup>, LUTZ MARDER<sup>1</sup>, DANA BLOSS<sup>1</sup>, CATMARNA KÜSTNER-WETEKAM<sup>1</sup>, PETER BAUMGÄRTEL<sup>3</sup>, ANDREAS HANS<sup>1</sup>, and ARNO EHRESMANN<sup>1</sup> — <sup>1</sup>Institut für Physik und CINSaT, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Deutschland — <sup>2</sup>European XFEL, Holzkoppel 4, 22869 Schenefeld, Deutschland — <sup>3</sup>Helmholtz-Zentrum Berlin, BESSY II, Abteilung Optik und Strahlrohre, Albert-Einstein-Str. 15,

12489 Berlin, Deutschland

The H<sub>2</sub> *B-X* system is investigated using its Condon diffraction bands in order to test for a shift between the independently calculated electronic potential energy curves of the *B* and *X* electronic states. The Condon diffraction bands for the rovibronic states *B* (*v'* = 8, 9, ..., 13, *J'* = 0) are calculated from literature potentials, to which internuclear distance shifts are applied. Data from a first measurement of the H<sub>2</sub> photon-excitation photon-emission map is analyzed and compatible with the unmodified literature potentials. Planned future measurements will significantly improve the accuracy and uncertainty.

MO 11.3 Wed 11:30 HS 3044

**X-ray absorption of CF<sub>4</sub> driven by x-ray free-electron laser**

— •RUI JIN<sup>1</sup>, ADAM FOUA<sup>2</sup>, ALEXANDER MAGUNIA<sup>1</sup>, MARC REBHOLZ<sup>1</sup>, ALBERTO DE FANIS<sup>3</sup>, KAI LI<sup>2</sup>, GILLES DOUMY<sup>2</sup>, JAN-ERIK RUBENSSON<sup>4</sup>, MARIA NOVELLA PIANCASTELLI<sup>5</sup>, MARC SIMON<sup>5</sup>, THOMAS BAUMANN<sup>3</sup>, MICHAEL STRAUB<sup>1</sup>, SERGEY USENKO<sup>3</sup>, YEVHENY OVCHARENKO<sup>3</sup>, TOMMASSO MAZZA<sup>3</sup>, NINA ROHRINGER<sup>6,7</sup>, MICHAEL MEYER<sup>3</sup>, LINDA YOUNG<sup>2,8</sup>, CHRISTIAN OTT<sup>1</sup>, and THOMAS PFEIFER<sup>1</sup> — <sup>1</sup>MPIK, Heidelberg — <sup>2</sup>Argonne National Laboratory, USA — <sup>3</sup>European XFEL, Schenefeld — <sup>4</sup>Uppsala University, Sweden — <sup>5</sup>Sorbonne Universités, Paris — <sup>6</sup>DESY, Hamburg — <sup>7</sup>Universität Hamburg — <sup>8</sup>University of Chicago, USA

X-ray absorption spectroscopy (XAS) is widely used to study atomic and molecular structure and dynamics, especially at the core level. X-ray free electron lasers (XFEL) have introduced coherent, high-brilliance, and ultrashort laser pulses with tunable energies, thereby enabling the study of multiphoton x-ray matter interactions. In this work, the EuXFEL is combined with a grating spectrometer for a single-pulse transient absorption study of the intermediate electronic states arising from multiphoton-induced molecular dynamics in CF<sub>4</sub>. The central photon energy is tuned to initiate dynamics with and without producing fluorine K-holes. The spectra are measured at different FEL intensities to study multiphoton effects. Two main findings are: (1) neutral fluorine atoms are observed within the pulse duration (40 fs), and (2) short-lived molecular fragments with fluorine 1s-core hole state are observed for high intensities. Precise atomic and molecular structure calculations as well as semi-classical molecular dynamics are used to interpret the results. Overall, this study demonstrates the potential of XFEL-driven transient-absorption spectroscopy to study ultrafast multiphoton dynamics of molecular systems.

MO 11.4 Wed 11:45 HS 3044

**Simulation of X-ray photoelectron spectroscopy in atoms, molecules, and clusters: Core-electron excitation from ab initio many-body approach**

— •ISKANDER MUKATAYEV<sup>1</sup>, GABRIELE D'AVINO<sup>2,3</sup>, FLORIENT MOEVUS<sup>1</sup>, BENOÎT SKLÉNARD<sup>1,4</sup>, VALERIO OLEVANO<sup>2,3,4</sup>, and JING LI<sup>1,4</sup> — <sup>1</sup>Université Grenoble Alpes, CEA, Leti, F-38000, Grenoble, France — <sup>2</sup>Université Grenoble Alpes, F-38000 Grenoble, France — <sup>3</sup>CNRS, Institut Néel, F-38042 Grenoble, France — <sup>4</sup>European Theoretical Spectroscopy Facility (ETSF)

X-ray photoelectron spectroscopy (XPS) technique, measuring directly core-electrons binding energies (BEs), provides information about electronic structure, chemical bonding, and stoichiometry for molecules/solids. This work presents the benchmark study of core electrons BEs in noble gas atoms between theories, including density functional theory (DFT), Hartree-Fock (HF) and many-body theory perturbation theory (GW approach) against experiments first, pointing out significant improvement of computed BEs from HF/DFT to GW. Furthermore, XPS of noble gas clusters with 3000 atoms were studied with embedded many-body theory to estimate the environmental polarization effect on relative BEs (chemical shifts). An analytical formula derived from classical electrostatics accurately describes these polarization effects, aligning well with experimental XPS for noble gas clusters. Finally, by investigating the core-electron excitation in carbon 1s among various molecules, we found that the main contribution to chemical shift comes from classical electrostatic interaction and is one order of magnitude larger than the correlation effects.

MO 11.5 Wed 12:00 HS 3044

**Soft X-Ray-induced Dimerization of Methane** — •SIMON

REINWARDT<sup>1</sup>, IVAN BAEV<sup>1</sup>, PATRICK CIESLIK<sup>1</sup>, KAROLIN BAEV<sup>2</sup>, TICIA BUHR<sup>3</sup>, ALEXANDER PERRY-SASSMANNSHAUSEN<sup>3</sup>, STEFAN SCHIPPERS<sup>3</sup>, ALFRED MÜLLER<sup>3</sup>, FLORIAN TRINTER<sup>4</sup>, JENS VIEFHAUS<sup>5</sup>, and MICHAEL MARTINS<sup>1</sup> — <sup>1</sup>Universität Hamburg, Hamburg, Deutschland — <sup>2</sup>Deutsches Elektronen-Synchrotron, Hamburg, Deutschland — <sup>3</sup>Justus-Liebig-Universität Gießen, Gießen, Deutschland — <sup>4</sup>Fritz-Haber-Institut, Berlin, Deutschland — <sup>5</sup>Helmholtz-Zentrum Berlin für Material und Energie, Berlin, Deutschland

Carbon 1s photo excitation of methane, CH<sub>4</sub> and subsequent multimerization in CH<sub>4</sub> gas has been studied using the PIPE ion-trap setup [1] at PETRA III of DESY. Photoions resulting from the decay of the 1s vacancy are stored within the ion trap so that they can undergo reactions with the surrounding neutral methane molecules. The experimental results clearly show that the initial photoionization event leads to the formation of reaction products with up to three carbon atoms [2]. Accordingly, such addition reactions can play an important role in the formation of larger molecular ions in planetary ionospheres. A better understanding of photoinduced reactions forming larger molecules is essential for understanding the chemistry and chemical composition of atmospheres such as those of exoplanets, whose investigation is planned with celestial observatories like the James Webb Space Telescope.

[1] S. Reinwardt et al., *Rev. Sci. Instrum.* **94**, 023201 (2023).

[2] S. Reinwardt et al., *Astrophys. J.* **952**, 39 (2023).

MO 11.6 Wed 12:15 HS 3044

**Multi-electron emission from irradiated iodide anion in solution**

— •YUSAKU TERAOKA, DANA BLOSS, GABRIEL KLASSEN, JOHANNES VIEHMANN, ADRIAN KRONE, NIKLAS GOLCHERT, ARNO EHRESMANN, and ANDREAS HANS — Institut für Physik und Center for Interdisciplinary Nanostructure Science and Technology (CINSA-T), Universität Kassel, Heinrich-Plett-Straße 40, 34132 Kassel

Radiation effects in solvated matter is of great interests, since many aspects of them are still poorly understood and better knowledge can be beneficial for radiation protection and radiation therapy. Recently, interatomic/intermolecular processes, that transfer deposited excess energy or charge from the initially ionized size to so surrounding water molecules, were newly discovered. Some of these mechanisms have been experimentally observed in our group for the decay of the ionized Mg dication in aqueous solution by multi-electron coincidence spectroscopy. Here, we focused on iodide anion in solution, which has a large photoionization cross-section at its 3d ionization region and decays by multiple Auger steps, competitors to interatomic/intermolecular processes. For a better understanding of complex decay processes of irradiated atoms and molecules in solution, multi-electron emission from iodide anion in aqueous solution by ionizing 3d iodide electrons was studied by combining the liquid micro jet technique and multi-electron coincidence spectroscopy.

MO 11.7 Wed 12:30 HS 3044

**Solvated pyrimidine molecules as donor or acceptor of X-ray induced intermolecular energy transfer**

— •DANA BLOSS<sup>1</sup>, FLORIAN TRINTER<sup>2,3</sup>, NIKOLAI V. KRZYŻEWOI<sup>4</sup>, ALEXANDER KULEFF<sup>4</sup>, LORENZ S. CEDERBAUM<sup>4</sup>, ARNO EHRESMANN<sup>1</sup>, and ANDREAS HANS<sup>1</sup> — <sup>1</sup>Institute of Physics and Center for Interdisciplinary Nanostructure Science and Technology (CINSA-T), University of Kassel, Heinrich-Plett-Straße 40, 34132 Kassel, Germany — <sup>2</sup>Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany — <sup>3</sup>Institut für Kernphysik, Goethe-University, Max-von-Laue-Str. 1, 60438 Frankfurt am Main, Germany — <sup>4</sup>Institute of Physical Chemistry, University of Heidelberg, Im Neuenheimer Feld 229, 69120 Heidelberg, Germany

We investigated the effect of the presence of an aqueous environment for small bio-relevant organic molecules after their exposure to X-ray irradiation in a photoelectron-ion-ion coincidence experiment performed at the P04 beamline of PETRA III. In the decay of electronic inner-shell vacancies located in the solvated pyrimidine itself or in the water environment of the molecule we found evidence for intermolecular energy transfer in both directions. These processes can protect the molecule from reaching dicationic states via Auger decay and their inevitable fragmentation. The observations are compared with the results of theoretical calculations for a deeper understanding of the occurring effects.

Time: Wednesday 13:00–14:00

Location: HS 3044

All members of the Molecular Physics Division are invited to participate.

**MO 13: Interaction with Strong or Short Laser Pulses II (joint session A/MO)**

Time: Wednesday 14:30–16:30

Location: HS 1010

MO 13.1 Wed 14:30 HS 1010

**Focal volume reduction in pulsed standing waves for xenon multiphoton ionization** — ●TOBIAS HELDT, JAN-HENDRIK OELMANN, LENNART GUTH, NICK LACKMANN, LUKAS MATT, FIONA SIEBER, JANKO NAUTA, THOMAS PFEIFER, and JOSÉ R. CRESPO LÓPEZ-URRUTIA — Max-Planck-Institut für Kernphysik, Heidelberg, Germany

To study the highly nonlinear light-matter interaction of multiphoton or tunnel ionization, intense light fields are needed. We use a femtosecond enhancement cavity to fulfill this requirement by reaching intensities of  $> 10^{13}$  W/cm<sup>2</sup>, even at the high 100 MHz repetition rate of a near-infrared frequency comb. The bow-tie cavity supports counter-propagating pulses, leading to a pulsed standing wave when two pulses overlap in the focus. There, we have integrated a gas nozzle and a velocity-map imaging (VMI) spectrometer to study the angular distribution of the emitted photoelectrons [1].

The joint focus of the counter-propagating pulses leads to a doubling of the maximum intensity. In addition, the ionization region along the beam propagation is also reduced because it no longer depends on the Rayleigh length but on the  $< 200$  fs overlap of the pulses. Our experimental data show that this reduction of the focal volume renders the electrostatic focusing in the VMI technique unnecessary. Furthermore, the standing wave influences the emitted electrons over the structured ponderomotive potential, leading to the Kapitza-Dirac effect.

[1] J.-H. Oelmann et al., *Rev. Sci. Instrum.*, 93(12), 123303 (2022).

MO 13.2 Wed 14:45 HS 1010

**Controlling ionization with chirped circularly-polarized laser pulses** — ●ULF SAALMANN — Max-Planck-Institut für Physik komplexer Systeme, Dresden/Germany

We show that controlling two-photon ionization with a chirp, originally predicted for linearly-polarized pulses [X], applies to circular polarization as well. In this case the underlying mechanism is particularly transparent in the rotating frame. Experimental demonstration of this mechanism for the Helium atom has been achieved at FERMI by the Freiburg group and is presented elsewhere.

[X] Saalman & Giri & Rost, *Phys. Rev. Lett.* 121 (2018) 153203.

MO 13.3 Wed 15:00 HS 1010

**Coulomb-correlated multi-electron states generated by femtosecond laser-triggered nanotip photoemission** — ●RUDOLF HAINDL<sup>1,2</sup>, ARMIN FEIST<sup>1,2</sup>, TILL DOMRÖSE<sup>1,2</sup>, MARCEL MÖLLER<sup>1,2</sup>, JOHN H. GAIDA<sup>1,2</sup>, SERGEY V. YALUNIN<sup>1,2</sup>, and CLAUS ROPERS<sup>1,2</sup> — <sup>1</sup>Department of Ultrafast Dynamics, Max Planck Institute for Multidisciplinary Sciences, Göttingen, Germany — <sup>2</sup>4th Physical Institute, University of Göttingen, Göttingen, Germany

Correlations between electrons are at the core of numerous phenomena in atomic, molecular, and solid-state systems. For free particles, detecting inter-particle correlations remains challenging, as ensemble-averaged detection typically conceals few-body effects.

A powerful approach to induce strong electron-electron correlations is spatio-temporally confined photoemission from field emitters employed in ultrafast electron microscopes. When  $n$  electrons are generated by the same laser pulse at the emitter, their initially meV-scale inter-particle Coulomb repulsion is acceleration-enhanced in a static electric field to an energy exchange of about 2 eV, as confirmed by trajectory simulations.

In our experiment, we measure distinct energy correlations of pair, triple and quadruple free-electron states in transverse and longitudinal direction [1]. Furthermore, we demonstrate control over the magnitude of Coulomb correlations and discuss how they can facilitate non-Poissonian electron pulse statistics with applications in free-electron quantum optics.

[1] R. Haindl et al., *Nat. Phys.* 19, 1410-1417 (2023).

MO 13.4 Wed 15:15 HS 1010

**Strong-field Electron Emission of metal Nanotips with op-**

**tical Single-Cycle Pulses** — ●ANNE HERZIG, LENNART SEIFFERT, and THOMAS FENNEL — University of Rostock, Institute of physics, Albert-Einstein-Straße 23, 18059 Rostock

Exposing nanostructures to strong fields enables the emission of energetic electrons via near-field driven elastic backscattering [1]. The availability of intense single cycle or sub-single cycle waveforms [2, 3] enables to explore the formation and propagation of attosecond electron pulses in previously inaccessible regimes of the strong-field interaction. Recent experimental studies [4] have shown promising results on analyzing the short backscattering electron signal. In this talk, the electron emission from tungsten nanotips under intense single-cycle pulses is inspected theoretically via one-dimensional single-active TDSE simulations. The calculated carrier-envelope phase-dependent photoelectron energy spectra reveal prominent signatures with pronounced differences to previous studies performed with many-cycle pulses [5]. The physical origins behind the observed spectral features are disentangled by extending the famous Simple Man's Model of strong-field physics.

[1] M. F. Ciappina et al., *Rep. Prog. Phys.* 80, 054401 (2017)

[2] A. Wirth et al., *Science* 334, 195 (2011)

[3] M. T. Hassan et al., *Nature* 530, 66 (2016)

[4] H. Y. Kim et al., *Nature* 613, 7945 (2023)

[5] L. Seiffert et al., *J. Phys. B* 51, 134001 (2018)

MO 13.5 Wed 15:30 HS 1010

**Observing Laser-Induced Plasma Dynamics by Time-Resolved Coherent-Diffractive-Imaging** — ●TOM BÖTTCHER, RICHARD ALTENKIRCH, CHRISTIAN PELTZ, THOMAS FENNEL, FRANZISKA FENNEL, and STEFAN LOCHBRUNNER — University of Rostock, Institute of Physics, Albert-Einstein-Str. 23, 18059 Rostock

Resolving the excitation and relaxation dynamics of laser-induced solid state plasmas is crucial for a fundamental understanding of the response of condensed matter targets to intense laser radiation. Knowledge about the influence of laser parameters like the spatial, temporal and spectral pulse structure on the plasma dynamics is essential for tailored laser machining applications. We present a method for observing the plasma dynamics in laser-excited thin gold foils using single-shot pump-probe coherent diffractive imaging. By employing a phase retrieval algorithm, we can reconstruct the 2D-spatial and time resolved complex transmission from recorded diffraction patterns. Our targets are 30 nm thick, free-standing gold foils that are excited by a focused femtosecond (fs)-800 nm pump pulse and subsequently imaged by a low intensity fs-400 nm pulse. The plasma dynamics are monitored on a time scale from 50 fs to 2 ns giving access to the ultrafast excitation (fs-ps regime) as well as the melting and ablation (ps-ns regime) dynamics.

MO 13.6 Wed 15:45 HS 1010

**Extreme-UV microscopy at ultimate spatial and temporal scales** — ●SERGEY ZAYKO<sup>1</sup>, HUNG-TZU CHANG<sup>1</sup>, OFER KFIR<sup>2</sup>, MURAT SIVIS<sup>1</sup>, and CLAUS ROPERS<sup>1</sup> — <sup>1</sup>Department of Ultrafast Dynamics, Max-Planck-Institute for Multidisciplinary Sciences, 37077 Göttingen, Germany — <sup>2</sup>School of Electrical Engineering, Faculty of Engineering, Tel Aviv University, 69978Tel Aviv, Israel

Future developments in logic and storage devices heavily rely on versatile research tools operating at the relevant spatio-temporal scales. In applied research fields such as spintronics and strongly correlated electronic materials, these extend into previously unreachable femtosecond-nanometer regimes [1]. In this work, we demonstrate an experimental advance towards such capabilities with femtosecond element-specific, spin-sensitive microscopy at ultimate spatio-temporal scales, achieving simultaneous 18 nm spatial and 35 fs temporal resolution. This allows for a close examination of ultrafast phenomena in real space, providing, deeper insights into the puzzles surrounding ultrafast spin dynamics in the presence of nanoscale magnetic domains [2]. By optimizing the experimental conditions for static imaging, we

demonstrate real-space resolutions of 13.5 nm and 12.5 nm for spin and charge scattering, using probe wavelengths close to the m-edges of Co and Ni, respectively. These results from our compact high-harmonic based microscope establish a set of new benchmarks for photon-based imaging techniques.

- [1] Zayko et al., Nat. Commun. 12, 6337 (2021)
- [2] Koopmans et al., Nat. Materials 9, 259-265 (2010)

MO 13.7 Wed 16:00 HS 1010

**Tracing attosecond electron emission from a nanometric metal tip** — ●LENNART SEIFFERT<sup>1</sup>, PHILIP DIENSTBIER<sup>2</sup>, TIMO PASCHEN<sup>2</sup>, ANDREAS LIEHL<sup>3</sup>, ALFRED LEITENSTORFER<sup>3</sup>, THOMAS FENNEL<sup>1,4</sup>, and PETER HOMMELHOFF<sup>2</sup> — <sup>1</sup>University of Rostock — <sup>2</sup>University of Konstanz — <sup>3</sup>Friedrich-Alexander-Universität Erlangen-Nürnberg — <sup>4</sup>Max Born Institute Berlin

Solids exposed to intense electric fields release electrons through tunnelling. This fundamental quantum process lies at the heart of various applications such as petahertz vacuum electronics where electron wavepackets undergo semiclassical dynamics in an intense laser field, similar to strong-field physics in the gas phase. Recently, we measured the subcycle-dynamics at solids, including the duration of the emission time window [1] and the temporal width of the recolliding wavepacket [2]. Here I present how the suboptical-cycle strong-field emission dynamics from a metallic nanotip is uncovered via two-colour modulation spectroscopy [1,3], where energy spectra of emitted photoelectrons are measured as function of the relative phase between the colors. Projecting the solution of the time-dependent Schrödinger equation onto classical trajectories relates phase-dependent signatures

in the spectra to the emission dynamics and yields an emission duration of  $710 \pm 30$  attoseconds.

- [1] P. Dienstbier et al., Nature 616, 702-706 (2023)
- [2] H. Y. Kim et al., Nature 613, 662-666 (2023)
- [3] L. Seiffert et al., J. Phys. B 51, 134001 (2018)

MO 13.8 Wed 16:15 HS 1010

**Axially Polarized Photoelectrons in Strong-Field Ionization** — ●PEI-LUN HE, ZHAO-HAN ZHANG, KAREN Z. HATSAGORTSYAN, and CHRISTOPH H. KEITEL — Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany

The spin effects in strong-field ionization induced by a linearly polarized laser field are investigated, demonstrating that the photoelectrons exhibit axial polarization relative to the laser polarization axis typically. While the total polarization vanishes upon averaging over the photoelectron momentum, significant momentum-resolved spin polarization is found. The polarization originates from the spin-orbit coupling in the bound state, establishing a correlation between the orbital angular momentum and the spin of the valence shell electron. Consequently, the correlation extends to the spin and the initial transverse velocity of the photoelectron at the tunnel exit. The electron trajectories are thus spin-dependent and are scattered into different directions upon recollisions, resulting in the entanglement of the angular distribution with the electron spin. Furthermore, the interference between direct and rescattered electrons leads to the feasibility of spin-polarized electron holography, offering structural information about the atom.

## MO 14: Atomic Clusters (joint session A/MO)

Time: Wednesday 14:30–16:30

Location: HS 1015

MO 14.1 Wed 14:30 HS 1015

**Experimental studies on core-level interatomic Coulombic decay in heterogeneous rare gas clusters** — ●CATMARNA KÜSTNER-WETEKAM<sup>1</sup>, LUTZ MARDER<sup>1</sup>, DANA BLOSS<sup>1</sup>, CHRISTINA ZINDEL<sup>1</sup>, UWE HERGENHAHN<sup>2</sup>, ARNO EHRESMANN<sup>1</sup>, PŘEMYSL KOLORENČ<sup>3</sup>, and ANDREAS HANS<sup>1</sup> — <sup>1</sup>Institut für Physik und CINSaT, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany — <sup>2</sup>Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany — <sup>3</sup>Institute of Theoretical Physics, Charles University, V Holesovickach 2, 180 00 Prague, Czech Republic

To understand the fundamental mechanisms of radiation chemistry in realistic environments, it is crucial to examine prototypical systems where molecules or atoms interact with their surroundings. Weakly bound van der Waals clusters serve as promising model systems for investigating novel relaxation pathways. In contrast to isolated atoms, electronically excited states may now decay via different interatomic processes such as interatomic Coulombic decay (ICD) or radiative charge transfer (RCT). Due to the relatively low probability of ICD following inner-shell ionization in rare gas clusters, multicoincidence spectroscopy is essential for its detection. Here, we present the observation of changes in branching ratios when going from homogeneous Ar and Kr clusters to heterogeneous ArKr clusters. This transition effectively introduces a distinct environment for the excited atom in each cluster, providing valuable insights into the influence of cluster composition on interatomic decay pathways.

MO 14.2 Wed 14:45 HS 1015

**Self-organized supersolidity in ion doped Helium droplets** — ●JUAN CARLOS ACOSTA MATOS, PANAGIOTIS GIANNAKEAS, and JAN MICHAEL ROST — Max Planck Institute for the Physics of Complex Systems, Dresden, Germany

It is well known that crystallized shells, of Helium atoms, a so called snowball, forms around the ion in the otherwise (super-)fluid Helium droplet [1]. Here, we show that for sufficiently large droplets a third regime appears between the snowball and the liquid one with a supersolid structure where the Helium density exhibits a periodic modulation of the particle density on a spherical shell. The periodic modulation emerges due to the inner shell snowball structure that provides a lattice substrate for the outer droplet shells yielding an accumulation of superfluid particles. To identify supersolidity in a geometrically confined scenario of a droplet we combine modified density functional

theory (DFT), allowing us to describe large enough droplets, with a Gaussian Imaginary Time Dependent Hartree (G-ITDH)[2] method which traces the emergence of crystallized structures. Our approach works well as a comparison to Quantum Monte Carlo results [3] for smaller droplets reveals. [1] D. E. Galli et al, J. Phys. Chem. A 2011, 115, 7300-7309 [2] W. Unn-Toc et al, J. Chem. Phys. 137, 054112 (2012) [3] M. Rastogi et al, Phys. Chem. Chem. Phys. 2018, 20, 25569

MO 14.3 Wed 15:00 HS 1015

**Disentangling the decay cascade of inner-shell vacancies in krypton clusters** — ●LUTZ MARDER, CATMARNA KÜSTNER-WETEKAM, NIKLAS GOLCHERT, JOHANNES VIEHMANN, EMILIA HEIKURA, NILS KIEFER, ARNO EHRESMANN, and ANDREAS HANS — Institut für Physik und CINSaT, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany

Noble gas clusters represent prototype systems well-suited for the investigation of fundamental atomic and molecular processes; their van der Waals bonds enable new relaxation pathways not available in isolated systems. Many of these have been studied during the recent years, often using coincidence measurement techniques.

Our state-of-the-art experiment, where electrons and photons are detected in coincidence, allows for investigation of multi-particle decay pathways after ionization with synchrotron radiation. Upon introduction of an inner-shell vacancy in a homogeneous Kr cluster, the well-known atomic relaxation pathways – consisting of Auger-Meitner decays and fluorescence – is altered significantly by the opening of new interatomic relaxation mechanisms such as interatomic Coulombic decay (ICD), electron-transfer mediated decay (ETMD) and radiative charge transfer (RCT), all of which have been observed and are presented here.

MO 14.4 Wed 15:15 HS 1015

**Measurements of Electron-Photon Coincidences from Local and Non-Local Electronic Relaxation Processes in Rare-Gas Clusters after Excitation with Synchrotron Radiation from Multi-Bunch Operation Mode** — ●JOHANNES VIEHMANN<sup>1</sup>, ANDREAS HANS<sup>1</sup>, CHRISTIAN OZGA<sup>1</sup>, NILS KIEFER<sup>1</sup>, EMILIA HEIKURA<sup>1</sup>, LUTZ MARDER<sup>1</sup>, CATMARNA KÜSTNER-WETEKAM<sup>1</sup>, UWE HERGENHAHN<sup>2</sup>, and ARNO EHRESMANN<sup>1</sup> — <sup>1</sup>Institut für Physik und CINSaT, Universität Kassel, Heinrich-Plett-Straße 40, 34132 Kassel, Germany — <sup>2</sup>Fritz Haber Institute of the Max Planck Society, Faradayweg 4-6 14195 Berlin Germany

Investigating interatomic (or intermolecular) processes in dense media is of interest for understanding the emergence of new properties in conglomerates of interacting particles. This is a stepping stone in bottom up approaches to describe complex environments like biological relevant systems. Our group has used electron-photon coincidence measurements to investigate local and non-local electronic relaxation processes after inner-valence excitation with synchrotron radiation of rare gas clusters. Coincidence measurements at synchrotrons have been restricted to single bunch operation modes of the facilities due to necessities of proper time references. Here, we suggest a technique to expand the use of such electron-photon-coincidence measurements to arbitrary synchrotron filling patterns and show first benchmark results.

MO 14.5 Wed 15:30 HS 1015

**Extreme shift of Auger cascade energies after deep inner-shell ionization in rare-gas clusters** — ●NIKLAS GOLCHERT<sup>1</sup>, NILS KIEFER<sup>1</sup>, CATMARN KÜSTNER-WETEKAM<sup>1</sup>, LUTZ MARDER<sup>1</sup>, MINNA PATANEN<sup>2</sup>, CHRISTINA ZINDEL<sup>1</sup>, ARNO EHRESMANN<sup>1</sup>, and ANDREAS HANS<sup>1</sup> — <sup>1</sup>Institut für Physik und CINSA<sup>T</sup>, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany — <sup>2</sup>Nano and Molecular Systems Research Unit, Faculty of Science, P.O. Box 3000, FI-90014, University of Oulu, Oulu, Finland

Closing the gap between isolated atoms and macroscopic objects, clusters serve as ideal prototype systems for fundamental research of local and non-local processes in dense media. By investigating their electron emission spectra after photoionization, detailed insights about the interactions between the constituents of a medium are gained.

Here, we present recent experimental results obtained by multi-electron coincidence spectroscopy showing the strong dependence of Auger cascade energies in clusters on the charge state of the emitting ion caused by the polarization of its surrounding. These findings will deliver valuable information for future spectroscopic experiments on dense media such as clusters or liquids using high-energetic light sources.

MO 14.6 Wed 15:45 HS 1015

**Reconstructing the anisotropic expansion of a laser driven nanoplasma** — ●PAUL TUEMMLER<sup>1</sup>, FELIX GERKE<sup>2</sup>, CHRISTIAN PELTZ<sup>1</sup>, HENDRIK TACKENBERG<sup>1</sup>, BJÖRN KRUSE<sup>1</sup>, BERNHARD WASSERMANN<sup>2</sup>, THOMAS FENNEL<sup>1</sup>, and ECKART RÜHL<sup>2</sup> — <sup>1</sup>University of Rostock, D-18059 Rostock, Germany — <sup>2</sup>Freie Universität Berlin, D-14195 Berlin, Germany

Coherent diffractive imaging (CDI) at X-ray free-electron lasers (FELs) has evolved into a well-established method for the structural investigation of unsupported nanoparticles. This inherently static method can be readily adopted to time-dependent studies by incorporating a second pulse in a pump-probe scheme.

In a recent experiment at LCLS, we utilized this method to study the fundamental process of free plasma expansion into vacuum using the example of laser-pumped SiO<sub>2</sub> nanospheres. The resulting plasma expansion rapidly and isotropically softens the initial surface density step. This, in turn, increases the radial decay of the scattering signal eventually precluding meaningful measurements due to a diminishing signal-to-noise ratio within only a few hundred femtoseconds [1].

Here, we present the results of a follow-up experiment at the Eu-

ropean XFEL where we revisited SiO<sub>2</sub> as a target, but operated in a weaker excitation regime. This approach allowed us to record images over far longer timescales and revealed a strong anisotropic expansion dynamic, as predicted by theory [2].

[1] C. Peltz *et al.*, New J. Phys. **24**, 043024 (2022).

[2] C. Peltz *et al.*, Phys. Rev. Lett. **113**, 133401 (2014).

MO 14.7 Wed 16:00 HS 1015

**Superradiant parametric Mössbauer radiation** — ●ZE-AN PENG, CHRISTOPH H. KEITEL, and JÖRG EVERS — Max-Planck-Institut für Kernphysik, Heidelberg, Germany

Mössbauer nuclei facilitate a broad range of applications based on their spectrally narrow resonances at energies of hard X-rays. However, the narrow resonances render a strong excitation via intense X-ray beams challenging. This motivates a search for alternative excitation sources.

Parametric X-ray radiation (PXR) is a well-known mechanism for generating high-quality x-ray beams, which is based on intense relativistic electron beams passing through crystals. If the crystal contains Mössbauer nuclei, then under suitable conditions spectrally narrow parametric Mössbauer radiation (PMR) can be emitted [1]. Recently, a new scheme of superradiant PXR was proposed which employs coherently modulated electron bunches produced in X-ray free-electron laser accelerators [2]. This boosts the PXR intensity generated from the crystal by orders of magnitude.

Here, we construct a superradiant parametric Mössbauer radiation source, which is rendered possible by an extended configuration in which the conditions for superradiant PXR and the Mössbauer resonance condition can be satisfied simultaneously. After illustrating the operation principle of the source, the properties of the generated X-ray beam and possible applications will be discussed.

[1] O. D. Skoromnik, I. D. Feranchuk, J. Evers, and C. H. Keitel, Phys. Rev. Accel. Beams **25**, 040704 (2022). [2] I. D. Feranchuk, N. Q. San, and O. D. Skoromnik, Phys. Rev. Accel. Beams **25**, 120702 (2022).

MO 14.8 Wed 16:15 HS 1015

**Nonlinear effects in the charge fractionalization of critical chains** — ●FLÁVIA BRAGA RAMOS<sup>1</sup>, IMKE SCHNEIDER<sup>1</sup>, SEBASTIAN EGGERT<sup>1</sup>, and RODRIGO PEREIRA<sup>2</sup> — <sup>1</sup>University of Kaiserslautern-Landau, Kaiserslautern, Germany — <sup>2</sup>International Institute of Physics, Natal, Brazil

Using the density matrix renormalization group we investigate how a single particle excitation is accommodated in a strongly correlated chain using an out-of-equilibrium protocol. By creating an initial Gaussian wave packet with fixed momentum, we are able to control the regime of energy excitations. Remarkably, the late-time dynamics of the wave packet comprises up to three descendent humps: two counter-propagating low-energy modes and an additional high-energy contribution, whose existence depends on the energy scale set in the initial state. We interpret this unconventional charge fractionalization in terms of the nonlinear Luttinger liquid theory which has attracted great theoretical interest in recent years. Our results provide a new perspective to observe the dynamics of critical chains in the whole range of energy excitations which could potentially be realized in ultracold atomic gases.

## MO 15: Spectroscopy of Metal Clusters

Time: Wednesday 14:30–16:30

Location: HS 3042

### Invited Talk

MO 15.1 Wed 14:30 HS 3042

**Metal Cluster opportunities** — ●GEREON NIEDNER-SCHATTEBURG — Fachbereich Chemie, RPTU Kaiserslautern-Landau

Isolated Main group Metal Clusters (MMC) and those of Transition Metals (TMC), both provide for unique physical properties and chemical activities which are neither found in single atoms nor in bulk metals. Variation of cluster sizes provide for a scalable tuning of these properties, with some cluster size and shape related non-scalable exceptions superimposed. Empirical scaling laws often find a descriptive interpretation while the non-scalable exceptions receive or await support by explicit quantum chemical modelling. It is a major prevailing challenge to record and to describe appropriately TMCs electronics and their spin couplings. Upon deposition onto strongly interacting surfaces the properties of MMCs and TMCs may change significantly,

and much less by weakly interacting surfaces.

The presentation reviews and exemplifies some of these aspects in complement to the prior Symposium on the Spectroscopy of Metal Clusters (SYMC). It concludes with a short outline of likely applications and limitations.

MO 15.2 Wed 15:00 HS 3042

**Ultrafast Dynamics of Mass-selected Neutral Cerium Clusters Probed by Femtosecond NeNePo Spectroscopy** — MAX GRELLMANN<sup>1</sup>, NIKITA KAVKA<sup>2</sup>, ●JIAYE JIN<sup>1</sup>, ROLAND MITRIC<sup>2</sup>, and KNUT R. ASMIS<sup>1</sup> — <sup>1</sup>Wilhelm-Ostwald-Institut für Physikalische und Theoretische Chemie, Universität Leipzig — <sup>2</sup>Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Germany

Cerium clusters have unique physical properties, making them a subject of current research. The studies on sizes-selected clusters isolated

in the gas phase provides information on the inherent cluster properties in the absence of a perturbing environment. Here, we report our results on electronic-state-selected vibrational wave-packet dynamics for the mass-selected neutral cerium clusters ( $Ce_{2-4}$ ) in a cryogenic ion trap probed by two-color femtosecond pump-probe spectroscopy involving the negative-neutral-positive excitation scheme (fs-NeNePo). High-level CASSCF calculations and quantum dynamic simulations are performed to disentangle the observed oscillatory nuclear wave-packet dynamics on the dense potential energy surface (PES) for the neutral  $Ce_2$ , which arises mainly from two electronic states containing super-configurations in  $\Pi$  symmetry. Both of the fs-NeNePo spectra for  $Ce_3$  and  $Ce_4$  show coherent vibrational wave-packet dynamics that decay rapidly within 2 picoseconds, suggesting the presence of crossed PESs. These valuable results demonstrate complex electronic and vibrational structures of neutral cerium clusters and the potential of fs-NeNePo to study femtosecond dynamics of mass-selected, neutral and tag-free lanthanide clusters with high density of states.

MO 15.3 Wed 15:15 HS 3042

**Magnetic nanodoping: cobalt doped silver clusters** — ●V. ZAMUDIO-BAYER<sup>1</sup>, K. HIRSCH<sup>1</sup>, L. MA<sup>2</sup>, K. DE KNIJF<sup>3</sup>, X. XU<sup>4</sup>, A. LAWICKI<sup>1</sup>, A. TERASAKI<sup>5</sup>, P. FERRARI<sup>3</sup>, B. VON ISSENDORFF<sup>6</sup>, P. LIEVENS<sup>3</sup>, W.A. DE HEER<sup>7</sup>, J.T. LAU<sup>1,6</sup>, and E. JANSSENS<sup>3</sup> — <sup>1</sup>HZB, DEU — <sup>2</sup>TCNN, CHN — <sup>3</sup>KU Leuven, BEL — <sup>4</sup>UNL, USA — <sup>5</sup>Kyushu U., JPN — <sup>6</sup>U. Freiburg, DEU — <sup>7</sup>Georgia Tech, USA

The magnetic properties of neutral and charged silver metal clusters with a magnetic cobalt atom impurity were investigated experimentally by exploiting the complementary methods of Stern-Gerlach cluster beam deflection and XMCD action spectroscopy and are accompanied by DFT calculations and charge transfer multiplet simulations [*Phys. Rev. Research* **5**, 033103 (2023)]. The influence of the number of valence electrons and the consequences of impurity encapsulation were addressed in free size-selected, singly cobalt-doped silver clusters  $CoAg_n^{0,+}$  ( $n = 2-15$ ). Encapsulation of the dopant facilitates the formation of delocalized electronic shells with complete hybridization of the impurity  $3d$ - and the host  $5s$ -derived orbitals, which results in impurity valence electron delocalization, effective spin relaxation, and a low-spin ground state. Doped clusters with more than nine silver atoms are low-spin systems independent of their charge state, coincident with the increase in stability and decrease in reactivity of endohedrally doped silver clusters. In the exohedral cluster size range, spin pairing in the free electron gas formed by the silver  $5s$  electrons is the dominating driving force determining the local  $3d$  occupation of the impurity and thus the cluster's spin multiplicity.

MO 15.4 Wed 15:30 HS 3042

**Electronic structure of reactive transition metal-oxygen cations** — ●MAYARA DA SILVA SANTOS<sup>1,2</sup>, ROBERT MEDEL<sup>3</sup>, SIMON KRUSE<sup>2,4</sup>, MAX FLACH<sup>1,2</sup>, OLESYA S. ABLYASOVA<sup>1,2</sup>, MARTIN TIMM<sup>2</sup>, BERND VON ISSENDORFF<sup>1</sup>, KONSTANTIN HIRSCH<sup>2</sup>, VICENTE ZAMUDIO-BAYER<sup>2</sup>, TONY STÜCKER<sup>3</sup>, SEBASTIAN RIEDEL<sup>3</sup>, and TOBIAS LAU<sup>1,2</sup> — <sup>1</sup>Universität Freiburg, Freiburg, Germany — <sup>2</sup>Helmholtz-Zentrum Berlin, Berlin, Germany — <sup>3</sup>Freie Universität Berlin, Berlin, Germany — <sup>4</sup>Humboldt-Universität zu Berlin, Berlin, Germany

Discovering compounds that present transition metals with unusual oxidation states or reactive oxygen species (superoxide, peroxide and oxygen-centered radical) is of great scientific and technological interests, as they have key applications as oxidizing agents, catalysts, or reaction intermediates. Here, we use X-ray absorption spectroscopy (XAS) at the oxygen K and metal  $M_3$  or  $N_3$  edges of gas-phase  $[MO_n]^+$  systems ( $M =$  transition metal,  $n =$  integer), in their ground state, to identify the spectroscopic signatures of oxygen ligands and assign the oxidation state of the metal.[1,2] Experiments were performed at the cryogenic ion trap endstation at the beamline UE52-PGM at the Berlin synchrotron radiation facility BESSY II.[3] The highly oxidized and reactive  $[Rh^{VII}O_3]^+$ ,  $[Ru^{VIII}O_4]^{\bullet+}$  and  $[Re^{VII}O_4]^{\bullet+}$  are here investigated via XAS for the first time. References: [1] M. da S. Santos, et al., *Angew. Chem. Int. Ed.* **61**, e202207688 (2022); [2] M. da S. Santos, et al., *ChemPhysChem* **24**, e202300390 (2023); [3] K. Hirsch, et al., *J. Phys. B: At., Mol. Opt. Phys.* **42**, 154029 (2009).

MO 15.5 Wed 15:45 HS 3042

**Electronic state of a dioxidomanganese(V) and bis(mu-oxo) di-manganese oxide cluster revealed by XAS and XMCD** — ●O.S. ABLYASOVA<sup>1,2</sup>, E.B. BOYDAS<sup>3</sup>, M. UGANDI<sup>3</sup>, V. ZAMUDIO-BAYER<sup>1</sup>, K. HIRSCH<sup>1</sup>, M. FLACH<sup>1,2</sup>, M. DA SILVA SANTOS<sup>1,2</sup>, M. TIMM<sup>1</sup>, B. VON ISSENDORFF<sup>2</sup>, M. RÖMELT<sup>3</sup>, and J.T. LAU<sup>1,2</sup> — <sup>1</sup>HZB, Albert-Einstein-Straße 15, 12489 Berlin, Germany; — <sup>2</sup>Universität Freiburg, Hermann-Herder-Straße 3, 79104 Freiburg, Germany — <sup>3</sup>Humboldt Universität, Brook-Taylor-Straße 2, 12489 Berlin, Germany

The  $CaMn_4O_5$  cluster's electronic structure plays a crucial role in understanding dioxygen formation in the Kok cycle. The  $S_4$  state, responsible for  $O_2$  formation, is difficult to observe because of the millisecond time scale of the transition. Two main models for the  $S_4$  state feature different oxidation states of +4 and +5 of the manganese atom at the reaction site. We report on the characterization of cold cationic gas-phase manganese oxide complexes via XAS and XMCD. We identify oxidation and spin states by comparison with reference spectra of manganese compounds with known oxidation states, accompanied by multireference and density functional theory calculations. We demonstrate that cationic  $Mn(V)O_2^+$  is only the second manganese oxo complex to exist in a high-spin state. Our most important result is the identification of a  $Mn_2O_3^+$  species with Mn(V) in a high-spin state. This is the first observation of the elusive Mn(V) high-spin state in a polymanganese oxido complex, which may have implications for the future study of  $CaMn_4O_5$  complex structure.

MO 15.6 Wed 16:00 HS 3042

**The highest oxidation states of iridium probed by soft x-ray absorption spectroscopy** — ●JOAO P. M. DE ARCANTO<sup>1</sup>, M. DA SILVA SANTOS<sup>1,2</sup>, V. ZAMUDIO-BAYER<sup>1</sup>, S. KRUSE<sup>1,3</sup>, M. TIMM<sup>1</sup>, M. FLACH<sup>1,2</sup>, O. S. ABLYASOVA<sup>1,2</sup>, K. HIRSCH<sup>1</sup>, R. MEDEL<sup>4</sup>, S. RIEDEL<sup>4</sup>, and J. T. LAU<sup>1,2</sup> — <sup>1</sup>HZB, Berlin, Germany — <sup>2</sup>Universität Freiburg, Germany — <sup>3</sup>HU, Berlin, Germany — <sup>4</sup>FU, Berlin, Germany

Iridium stands out prominently in the pursuit of higher oxidation states (OS). Recently, a combined theoretical and experimental effort has confirmed the existence of Ir in the OS +VII, +VIII, and the highest known OS for an element, +IX with  $5d^2$ ,  $5d^1$  and  $5d^0$  local electron configuration, but only a few compounds are known [1]. Utilizing the potential of the Ion Trap endstation at the beamline UE52-PGM of the BESSY II synchrotron radiation facility, our group has successfully generated a series of cationic iridium oxides  $[IrO_n]^+$  ( $n = 0-4$ ) in the gas phase using a magnetron sputtering source. Employing a mass filter, Ir species of interest are selected and cooled in an ion trap. We performed X-ray absorption spectroscopy at the iridium  $N_3$ -edge allowing to study the  $5d$  derived valence states as well as the oxygen K-edge, facilitating a comparative analysis across the series of iridium-oxo species [2]. [1] Wang et al., *Nature* **514**, 475 (2014) [2] Da Silva Santos et al., *Angew. Chem.*, **61**, no. 38, e202207688, 2022

MO 15.7 Wed 16:15 HS 3042

**Metal cluster mediated  $N_2$  activation and cleavage** — ●GEREON NIEDNER-SCHATTEBURG — Dept. of Chemistry and State Research Center OPTIMAS, RPTU Kaiserslautern-Landau, 67663 Kaiserslautern

Transition metal clusters (TMC) serve a model systems for chemically active surfaces as e.g. of catalytically active nano particles. Size selected TMCs were characterized before to activate and cleave Dinitrogen ( $N_2$ ) spontaneously and under isothermal cryo conditions [1,2,3]. Here, we report on the current state of understanding and on new findings which elaborate the influence of cluster charge state, and which elaborate on cooperative effects amongst multiple adsorbate molecules on the surface of clusters. We discuss the validity of the across-edge above-surface (AEAS) mechanism of  $N_2$  cleavage, and we report on current insights of the underlying interactions. Eventually, we show some results on Dihydrogen ( $H_2$ ) activation, and we give reference to initial experiments on  $N_2 - H_2$  coadsorption [4].

The presentation concludes with an outline of the current and future experimental schedule of TMC investigation in progress.

[1] <https://doi.org/10.1039/D0CP06208A>

[2] <https://doi.org/10.1063/5.0157217>

[3] <https://doi.org/10.1063/5.0157218>

[4] <http://dx.doi.org/10.1021/acs.jpcclett.8b00093>

## MO 16: Ultrafast Dynamics I

Time: Wednesday 14:30–16:30

Location: HS 3044

MO 16.1 Wed 14:30 HS 3044

**Imaging thermal-energy chemical dynamics of solvated (bio)molecular complex system** — ●MUKHTAR SINGH<sup>1,2,3</sup>, MATTHEW SCOTT ROBINSON<sup>1,2,3</sup>, HUBERTUS BROMBERGER<sup>1,2</sup>, SEBASTIAN TRIPPEL<sup>1,2</sup>, and JOCHEN KÜPPER<sup>1,2,3</sup> — <sup>1</sup>Center for Free-Electron Laser Science CFEL, Deutsches Elektronen-Synchrotron DESY, Hamburg — <sup>2</sup>Center for Ultrafast Imaging, Universität Hamburg — <sup>3</sup>Department of Physics, Universität Hamburg

We present the imaging of ultrafast thermal-energy-induced chemical dynamics of a micro-solvated (bio) molecular complex probed with a time-dependent strong field ionization, and ion mass spectroscopy [1]. We produce a pure gas-phase indole-water sample using a combination of a cold molecular beam and the electrostatic deflector [2]. By employing a mid-IR pump to excite a single vibrational mode [3], the resulting thermal-energy chemical dynamics between the indole and water moieties were investigated. The dissociation of the micro-solvated system was monitored using strong-field multi-photon ionization by 1.3  $\mu\text{m}$  wavelength light from a femtosecond pulsed laser, tracking the time-dependent ion signals of the indole-water cluster as well as the individual indole and water ionic products.

- [1] J Onvlee, *et al.*, *Nat Commun.* **13**, 7462 (2022)  
 [2] S. Trippel, *et al.*, *Rev. Sci. Instrum.* **89**, 096110 (2018)  
 [3] M.S. Robinson, *et al.*, *Phys. Chem. Chem. Phys.* (2023)

MO 16.2 Wed 14:45 HS 3044

**Probing ultrafast nonadiabatic dynamics of NO<sub>2</sub> with time-resolved X-ray absorption spectroscopy at N K-edge** — ●LORENZO RESTAINO<sup>1</sup>, ZHUAN-YANG ZHANG<sup>2</sup>, MICHAEL COATES<sup>1</sup>, MICHAEL ODELIUS<sup>1</sup>, MARKUS KOWALEWSKI<sup>1</sup>, ERIK T. J. NIBBERING<sup>2</sup>, and ARNAUD ROUZÉ<sup>2</sup> — <sup>1</sup>Stockholm University, AlbaNova University Center, SE-106 91 Stockholm, Sweden — <sup>2</sup>Max-Born-Institute, Max-Born-Str. 2A, 12489 Berlin, Germany

Time-resolved X-ray absorption spectroscopy (tr-XAS) is a chemically sensitive method well-suited for investigating the intricate behavior of electronically excited molecules. We employed tr-XAS to explore the ultrafast nonadiabatic processes occurring at the conical intersection (CI) between the electronic ground and first excited state of nitrogen dioxide. Despite the success of the experimental tr-XAS spectrum in revealing photodissociation, its capacity to capture the CI dynamics was hindered by the restricted temporal resolution of the 60 fs UV pump pulse. To overcome this constraint, we performed high-level quantum dynamics simulations with a shorter pump pulse, allowing us to access the complex nonadiabatic relaxation events. In our study, an 8-fs UV pump pulse at 400 nm excited the system from the ground state, followed by a 1-fs Gaussian probe pulse at the nitrogen K-edge to probe the core-hole states. By using this temporally resolved pump, we were able to map the system's passage through the conical intersection, identified through a spectral signature indicative of wave packet bifurcation. Consequently, we achieved a detailed tracing of the NO+O dissociation process.

MO 16.3 Wed 15:00 HS 3044

**Investigating the Ultrafast Molecular Relaxation of 4-Thiouracil Using Time-Resolved X-Ray Photoelectron Spectroscopy** — ●DENNIS MAYER<sup>1</sup>, DAVID PICCONI<sup>2</sup>, MATTEO BONANOMI<sup>3,4</sup>, MILTCHO DANAILOV<sup>5</sup>, ALEXANDER DEMIDOVICH<sup>5</sup>, MICHELE DEVETTA<sup>4</sup>, MICHELE DI FRAIA<sup>5</sup>, DAVIDE FACCIOLA<sup>4</sup>, RAIMUND FEIFEL<sup>6</sup>, CESARE GRAZIOLI<sup>7</sup>, FABIANO LEVER<sup>1</sup>, NITSH PAL<sup>8</sup>, VASILIS PETROPOULOS<sup>3</sup>, KEVIN PRINCE<sup>5</sup>, OKSANA PLEKAN<sup>5</sup>, RICHARD SQUIBB<sup>6</sup>, CATERINA VOZZI<sup>4</sup>, GIULIO CERULLO<sup>3,4</sup>, and MARKUS GÜHR<sup>1,9</sup> — <sup>1</sup>DESY, Hamburg, Germany — <sup>2</sup>University of Groningen, The Netherlands — <sup>3</sup>Politecnico di Milano, Italy — <sup>4</sup>CNR-IFN, Milan, Italy — <sup>5</sup>Elettra-Sincrotrone Trieste, Italy — <sup>6</sup>University of Gothenburg, Sweden — <sup>7</sup>CNR-IOM, Trieste, Italy — <sup>8</sup>Heriot-Watt University, Edinburgh, UK — <sup>9</sup>University of Hamburg, Germany

Recent experiments on 4-thiouracil observed different time constants for the UV-induced relaxation into its triplet state that go beyond the difference between experiments in the gas and solution phase [1,2]. Utilizing the element- and site-selectivity of x-rays, we studied the relaxation process 4-thiouracil using gas-phase time-resolved x-ray photoelectron spectroscopy (XPS) at the free-electron laser FERMI. Lifetimes of the chemical shifts at the S 2p edge support previous gas-

phase experiments [1]. In comparison to its isomer 2-thiouracil [3], the molecule shows an additional excited-state spectral feature.

- [1] *Chem. Phys.* **515**, 572 (2018); [2] *J. Am. Chem. Soc.* **140**, 16087-16093 (2018); [3] *Nat. Comm.* **13**, 198 (2022)

MO 16.4 Wed 15:15 HS 3044

**Investigating the ultrafast dynamics of photoexcited azobenzene with an x-ray free electron laser** — ●FABIANO LEVER<sup>1</sup>, DENNIS MAYER<sup>1</sup>, ATIA TUL NOOR<sup>1</sup>, GESA GOETZKE<sup>1</sup>, JULIUS SCHWARZ<sup>2</sup>, MICHAEL MARTINS<sup>2</sup>, REBECCA INGLE<sup>3</sup>, STEFAN DUESTERER<sup>1</sup>, STEFFEN PALUTKE<sup>1</sup>, TARAN DEXTER CYRUS DRIVER<sup>4</sup>, ULRIKE FRUEHLING<sup>1</sup>, and MARKUS GÜHR<sup>1</sup> — <sup>1</sup>DESY, Hamburg, DE — <sup>2</sup>Hamburg Universität, DE — <sup>3</sup>UCL, London, UK — <sup>4</sup>SLAC, Menlo Park, USA

Ultraviolet excitation triggers a light-induced isomerization reaction in the molecular switch azobenzene, changing its geometry from the trans ground state into the cis isomer. This work presents results from an ultrafast UV pump / soft x-ray probe experiment at the Free-Electron Laser FLASH. The electronic state dynamics of photoexcited azobenzene is probed with time-resolved x-ray absorption and photoelectron spectroscopy. In both observables, we measure dynamical features on a sub-picosecond scale. Comparing the experimental results to theoretical calculations for both datasets, we identify the ultrafast relaxation of the initially photoexcited S<sub>2</sub> ( $\pi\pi^*$ ) to lower states.

MO 16.5 Wed 15:30 HS 3044

**Capturing ultrafast dynamics of bio-relevant molecules combining few-femtosecond UV pulses with electropray ionization** — ●SERGEY RYABCHUK<sup>1,2</sup>, AARATHI NAIR<sup>1</sup>, JOSINA HAHNE<sup>1,2</sup>, LAURA PILLE<sup>3</sup>, JULIETTE LEROUX<sup>3,4</sup>, NICOLAS VELASQUEZ<sup>5</sup>, BART OOSTENRIJK<sup>1</sup>, ERIK P. MÅNSSON<sup>6</sup>, LUCAS SCHWOB<sup>3</sup>, VINCENT WANIE<sup>6</sup>, SADIA BARI<sup>7</sup>, and FRANCESCA CALEGARI<sup>1,2,6</sup> — <sup>1</sup>The Hamburg Centre for Ultrafast Imaging, Hamburg, Germany — <sup>2</sup>Universität Hamburg, Hamburg, Germany — <sup>3</sup>DESY, Hamburg, Germany — <sup>4</sup>Université de Caen Normandie, Caen, France — <sup>5</sup>Sorbonne Université, Paris, France — <sup>6</sup>CFEL, Hamburg, Germany — <sup>7</sup>University of Groningen, Groningen, The Netherlands

Ultraviolet (UV) light exposure induces various important chemical and biological processes in nature [1]. Electropray ionization (ESI) technique [2] allows to bring intact large and fragile bio-relevant molecules into the gas phase which is not feasible by other conventional methods. Time-resolved experiments have been recently developed to study the dynamics of such molecules following UV excitation by combining ESI devices with sources of ultrashort laser pulses [3]. However, the time resolution of these studies was limited by 80 fs UV pulses.

The present study involves the merging of an ESI source and an all-in-vacuum beamline, delivering few-fs UV pulses with few-cycle NIR pulses in a pump-probe scheme. This sophisticated experimental setup allows us to track the UV-induced dynamics and non-adiabatic processes in complex systems such as cobalt protoporphyrin IX and deprotonated nucleotides with an unprecedented temporal resolution.

MO 16.6 Wed 15:45 HS 3044

**Excited state dynamics of BN-9,10-naphthalene: The impact of replacing CC by BN** — ●FLORIANE STURM, MICHAEL BÜHLER, CHRISTOPH STAPPER, JOHANNES SCHNEIDER, HOLGER HELTEN, INGO FISCHER, and MERLE RÖHR — Julius-Maximilians-Universität Würzburg

Substituting CC by BN units is an excellent means to modify the optoelectronic properties of PAHs.<sup>1-3</sup> In our research, the excited state spectroscopy and dynamics of BN-9,10-naphthalene were studied by picosecond time-resolved photoionization in a supersonic jet. A REMPI spectrum reveals the S<sub>1</sub> origin at 33841 cm<sup>-1</sup>, which is in very good agreement with theory. Several vibrational bands were resolved and assigned by comparison with computations. A [1+1] photoelectron spectrum via the S<sub>1</sub> origin yielded an adiabatic ionization energy of 8.27 eV. Selected vibrational bands were investigated by pump-probe photoionization. While the origin as well as several low-lying vibronic states exhibit lifetimes in the ns-range, a monoexponential decay is observed at higher excitation energies, ranging from 400 ps, at +1710 cm<sup>-1</sup> to 13 ps at +3360 cm<sup>-1</sup>. Based on quantum chemical calculations, the deactivation is attributed to a conical intersection to the

ground state. In order to access it, an energy barrier has to be passed, which requires sufficient excess energy.

- [1] M. J. D. Bosdet, W. E. Piers, *Can. J. Chem.*, **87**, 8-29 (2009).  
 [2] H. L. van de Wouw, R. S. Klausen, *J. Org. Chem.*, **84**, 1117-1125 (2019).  
 [3] Z. Liu, T. B. Marder, *Angew. Chem. Int. Ed.*, **47**, 242-244 (2008).

MO 16.7 Wed 16:00 HS 3044

**Non-adiabatic electronic relaxation of tetracene studied by time-resolved photoelectron spectroscopy** — ●SEBASTIAN HARTWEG<sup>1</sup>, AUDREY SCOGNAMIGLIO<sup>1</sup>, KARIN S. THALMANN<sup>1</sup>, NICOLAS RENDLER<sup>1</sup>, AARON NGAI<sup>1</sup>, LUKAS BRUDER<sup>1</sup>, PEDRO B. COTO<sup>2</sup>, MICHAEL THOSS<sup>1</sup>, and FRANK STIENKEMEIER<sup>1</sup> — <sup>1</sup>Institute of Physics, University of Freiburg, Germany — <sup>2</sup>Materials Physics Center, Spanish National Research Council, Donostia-San Sebastian, Spain

Polycyclic aromatic hydrocarbons are assumed to be important sources of carbon in the interstellar medium. Additionally, some of these species, especially the acenes consisting of linearly-fused benzene units, are promising candidates for organic semiconductor applications. These applications motivate the fundamental study of the ultrafast excitation dynamics of the acenes and their aggregates to provide a fundamental understanding of the underlying processes and energetics.

I will present a femtosecond time-resolved photoelectron spectroscopy study of tetracene molecules supported by high-level ab initio calculations, revealing the ultrafast non-adiabatic dynamics following the excitation to a bright state in the UV range. The stepwise relaxation via an intermediate dark state to a low lying electronic excited state is accompanied by nuclear motion imprinted in the photoelectron

spectra by the time-dependent Franck-Condon factors of the delayed photoionization step.

MO 16.8 Wed 16:15 HS 3044

**Residue Size Dependency of the Geminate Recombination Dynamics of the Biologically Relevant Disulfide Moiety after UV-cleavage investigated by TRXAS** — ●JESSICA HARICH — Institute of Nanostructure and Solid State Physics, University of Hamburg and Center for Free-Electron Laser Science, Germany

The tertiary structure of proteins is stabilized by disulfide bonds formed from two spatially adjacent L-cysteine residues. These disulfide bridges are prone to UV radiation damage with potentially adverse effects. We employ time resolved X-ray absorption spectroscopy (TRXAS) to observe the UV photochemistry of the natural amino acid dimer L-cystine and the tripeptide Glutathione disulfide in aqueous solution to understand the photochemistry under physiological conditions. Furthermore, we have first exciting insights into the UV-photochemistry of the disulfide bridges within the protein hen egg white Lysozyme.

We find that upon UV irradiation, aliphatic disulfides immediately undergo S-S bond cleavage, leading to the formation of two identical thiyl radicals, followed by fast geminate recombination indicating a very effective recombination process for thiyl radicals to the ground state. This process is only possible in condensed phases and its speed increases with chain length. Our results show that L-cystine already captures the essence of the ultrafast photochemistry of the disulfide bridge, but that the size of the residue adjacent to the disulfide bonds has a strong influence on the immediate recombination dynamics of the photoproducts.

## MO 17: Poster: Cold Molecules

Time: Wednesday 17:00–19:00

Location: Tent C

MO 17.1 Wed 17:00 Tent C

**Photoassociation Spectroscopy of RbYb near the Yb intercombination line** — CHRISTIAN SILLUS<sup>1</sup>, ●ARNE KALLWEIT<sup>2</sup>, and AXEL GÖRLITZ<sup>3</sup> — <sup>1</sup>Heinrich-Heine-Universität Düsseldorf — <sup>2</sup>Heinrich-Heine-Universität Düsseldorf — <sup>3</sup>Heinrich-Heine-Universität Düsseldorf

Ultracold dipolar molecules constitute a promising system for the investigation of topics like ultracold chemistry, novel interactions in quantum gases, precision measurements and quantum information.

Here we report on experiments in our apparatus for the production of ultracold RbYb molecules. This setup constitutes an improvement of our old apparatus with the new approach of using the intercombination line of Yb for photoassociation. In the new setup a major goal is the efficient production of ground state RbYb molecules.

We employ optical tweezers to transport individually cooled samples of Rb and Yb from their separate production chambers to a dedicated science chamber. Here we start to study interspecies interactions of different isotopes by overlapping crossed optical dipole traps. To explore the pathways towards ground state molecules we start with photoassociation spectroscopy.

MO 17.2 Wed 17:00 Tent C

**Collisions in a quantum gas of bosonic  $^{23}\text{Na}^{39}\text{K}$  molecules** — ●MARA MEYER ZUM ALTEN BORGLÖH<sup>1</sup>, JULE HEIER<sup>1</sup>, PHILIPP GERSEMA<sup>1</sup>, KAI KONRAD VOGES<sup>3</sup>, CHARBEL KARAM<sup>2</sup>, LEON KARPA<sup>1</sup>, OLIVIER DULIEU<sup>2</sup>, and SILKE OSPELKAUS<sup>1</sup> — <sup>1</sup>Leibniz Universität Hannover, Institut für Quantenoptik — <sup>2</sup>Université Paris-Saclay, CNRS, Laboratoire Aimé Cotton — <sup>3</sup>Centre for Cold Matter, Blackett Laboratory, Imperial College London

We report on our experiments with quantum gases of polar  $^{23}\text{Na}^{39}\text{K}$  molecules. We discuss both atom-molecule and molecule-molecule collisions including the origin of loss processes in a cloud of chemically stable molecules. Furthermore, we discuss a method for suppressing molecular loss using a coherent two-photon transition to induce a potential barrier that protects the colliding molecules from reaching the short range.

MO 17.3 Wed 17:00 Tent C

**Towards cooling and thermalisation of trapped polyatomic molecules** — ●FLORIAN JUNG, JINDARATSAMEE PHROMPAO, and

GERHARD REMPE — Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Straße 1, 85748 Garching, Germany

Cold and controlled molecules offer a myriad of applications ranging from quantum computation to tests of fundamental physics. In particular, polyatomic molecules are of interest, as they exhibit emergent phenomena such as quasi-permanent electric dipole moments or chirality. Their applications are benefiting from or are even inconceivable without cooling the molecules to ultracold temperatures. To this end, increasing the ratio between elastic and inelastic collision rates to allow for collisional thermalisation is an important milestone.

Combining a cryogenic buffer-gas cell with a centrifuge decelerator and an electrostatic trap, trapping molecules for many seconds, we prepared densities of up to  $10^7\text{cm}^{-3}$  for  $\text{CH}_3\text{F}$  molecules at 350 mK, allowing for observation and control of losses from inelastic dipolar collisions [1]. We expect that those can be further suppressed by opto-electrical Sisyphus cooling [2] for which we resort to the  $\text{CF}_3\text{CCH}$  molecule, which seems suitable for this technique and exhibits a large electric dipole moment. This would pave the way for dense and ultracold samples of polyatomic molecules. However, the attractive properties of  $\text{CF}_3\text{CCH}$  come with increased theoretical and experimental complexity, which we present here alongside preliminary measurements.

- [1] M. Koller *et al.*, *Phys. Rev. Lett.* **128**, 203401 (2022).  
 [2] A. Prehn *et al.*, *Phys. Rev. Lett.* **116**, 063005 (2016).

MO 17.4 Wed 17:00 Tent C

**Design of a new apparatus for creating dipolar quantum gases strongly coupled to an optical cavity** — ●JOHANNES SEIFERT, MARIAN DUERBECK, DALILA ROBLEDO DE BASABE, GERARD MEIJER, and GIACOMO VALTOLINA — Fritz Haber Institute of the MPS, Berlin, Germany

We are designing of a new apparatus at the Fritz Haber Institute for studying dipolar systems of atoms and molecules strongly coupled to an optical cavity. Light-matter coupling can be used to realize exotic many body phases from the competition between different types of long-range interactions (dipolar vs light-mediated) or to control chemical reactions. We report on our efforts to create a quantum gas of dysprosium atoms in a preliminary version of the apparatus.

MO 17.5 Wed 17:00 Tent C

**Measurement of absolute partial and total ionization cross sections of fluorine-based ozone-damaging molecules** —

•MEVLUT DOGAN, DEEPTHY THOMAS MOOTHERIL, WANIA WOLFF, HUGO LUNA, THOMAS PFEIFER, and ALEXANDER DORN — Max-Planck-Institut für Kernphysik, 69117 Heidelberg, Germany

Electron impact dissociative ionization of fluorine based molecules is studied. In existing experiments mostly relative cross sections are obtained due to the difficult determination of absolute data. We have developed a calibration procedure to convert the relative cross sections measured into absolute values. Our experiments were carried out with a Reaction Microscope. A gas mixing device was implemented to add known quantities of the target gas and a reference gas with known absolute cross section. Using this setup, we minimized calibration errors and the absolute cross-sections of fluorine-based ozone-damaging molecules were measured by electron collision from threshold to the 1keV impact energy range. The ionization cross sections of each fragment ion was measured on the absolute scale.

It has been suggested that molecules containing the CF<sub>3</sub> group may cause fluorine-catalyzed ozone loss in the Earth stratosphere. For example, since CF<sub>3</sub> is stable, it can destroy significant amounts of ozone via catalytic cycles involving CF<sub>3</sub>Ox radicals. Important reactions that may occur in the stratosphere are given in the literature (Scientific Assessment of Ozone Depletion, Chapter 2: Hydrofluorocarbons, 2022). Our experimental results will be compared with theoretical and experimental studies in the literature.

MO 17.6 Wed 17:00 Tent C

**Towards a Fermi gas of lithium-rubidium molecules** — •CHRISTINE FRANK, YUNXUAN LU, and XIN-YU LUO — Max Planck Institute of Quantum Optics, Garching, Germany

I present our progress on building a new setup for producing a Fermi gas of lithium-rubidium (LiRb) molecules. LiRb, with its large dipole moment and high rotational constant, exhibits substantially longer lifetimes and field-linked resonances at lower microwave field strengths than NaK fermionic molecules. These traits facilitate studying the rich phase diagram of a molecular Fermi gas near a field-linked resonance, ranging from a p-wave superfluid of spin-polarized dimers to a Bose-Einstein condensate (BEC) of dipolar tetramers. Our compact vacuum setup comprises two sequential 2D magneto-optical traps and a science cell housing the dual-species 3D. We aim to create 10<sup>6</sup> degenerate LiRb Feshbach molecules in an optically levitated box potential, crucial for reaching temperatures below the critical temperature of tetramer BEC formation. To boost the Li flux in our compact arrangement, we're integrating a Zeeman slowing laser beam into the Li 2D MOT, counter-propagating to the atomic trajectories from the oven. Our simulation suggests a sixtyfold increase in Li atom flux, promising a good starting point for producing a large double-degenerate Bose-Fermi atomic mixture and subsequently a deeply degenerate Fermi gas of LiRb molecules.

MO 17.7 Wed 17:00 Tent C

**Microwave spectroscopy of cold CH<sub>3</sub>F molecules in a microstructured electrostatic trap** — •JINDARATSAMEE PHROMPAO, FLORIAN JUNG, MANUEL KOLLER, MARTIN ZEPPELFELD, ISABEL RABEY, and GERHARD REMPE — Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Straße 1, 85748 Garching, Germany

Polar molecules exhibit strong interaction with an external electric field as well as long-range and anisotropic interaction between themselves. These offer fascinating research opportunities ranging from quantum chemistry to quantum computation. Motivated by these, cooling techniques are improving rapidly to prepare cold and ultracold molecular ensembles. To perform the cooling, information about the rotational state distribution and trapping fields is crucial for theoretical and practical considerations, addressability and controllability of the molecules.

In our experiment [1], we employ state-selective depletion by using only microwaves to determine the rotational *M*-substate population of cold CH<sub>3</sub>F molecules in an electrostatic trap [2]. The used trap provides a strongly-peaked and narrow electric-field distribution. While driving one transition resonantly on the peak of the distribution, other transitions can be driven resonantly in higher or lower fields in the wings of the distribution. This renders direct observation of the electric-field distribution difficult. However, by choosing a suitable transition measurement of the distribution via depletion dynamics seems possible. Preliminary data are presented on the poster.

[1] M. Koller *et al.*, Phys. Rev. Lett. **128**, 203401 (2022).

[2] B. G. U. Englert *et al.*, Phys. Rev. Lett. **107**, 263003 (2011).

MO 17.8 Wed 17:00 Tent C

**Progress on Zeeman slowing and trapping CaF** — •TIMO POLL,

JULIUS NIEDERSTUCKE, PAUL KAEBERT, MIRCO SIERCKE, and SILKE OSPELKAUS — Institut für Quantenoptik, Leibniz Universität Hannover

Recently, great progress has been made in direct laser cooling of molecules to temperatures close to absolute zero [1,2]. However, experiments are limited by the number of molecules that can be captured from molecular beams using typical laser-based trapping methods [3,4]. Here we discuss our approaches to increase the number of molecules in the experiments. We show our experimental results on the Zeeman slower for directly laser-coolable molecules proposed by our group [5] as well as schemes and first experimental steps towards the realisation of a sub-Doppler cooling magneto-optical trap [6,7].

- [1] J. F. Barry *et al.* 2012
- [2] Y. Wu *et al.* 2021
- [3] S. Truppe *et al.* 2017
- [4] L. Anderegg *et al.* 2017
- [5] M. Petzold *et al.* 2018
- [6] S. Xu *et al.* 2021
- [7] S. Xu *et al.* 2022

MO 17.9 Wed 17:00 Tent C

**A new apparatus for investigating collisions and chemical processes with ultracold NaK molecules** — •JAKOB STALMANN<sup>1</sup>, KAI KONRAD VOGES<sup>2</sup>, SEBASTIAN ANSKEIT<sup>1</sup>, FRITZ VON GIERKE<sup>1</sup>, and SILKE OSPELKAUS<sup>1</sup> — <sup>1</sup>Institute of Quantum Optics, Leibniz University Hannover — <sup>2</sup>Centre for Cold Matter, Blackett Laboratory, Imperial College London

Ultracold molecular collisions feature many highly complex and still not understood phenomena, such as formation and loss of long-lived collisional complexes, molecular Feshbach resonances and chemical reactions.

Here, we present our efforts for the construction of a new experimental setup using ultracold <sup>23</sup>Na<sup>39</sup>K ground-state molecules as a platform to investigate such collisional phenomena.

For ground-state molecule creation, we first produce optically trapped ultracold atomic ensembles from a dual-species Zeeman slower and MOT setup. The atoms are optically transported to a science chamber, where molecule preparation takes place by creating weakly bound Feshbach molecules and subsequently transferring them into their ground state by a coherent Raman process. In the science chamber a time of flight-velocity map imaging mass spectrometer will be implemented for the detection of all educt and product particles of molecular collisions. Combined with state-selective pulsed laser ionization and fragmentation schemes this allows us to resolve chemical reaction pathways, explore ultracold reaction dynamics and develop new quantum control techniques for chemical reaction steering.

MO 17.10 Wed 17:00 Tent C

**An Experiment to Measure the Electron's Electric Dipole Moment Using an Ultracold Beam of YbF Molecules** — •MICHAEL ZIEMBA, FREDDIE COLLINGS, RHYS JENKINS, JONGSEOK LIM, BEN SAUER, and MIKE TARBUTT — Centre for Cold Matter, Imperial College London, London, SW7 2AZ, UK

The fact that more matter than antimatter has been produced in the early stages of the universe is unexplained [1]. One precondition is the combined violation of charge conjugation and parity (CP-violation) which is too small in the Standard Model. In almost all theories, CP-violation is also a precondition for the electron to have an electric dipole moment (*d<sub>e</sub>*). In this respect, a measurement of *d<sub>e</sub>* can be a test of theories beyond the Standard Model. The value of *d<sub>e</sub>* can be determined by measuring the precession rate of the electron spin in a strong electric field. Heavy polarized molecules with their high intramolecular fields have already set a limit of  $|d_e| < 4.1 \cdot 10^{-30}$  e cm [2]. To improve on this, we create a collimated, bright beam of laser cooled YbF molecules [3] and have built an experiment to measure *d<sub>e</sub>* with it [4]. I will report the first interferometer fringes recorded on it and present the experiment's key features which allow us to determine *d<sub>e</sub>* with a projected uncertainty of  $5 \cdot 10^{-30}$  e cm per day of measurement [3].

[1] L. Canetti *et al.* New J. Phys. **14** 095012 (2012). [2] T. Roussy, *et. al.* arXiv:2212.11841 (2022). [3] X. Alauze *et al.* Quantum Sci. Technol. **6**, 044005 (2021). [4] N J Fitch, *et al.* Quantum Sci. Technol., **6**, 014006, (2021).

MO 17.11 Wed 17:00 Tent C

**Ionization and Dissociation Energies of Dysprosium Monox-**

ide — ●SASCHA SCHALLER, JOHANNES SEIFERT, GIACOMO VALTOLINA, ANDRÉ FIELICKE, BORIS G. SARTAKOV, and GERARD MEIJER — Fritz-Haber-Institut der Max-Planck-Gesellschaft

Previous reports for the ionization and dissociation energies of dysprosium monoxide are contradictory. Thermochemical studies and electron impact ionization led to estimates for IE and  $D_0$  of DyO, but the values are associated with large uncertainties. Furthermore, a recent measurement of  $D_0(\text{DyO}^+)$  implies  $\Delta H_0 = +0.33(2)$  eV, however, this conflicts with the earlier reported values for IE and  $D_0$  [1]. Here we report on the characterization DyO and  $\text{DyO}^+$  in a supersonic molecular beam by applying a variety of spectroscopic approaches using different REMPI and PFI schemes, MATI, and (V)UV single-photon ionization. Isotope specific excitation schemes allow to obtain rotationally resolved spectra, and several Rydberg-series converging to the ionization limits of different rotational states of  $\text{DyO}^+$ . The Rydberg series can be clearly assigned starting with the lowest  $J=7.5$  state. Beside these long-living Rydberg molecules, a number of short-lived molecular states are found. From the spectroscopic data obtained for the fermionic  $^{161}\text{DyO}$  and the bosonic  $^{162}\text{DyO}$ , the values of IE and  $D_0$  are determined with a high precision. This leads to the conclusion that the reaction  $\text{Dy} + \text{O} \rightarrow \text{DyO}^+ + \text{e}^-$  clearly proceeds exothermic.

[1] M. Ghiasee et al., J. Phys. Chem. A 127 (2023), 169

MO 17.12 Wed 17:00 Tent C  
**Characterization of  $4f^{13}6s^2$  hole states of ytterbium fluoride using resonant multiphoton ionization spectroscopy** — ●LUCA DIACONESCU<sup>1</sup>, STEFAN POPA<sup>2</sup>, SASCHA SCHALLER<sup>1</sup>, ANDRÉ FIELICKE<sup>1</sup>, and GERARD MEIJER<sup>1</sup> — <sup>1</sup>Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — <sup>2</sup>Imperial College London, London, UK

The ionically bonded ytterbium monofluoride molecule YbF, used for measuring the electron's electric dipole moment (eEDM), is capable of entering a  $\text{Yb}^+[4f^{13}6s^2]$  "hole" configuration, defined by excitation of one of the inner 4f shell electrons of the constituent ytterbium ion into the outer shell. The electronic levels derived from this configuration, along with the ones derived from the "normal"  $\text{Yb}^+[4f^{14}6s^1]$  configuration, coexist within the molecule's energy landscape. Using resonance enhanced multiphoton ionization spectroscopy (REMPI), we have rotationally characterized the low lying  $4f_{7/2,1/2}^{-1}$  hole state for  $\nu = 0, 1$ . This knowledge will help improve laser cooling schemes for YbF, thus enabling more precise eEDM measurements. Furthermore, significant differences in the ionization behavior of YbF between the normal and the 4f hole configurations were observed and ionization energies for both configurations were determined.

## MO 18: Poster: Cluster

Time: Wednesday 17:00–19:00

Location: Tent C

MO 18.1 Wed 17:00 Tent C  
**Nickel  $L_3$  excitation energy shifts and spectroscopic signatures revealing different electronic characteristics within cationic nickel halides** — ●MAX FLACH<sup>1,2</sup>, KONSTANTIN HIRSCH<sup>1</sup>, TIM GITZINGER<sup>2</sup>, MARTIN TIMM<sup>1</sup>, MAYARA DA SILVA SANTOS<sup>1,2</sup>, OLESYA ABLASOVA<sup>1,2</sup>, MARKUS KUBIN<sup>1</sup>, TOBIAS LAU<sup>1,2</sup>, BERND VON ISSENDORFF<sup>2</sup>, and VICENTE ZAMUDIO-BAYER<sup>1</sup> — <sup>1</sup>Helmholtz-Zentrum Berlin — <sup>2</sup>Universität Freiburg

Electronic configurations play an important role for the catalytic abilities of late transition metals. Late first row transition metal halides like nickel halides have been of interest in various studies regarding their possible use in catalytic reactions and reactivity studies reveal differences in their reactivity with respect to the halogen ligand. In this study we use x-ray absorption spectroscopy at the nickel  $L_3$ -edge of  $[\text{NiX}]^+$  ( $X=\text{F,Cl,Br,I}$ ) and mono atomic nickel cations in two well defined electronic configuration to show the change from  $\text{NiF}^+$  with a predominant 3d8 configuration to  $[\text{NiX}]^+$  ( $X=\text{Cl,Br,I}$ ) with a predominant 3d9  $\underline{L}$  configuration. Experimentally obtained  $L_3$ -edge shifts of the mono atomic species correspond well to the well established theory for exact one integer change in 3d occupation in atomic core level spectroscopy. The obtained  $L_3$ -edge shifts of the  $[\text{NiX}]^+$  ( $X=\text{F,Cl,Br,I}$ ) series shows shifts associated with an integer change in oxidation state in literature. Comparison between mono atomic with valence electrons in purely atomic orbitals and the diatomic samples with valence electrons contributing to molecular orbitals provides insight on the influence of 3d electrons participating in bonding on  $L_3$  excitation energy shifts.

MO 18.2 Wed 17:00 Tent C  
**DFT and TD-DFT study of the gas-phase nickel tetracarbonyl complex** — ●A. HREBEN<sup>1,2</sup>, O. S. ABLASOVA<sup>2,3</sup>, M. FLACH<sup>2,3</sup>, M. TIMM<sup>2</sup>, M. DA SILVA SANTOS<sup>2,3</sup>, V. ZAMUDIO-BAYER<sup>2</sup>, K. HIRSCH<sup>2</sup>, and J. T. LAU<sup>2,3</sup> — <sup>1</sup>Humboldt-Universität zu Berlin, Brook-Taylor-Straße 2, 12489 Berlin, Germany — <sup>2</sup>HZB, Albert-Einstein-Straße 15, 12489 Berlin, Germany — <sup>3</sup>Universität Freiburg, Hermann-Herder-Straße 3, 79104 Freiburg, Germany

Investigations of transition metal carbonyl complexes have a significant role in the rapid development of coordination chemistry. Metal carbonyls are widely used as catalysts in synthesis and industrial processes and also find applications in electrochemistry and laser chemistry as precursors. The first discovered homoleptic carbonyl complex was  $\text{Ni}(\text{CO})_4$ , whose structure and binding properties are still under discussion. In this research, we focused on calculating the most stable structure of  $\text{Ni}(\text{CO})_4^+$  using DFT methods. Four possible multiplicities of the given complex were analyzed using three different functionals (B3LYP, M06L, TPSSH) and two basis sets (def2-TZVP, 6-311+G(3df)). Based on the comparison of the obtained final total energies of optimized geometries, the method giving minimal energy

was used for further TD-DFT calculations. The obtained simulated oxygen K-edge of the X-ray absorption spectra (XAS) are compared to experimental data. For that matter, gas-phase  $\text{Ni}(\text{CO})_4^+$  complex measurements were performed via XAS in ion yield mode at the oxygen K-edge on mass-selected ions, cryogenically cooled in the Ion-Trap end station located in BESSY II.

MO 18.3 Wed 17:00 Tent C  
**Detailed investigation of unexpected photoelectron spectra via angle-resolved spectroscopy of Gold clusters** — ●STEVE TAKOUAN TCHOUNGA, LUKAS WEISE, and BERND VON ISSENDORFF — Physikalisches Institut der Albert-Ludwigs-Universität Freiburg, Germany

Angle-resolved spectroscopy provides an important test of the theoretical description of clusters since these spectra carry more information than the bare electron binding energies. Specifically, the anisotropy of photoelectron spectra depends on the angular momentum state. [1]. In the experiment cluster anions are produced in a magnetron sputter source, cooled to 7K, and enter a time-of-flight spectrometer for mass selection. Electrons are then detached by linear polarised laser light and projected onto an MCP detector in a velocity map imaging setup.

The presented analysis utilizes the additional information from angle-resolved spectroscopy to gain a better understanding of the electronic structure of the cluster. For  $\text{Au}_{33}^-$  an electronic shell closing is expected, leading to the opening of a new shell for  $\text{Au}_{34}^-$ . The angular momentum character of this new shell is not in accordance with a simple shell model. It also differs from the mixed character as observed for Sodium clusters of the same size [2]. Possible influences of the high-lying d-band are discussed.

[1] A. Piechaczek, C. Bartels, C. Hock, J.-M. Rost, and B. v. Issendorff, Phys. Rev. Lett. 126, 233201 (2021). [2] C. Bartels, C. Hock, R. Kuhnen, M. Walter, and B. v. Issendorff, Physical Review A 88, 043202 (2013).

MO 18.4 Wed 17:00 Tent C  
**New setup for synchrotron x-ray photoelectron spectroscopy on gas-phase size selected clusters** — ●LOTAR KURTI, PHILLIP STÖCKS, FABIAN BÄR, LUKAS WEISE, and BERND V. ISSENDORFF — Physikalisches Institut der Albert-Ludwigs-Universität Freiburg, Germany

A new apparatus has been constructed which will allow performing X-ray photoelectron spectroscopy on mass selected cluster ions at synchrotrons for the first time. The core of the setup is a liquid nitrogen cooled linear Paul trap, in which stored cluster ions will interact with synchrotron radiation. Emitted electrons will be guided by a specially designed magnetic field into a Hemispherical Energy Analyser, where photoelectron spectra are recorded. The clusters will be produced in

a magnetron cluster source and mass selected by a quadrupole mass spectrometer before insertion into the linear ion trap. Measuring element specific binding energies of core levels is expected to yield specific information about the chemical bonding in pure and mixed metal and semiconductor clusters.

MO 18.5 Wed 17:00 Tent C

**High-resolution photoelectron spectroscopy on tantalum and gold clusters** — ●MAZIYAR KAZEMI, FABIAN BÄR, and BERND V. ISSENDORFF — Institute of Physics University of Freiburg

The characteristics of deeply cold tantalum clusters (Ta4\* to Ta23\*) and gold clusters (Au3\* to Au40\*) have been studied using high-resolution photoelectron spectroscopy at 3.9K. Our magnetic bottle time-of-flight photoelectron spectrometer, which employs a time-dependent deceleration for electron package focusing, possesses an energy resolution of  $\Delta E/E = 0.22\%$  (5.5 meV at 2.0 eV kinetic energy for Pt ions). This is five times better than a conventional magnetic bottle spectrometer and competitive with hemispherical energy analyzers, which have the disadvantage of a significantly smaller collection efficiency. Combining the improved spectrometer with a low-jitter, short-pulse picosecond laser operating at 211 nm enables us to inspect states bound with up to 5.9eV binding energy with unprecedented resolution. This allows us to observe features like vibrational progressions or contributions from different isomers that have not been resolved before.

MO 18.6 Wed 17:00 Tent C

**Single-shot electron and ion coincidence spectroscopy of rare gas clusters.** — ●FREDERIC USSLING, YVES ACREMANN, ALESSANDRO COLOMBO, LINOS HECHT, KATHARINA KOLATZKI, MARIO SAUPE, JOSÉ GÓMEZ TORRES, ALEXANDRE ROSILLO VORSIN, and DANIELA RUPP — ETH Zurich, Laboratory for Solid State Physics, John-von-Neumann-Weg 9, 8093 Zurich, Switzerland

Intense short-wavelength pulses from free-electron lasers (FELs) or lab-based high harmonic generation (HHG) sources enable structural investigation of individual nanometre-sized specimens like viruses [1] or clusters [2] via diffraction imaging (CDI). The intense short-wavelength pulses lead to a highly ionized system followed by complex dynamics covering many different time scales. In this context, atomic and molecular clusters can serve as ideal model systems to study light-matter interaction on the nanoscale. Each interaction residual, such as ejected electrons or ions, gives insights into different processes inside the cluster: Direct electron measurement allows to probe (sub-)femtosecond dynamics, including ionization and nanoplasma formation [3]. Cluster dissociation dynamics, proceeding on longer timescales, can typically be studied via ion time-of-flight spectroscopy [3]. Consequently, the simultaneous measurement of ejected electrons and ions through coincidence spectroscopy is a powerful tool to study light-matter interaction. We present first tests towards single-shot electron and ion coincidence spectroscopy combined with CDI of large rare-gas clusters. [1] M. Seibert et al., *Nature* 470,(2011) [2] D. Rupp et al., *Nat Commun* 8, 493 (2017) [3] M. Arbeiter and T. Fennel, *New J. Phys.* 13 053022 (2011)

MO 18.7 Wed 17:00 Tent C

**A new helium droplet source setup for nanoparticle deposition** — ●FABIO ZAPPA, ANNA-MARIA REIDER, THOMAS POHL, JAN MAYERHOFER, MASOOMEH MAHMOODI-DARIAN, ELISABETH GRUBER, and PAUL SCHEIER — Institut für Ionenphysik und Angewandte Physik - Universität Innsbruck

Helium droplets have been extensively used in the last years to produce Taylor-made clusters of various atomic and molecular species, which can be analysed both in \*in flight\* or deposited on surfaces. Surface deposition with helium droplets presents various challenges as well as opportunities which our group is presently exploring. The present communication gives an overview of a new setup that is being developed in our group, which will allow the deposition of doped helium droplets both in neutral or multiply charged state. Various benchmarks and comparisons with other instruments in our lab will be presented, as well as preliminary deposition results with gold as dopant of the droplets.

The work was supported by the Standort Agentur Tirol, K-Regio Project SupremeByNano

MO 18.8 Wed 17:00 Tent C

**Density optimization of a pure indole-water molecular beam for thermal-energy studies** — ●HOSSEIN SABERIANI<sup>1,2,3</sup>, MATTHEW SCOTT ROBINSON<sup>1,2,3</sup>, MUKHTAR SINGH<sup>1,2,3</sup>, HUBERTUS

BROMBERGER<sup>1,2</sup>, SEBASTIAN TRIPPEL<sup>1,2</sup>, and JOCHEN KÜPPER<sup>1,2,3</sup> — <sup>1</sup>Center for Free-Electron Laser Science CFEL, Deutsches Elektronen-Synchrotron DESY, Hamburg — <sup>2</sup>Center for Ultrafast Imaging, Universität Hamburg — <sup>3</sup>Department of Physics, Universität Hamburg

Microsolvated systems, wherein a small number of solvent molecules are clustered around a solute, provide a unique window into solvation effects present in the bulk [1]. In particular, the interaction of biomolecules such as indole with water provides ideal model systems for ultrafast dynamics studies of ambient-temperature/thermal-energy chemistry [2]. To study such systems, we need to start with a pure and high-density molecular beam. Using high-pressure supersonic expansions and the electrostatic deflector, we are able to produce such samples [3]. Here, I will detail how our molecular beam was optimized by tuning valve conditions, and how the absolute density measurements were done using strong-field ionization techniques.

[1] L. He, *et int.*(6 authors), J. Küpper, S. Trippel, *J. Phys. Chem. Lett.* **14**, 10499 (2023)

[2] M. S. Robinson and J. Küpper, *Phys. Chem. Chem. Phys.* **20**, 20205 (2023)

[3] Y.-P. Chang, *et int.*(2 authors), J. Küpper, *Int. Rev. Phys. Chem.* **34**, 557 (2015)

MO 18.9 Wed 17:00 Tent C

**Time-resolved laser photodissociation investigation of a cationic Iridium(III) complex in an ion trap** — ●PHILIPP WEBER<sup>1</sup>, MARCEL J. P. SCHMITT<sup>1</sup>, CHRISTOPH RIEHN<sup>1</sup>, and CHRISTOPH LAMBERT<sup>2</sup> — <sup>1</sup>Department of Chemistry, RPTU Kaiserslautern — <sup>2</sup>Institute for Organic Chemistry, Universität Würzburg

Iridium complexes are well known for their exceptional photophysical properties, which lead to their application in the fields of OLEDs and photovoltaics. [1] Transient laser photodissociation experiments were conducted on an isolated Ir(III) complex in the ion trap of an electrospray ionization mass spectrometer, giving dynamics on time scales from sub-ps to  $\mu$ s. We present preliminary results for intrinsic gas phase dynamics of an Iridium(III) two donor- one acceptor (D<sub>2</sub>-A) system based on a substituted triarylamine as electron donor (D, ppz-TAA) and tetramethyl-phenanthroline (A, tmp) as electron acceptor, which showed a long-lived (tens of ns) non-fluorescing charge-separated state in solution. [2] We observed, both, fragmentation and photoionization (generating the dication) upon photoexcitation, with the latter dominating the signal for pump-probe investigations. The observed dynamics ( $\sim 0.2$  ps,  $\sim 9$   $\mu$ s) point more towards the fast formation of a long-lived triplet electronic state rather than a charge-separated state, with the latter possibly destabilized with respect to solution. We discuss different scenarios of electronic state dynamics for gas phase and solution.

[1] Longhi; De Cola. Iridium(III) Complexes for OLED Application. John Wiley & Sons, 2017. [2] *Chem. Commun.*, 2009, 1670-1672.

MO 18.10 Wed 17:00 Tent C

**Towards Femtochemistry in a Micro-Solvated Environment** — ●DEEPAK K. PANDEY<sup>1</sup>, LILIANA M. RAMOS MORENO<sup>1</sup>, CLAUDIUS-SCHULZ<sup>2</sup>, and JOCHEN MIKOSCH<sup>1</sup> — <sup>1</sup>Institut für Physik, Universität Kassel, Heinrich Plett Str. 40, 34132 Kassel, Germany — <sup>2</sup>Ultrafast XUV-Physics, Max Born Institute (MBI), Max-Born-Straße 2A, 12489 Berlin, Germany

Chemical reactions, intrinsic to both natural processes and technological advancements, exhibit diverse dynamics, particularly within the realm of solution-phase environments. Understanding these dynamics is crucial, especially when employing gas-phase techniques to investigate reactions in solution, a trend that has gained prominence. In contrast to the popular liquid-jet technique, we aim to systematically introduce water molecules to a gas-phase photochemical reaction in a bottom-up approach - one at a time. Our experimental setup utilizes the water cluster technique pioneered by Udo Buck in combination with a Photoelectron Photoion Coincidence (PEPICO) spectrometer. At the University of Kassel, we aim to investigate the impact of microsolvation on chemical reactions, ultimately using VUV light as a probe. The initial focus is on the steric effects of water on photochemical dynamics, employing a conventional pump-probe experiment to study processes such as photodissociation and photo-induced isomerization reactions. Our poster will focus on the experimental approach, characterization of the water cluster source and the spectrometer, and experimental advance toward studying photochemical reactions.

MO 18.11 Wed 17:00 Tent C

**Supramolecular dynamics investigated on hydrogen-bonded pyrrole-water clusters upon site-specific x-ray photoionization** — ●WUWEI JIN<sup>1,2</sup>, IVO VINKLÁREK<sup>1</sup>, HUBERTUS BROMBERGER<sup>1</sup>, SEBASTIAN TRIPPEL<sup>1,3</sup>, REBECCA BOLL<sup>4</sup>, MICHAEL MEYER<sup>4</sup>, and JOCHEN KÜPPER<sup>1,2,3</sup> — <sup>1</sup>Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg — <sup>2</sup>Department of Physics, Universität Hamburg — <sup>3</sup>Center for Ultrafast Imaging, Universität Hamburg — <sup>4</sup>European XFEL GmbH, Schenefeld

The solvation of molecules crucially affects their photostability and introduces additional pathways for relaxation dynamics compared to isolated molecules. To gain molecular-level insights into the solvation effects on the photofragmentation dynamics of a supramolecular system, we investigate the dynamics of a spatially separated pure sample of pyrrole-water (pyr-H<sub>2</sub>O) clusters prepared by the electric deflector [1-2]. This is achieved through an IR-pump-x-ray-probe experiment at EuXFEL. An ionizing IR pulse triggers the (pyr-H<sub>2</sub>O)<sup>+</sup> fragmentation. The resulting dynamics are site-specifically probed by x-ray free-electron laser pulses [3]. This study of the hydrogen-bonded pyrrole-H<sub>2</sub>O system is particularly relevant to pyrrole-containing biomolecules and establishes a new approach to study the key role of intermolecular interactions in supermolecular dynamics.

[1] M Johnny, S Trippel, and J Küpper, *Chem. Phys. Lett.* **721**, 149 (2019) [2] YP Chang, D Horke, S Trippel, and J Küpper, *Int. Rev. Phys. Chem.* **557**, 34 (2015) [3] J Onvlee, S Trippel, and J Küpper, *Nat. Commun.* **7462**, 13 (2022)

MO 18.12 Wed 17:00 Tent C  
**UV photo-induced dissociation dynamics of solvated (bio)molecular complex system** — ●MUKHTAR SINGH<sup>1,2,3</sup>, MATTHEW SCOTT ROBINSON<sup>1,2,3</sup>, HUBERTUS BROMBERGER<sup>1,2</sup>, JOLIYN ONVLEE<sup>1,3</sup>, SEBASTIAN TRIPPEL<sup>1,2</sup>, and JOCHEN KÜPPER<sup>1,2,3</sup> — <sup>1</sup>Center for Free-Electron Laser Science CFEL, Deutsches Elektronen-Synchrotron DESY, Hamburg — <sup>2</sup>Center for Ultrafast Imaging, Universität Hamburg — <sup>3</sup>Department of Physics, Universität Hamburg

We present the investigation of ultrafast chemical dynamics induced by UV excitation in a micro-solvated indole-water-complex system probed by time-dependent strong-field ionization, and ion mass spectroscopy [1]. Indole-water is important due to indole's role as the chromophore of tryptophan, the strongest near UV absorber in proteins. The experimental setup contains a molecular beam and the electrostatic deflector to produce a pure gas-phase sample of indole-water [2]. We conducted a UV-IR pump-probe experiment, wherein we excited the system to the electronic excited state using 270 nm light. The dissociation dynamics of the system was monitored using strong-field multiphoton ionization by 1.3 μm wavelength light from a femtosecond laser, tracking the time-dependent ion signals of the indole and indole-water ions.

[1] J Onvlee, *et al.*, *Nat Commun.* **13**, 7462 (2022)

[2] S. Trippel, *et al.*, *Rev. Sci. Instrum.* **89**, 096110 (2018)

## MO 19: Chirality

Time: Thursday 11:00–12:45

Location: HS 3044

### Invited Talk

MO 19.1 Thu 11:00 HS 3044

**Controlling the internal quantum states of chiral molecules** — JUHYEON LEE, ELAHE ABDIHA, BORIS SARTAKOV, GERARD MEIJER, and ●SANDRA EIBENBERGER-ARIAS — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

Controlling the internal quantum states of chiral molecules enantiomer-specifically has a wide range of fundamental and practical applications. The recently developed method of enantiomer-specific state transfer [1,2] holds great promise in providing the crucial ingredient for enabling the first experimental measurement of the long-predicted parity violation in chiral molecules. It also has the potential of enabling spatial separation of enantiomers in the gas phase and opening new avenues in chiral studies. For this to be possible, (almost) perfect enantiomer-specific state transfer is necessary. However, previous studies have been limited by thermal population in all involved quantum states for all currently accessible molecular temperatures. I will present recent efforts of our group targeted at increased state-specific enantiomeric enrichment and quantitative understanding thereof [3,4]. I will also give an outlook on future experimental directions. [1] S. Eibenberger, J. Doyle, D. Patterson, *Phys. Rev. Lett.* **118**, 123002 (2017) [2] C. Pérez, A. L. Steber, S. R. Domingos, A. Krin, D. Schmitz, M. Schnell, *Angew. Chem. Int. Ed.* **56**, 12512 (2017) [3] J.H. Lee, J. Bischoff, A. O. Hernandez-Castillo, B. Sartakov, G. Meijer, S. Eibenberger-Arias, *Phys. Rev. Lett.* **128**, 173001 (2022) [4] J.H. Lee, J. Bischoff, A.O. Hernandez-Castillo, E. Abdiha, B. Sartakov, G. Meijer, Sandra Eibenberger-Arias, arxiv: 2310.11120 (2023)

MO 19.2 Thu 11:30 HS 3044

**Towards perfect enantiomer-specific state transfer of chiral molecules** — ●ELAHE ABDIHA, JUHYEON LEE, JOHANNES BISCHOFF, DANIEL FONTOURA BARROSO, BORIS SARTAKOV, GERARD MEIJER, and SANDRA EIBENBERGER-ARIAS — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany

Chiral molecules are important in many chemical and biological processes and are also at the heart of some fundamental physics questions. Recently, enantiomer-specific state transfer (ESST) was experimentally demonstrated [1,2]. Here, the application of three mutually orthogonally polarized microwave fields yields enantiomer-specific population control in a chosen quantum state that is part of a triad of rotational states. I will present our recent work on ESST, where we largely overcome the previous limitation due to initial thermal population by combining ESST with optical methods [3]. By depleting the target state using resonant UV light prior to ESST we achieve state-specific enantiomeric enrichment in the order of 50%. Importantly,

we quantitatively study ESST, explicitly including the role of spatial degeneracy. I will also discuss our recent study on the influence of microwave pulse conditions on ESST [4]. Extensions to our scheme will allow to create a molecular beam with an enantiomer-pure rotational level, holding great prospects for future spectroscopic and scattering studies.

[1] Eibenberger et al, *PRL* **118**, 123002 (2017) [2] Pérez et al, *Angew. Chem. Int. Ed.* **56**, 12512 (2017) [3] Lee et al, *PRL* **128**, 173001 (2022)

[4] Lee et al, arxiv: 2310.11120 (2023)

MO 19.3 Thu 11:45 HS 3044

**Purcell effect in chiral environments** — ●OMAR JESUS FRANCA SANTIAGO<sup>1</sup>, CLAUDIO SALVADOR RAPP<sup>1</sup>, JANINE CHRISTINE FRANZ<sup>2</sup>, and STEFAN YOSHI BUHMANN<sup>1</sup> — <sup>1</sup>Institute of Physics, University of Kassel, Germany — <sup>2</sup>University of Freiburg, Germany

The Purcell effect describes the modification of the spontaneous decay rate in the presence of electromagnetic media and bodies. In this work, we shed light on the dependencies and magnitude of this effect for chiral materials. Using the framework of macroscopic quantum electrodynamics [1,2] and Fermi's golden rule, we study a chiral bulk medium with and without local field corrections, an idealised chiral mirror and a chiral surface. The results imply that the chiral effect is greatest for large transition frequencies, molecules with large optical rotatory strength, and media with a strong cross-susceptibility. In the case of a surface, short distances from the molecule to the interface additionally enhance the effect.

[1] D.T. Butcher, S.Y. Buhmann, and S. Scheel, *New J. Phys.* **14**, 113013 (2012).

[2] S. Y. Buhmann, *Dispersion Forces II: Many-Body Effects, Excited Atoms, Finite Temperature and Quantum Friction*, (Springer, Berlin Heidelberg, 2012).

MO 19.4 Thu 12:00 HS 3044

**Silicon 2p inner-shell photoelectron circular dichroism in sec-butyl trimethylsilylether** — ●EMILIA HEIKURA, CHRISTINA ZINDEL, LUTZ MARDER, CATMARNA KÜSTNER-WETEKAM, NIKLAS GOLCHERT, JOHANNES VIEHMANN, DENIS KARGIN, RUDOLF PIETSCHNIG, ANDREAS HANS, and ARNO EHRESMANN — University of Kassel, Heinrich-Plett-Straße 40, 34132 Kassel

Photoelectron circular dichroism (PECD) is one of the most powerful methods for investigating molecular chirality in the gas phase. PECD is a forward-backward asymmetry of emitted photoelectrons from chiral molecules after interaction with a circularly polarized light. The asymmetry can be observed even in randomly oriented chiral molecules.

Site-selectivity of inner-shell photoelectrons enables the investigation on chirality as a function of a distance from a stereocenter. It is still unknown how the magnitude of PECD is affected by the distance of the emission site from the stereocenter. To be able to investigate this phenomenon, a series of specifically synthesized molecules was created. In *sec*-butyl trimethylsilylether and its derivatives the distance between stereocenter and a marker atom can be increased by inserting additional CH<sub>2</sub> groups while otherwise the structure of the molecule stays intact.

MO 19.5 Thu 12:15 HS 3044

**Control of circular dichroism in ion yield of 3-methyl cyclopentanone using femtosecond laser pulses** — •SAGNIK DAS, JAYANTA GHOSH, SUDHEENDRAN VASUDEVAN, HANGYEOL LEE, NICOLAS LADDA, SIMON RANECKY, TONIO ROSEN, TILL STEHLING, FABIAN WESTMEIER, ARNE SENFTLEBEN, THOMAS BAUMERT, and HENDRIKE BRAUN — Institut für Physik, Universität Kassel, Heinrich-Plett-Strasse 40, 34132 Kassel, Germany

Circular dichroism in ion yield (CDIY) is the difference in ion yield from left and right circular polarised light interacting with an enantiomer of a chiral molecule. The cyclic ketone 3-methyl cyclopentanone (3-MCP) has long been studied for its large circular dichroism in the  $\pi^* \leftarrow n$  transition. We used femtosecond laser pulses tuned in wavelength around one of the vibrational modes in the  $\pi^* \leftarrow n$  band to study the effect of pulse parameters like linear chirp and peak intensity on the CDIY. In the 3-photon resonance enhanced multiphoton ionization (1+1+1) of 3-MCP, we observed an enhancement of CDIY for chirped pulses. At the same time, almost no change in CDIY was detected for variation of the peak intensity of bandwidth-limited pulses. Not only the magnitude of the chirp but also its sign influences the CDIY, indicating more than the pulse duration or peak intensity

as the underlying cause. It highlights the role of frequency ordering in the observed enhancement of CDIY for chirped pulses. Moreover, the progression of the CDIY with regard to the applied linear chirp sensitively depends on the central wavelength of the laser pulses.

MO 19.6 Thu 12:30 HS 3044

**Photoelectrons from transiently populated nonresonant states** — •SIMON RANECKY, SUDHEENDRAN VASUDEVAN, HAN-GYEOL LEE, TILL STEHLING, NICOLAS LADDA, TONIO ROSEN, FABIAN WESTMEIER, SAGNIK DAS, JAYANTA GHOSH, HENDRIKE BRAUN, ARNE SENFTLEBEN, and THOMAS BAUMERT — Universität Kassel, Institut für Physik und CINSaT, D-34132 Kassel

When an intense laser pulse induces a two-photon electronic transition from one state to another, all other states are populated transiently with various proportions during the interaction time. This transient population in nonresonant states can absorb further photons and then become ionized so that these states become visible in the photoelectron spectrum. This effect was already observed for Sodium atoms [1].

Here, we report the observation of the same ionization mechanism for the first time on molecules using Fenchone and fs- to ns-lasers around 400 nm. The main ionization scheme for this molecule at this wavelength is 2+1 resonance-enhanced multiphoton ionization. And besides the photoelectrons from resonant s- and p-states, we could see photoelectrons from further, nonresonant states.

With this study, we want to encourage spectroscopists to have a close look at the background of their photoelectron spectra. Doing this for Fenchone and observing the weak signals from transiently populated, nonresonant states, we were able to determine the energies of these states.

[1] Krug et al. N.J. Phys. 11 (2009) 105051

## MO 20: Theoretical Molecular Physics

Time: Thursday 14:30–16:45

Location: HS 3042

MO 20.1 Thu 14:30 HS 3042

**Dynamics of AIF-AIF: Potential energy surface and intermolecular complex characterization** — •XIANGYUE LIU<sup>1</sup>, WEIQI WANG<sup>1</sup>, and JESÚS PÉREZ-RÍOS<sup>2,3</sup> — <sup>1</sup>Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany — <sup>2</sup>Department of Physics and Astronomy, Stony Brook University, Stony Brook 11794, New York, USA — <sup>3</sup>Institute for Advanced Computational Science, Stony Brook University, Stony Brook 11794-3800, New York, USA

Diatomic metal-fluorine molecules are crucial for cryogenic buffer gas cooling, widely used in precision spectroscopy and laser cooling. Recently, AIF has gained attention for laser cooling due to its efficient laboratory production and highly-diagonal Franck-Condon matrix. This study focuses on the undesirable formation of AIF-AIF dimer complexes. We developed an accurate machine-learning potential energy surface for the AIF-AIF complex at the coupled-cluster theory level. Based on the resulting PES, *ab initio* molecular dynamics simulations have been performed, revealing primary reaction mechanisms. The lifetime of the intermediate AIF-AIF complex at different temperatures has been estimated.

MO 20.2 Thu 14:45 HS 3042

**Exotic charged molecules and Rydberg glue** — •DANIEL J. BOSWORTH<sup>1,2</sup>, PETER SCHMELCHER<sup>1,2</sup>, and MATTHEW T. EILES<sup>3</sup> — <sup>1</sup>Zentrum für Optische Quantentechnologien, Universität Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany — <sup>2</sup>The Hamburg Centre for Ultrafast Imaging, Universität Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany — <sup>3</sup>Max Planck Institute for the Physics of Complex Systems, 01187 Dresden, Germany

Neutral-ion interactions are of fundamental interest in physics and chemistry. Recent experiments are starting to explore them within the quantum regime. The sensitivity of Rydberg atoms to electric fields enables them to form weakly-bound diatomic molecular ions. These dimers bind on micrometer length scales due to the Rydberg's large induced dipole moment, which is the leading-order term in the ion-neutral interaction series. In this theoretical work, we explore the role of the higher-order terms in this interaction series. We first consider a system of a pair of cations interacting with a Rydberg atom

at long-range. Surprisingly, we reveal that the Rydberg's quadrupole compensates the enormous Coulomb repulsion between the cation pair, forming a metastable trimer with mixing between states in neighbouring n-manifolds. We discuss the rapid decay of these trimers due to charge transfer of the Rydberg electron. Additionally, we reveal that the quadrupole interaction term introduces a dependence on the sign of the ion's charge, which can significantly alter non-adiabatic couplings between Rydberg states. Such modified couplings would not only affect vibrational dynamics, but also molecular lifetimes.

MO 20.3 Thu 15:00 HS 3042

**The effects of dipole self energy in a molecular Tavis-Cummings model** — •LUCAS BORGES, MARKUS KOWALEWSKI, and THOMAS SCHNAPPINGER — Department of Physics, Stockholm University, AlbaNova University Center, SE-106 91 Stockholm, Sweden

Theoretical studies in polariton photochemistry describe the molecular and photonic field interactions within the long-wavelength limit. The Pauli-Fierz Hamiltonian in the length gauge representation features a linear light-matter interaction term and a squared dipole self-energy (DSE) term, which assures a stable ground state for the total system. However, this representation blurs the distinction between photon and matter degrees of freedom. We performed nuclear wave functions dynamics from one to three MgH<sup>+</sup> molecules interacting with a single photonic mode in an optical cavity at the electronic strong-coupling regime. We investigate how additional DSE terms in the interaction Hamiltonian affect the system by producing new inter-molecular energy transfer pathways.

MO 20.4 Thu 15:15 HS 3042

**Cavity-Born-Oppenheimer Hartree-Fock: Vibronic-Strong-Coupling beyond a single molecule** — •THOMAS SCHNAPPINGER and MARKUS KOWALEWSKI — Department of Physics, Stockholm University, Sweden

When a molecule interacts with the vacuum field of a nanoscale cavity, strong coupling reshapes the potential energy surfaces to form hybrid light-matter states called polaritons. Recent experiments show that this strong coupling between light and matter is capable of modifying chemical and physical properties. The situation in which the quantized cavity modes are coupled via their characteristic frequency to

vibrational degrees of freedom of molecules is called vibrational strong coupling (VSC). In the VSC regime, the chemistry of a single electronic state (mostly the ground state) and its vibrational spectroscopy are influenced by the cavity interaction. In this theoretical contribution we use the *ab-initio* Cavity-Born-Oppenheimer-Hartree-Fock approach to study the effect of VSC on the ground state properties of single molecules and small ensembles of such molecules. We are able to optimize cavity-coupled molecular systems and can calculate vibropolaritonic IR spectra, since we have implemented analytical gradients. Our *ab-initio* treatment allows us to study the interactions between individual molecules mediated by the cavity. These interactions give rise to local strong coupling effects that allow the modification of chemical reactivity in the VSC context.

MO 20.5 Thu 15:30 HS 3042

**Multi-state mapping approach to surface hopping** — ●JOHAN RUNESON and DAVID MANOLOPOULOS — Physical and Theoretical Chemistry Laboratory, University of Oxford, UK

Many important problems in physics and chemistry involve non-adiabatic dynamics: nuclear motion on two or more coupled electronic potential energy surfaces. The most popular method to treat this problem is fewest-switches surface hopping (FSSH), which involves stochastic hops of classical nuclear trajectories between adiabatic electronic states. This method can be used with *ab initio* potentials and is widely applied in photochemistry. However, its long-standing problems are overcoherence and violation of detailed balance. A new ‘mapping approach to surface hopping’ (MASH) [1] appears to resolve many of these issues: it avoids *ad hoc* decoherence corrections and provably relaxes to the correct quantum–classical equilibrium. Although originally developed for two states, we have extended the method to any number of states and applied it to a variety of problems, including gas-phase photochemistry, spin-boson models, and exciton transfer in photosynthetic systems [2]. In all cases, we find MASH to be more accurate as well as more numerically tractable than FSSH. The talk will summarize this development and mention some current exciting applications, including charge transport in organic materials.

[1] J. R. Mannouch and J. O. Richardson, *J. Chem. Phys.* 158, 104111 (2023). [2] J. E. Runeson and D. E. Manolopoulos. *J. Chem. Phys.* 159, 094115 (2023).

MO 20.6 Thu 15:45 HS 3042

**Open quantum system approach to non-adiabatic molecular physics** — MICHAEL REITZ<sup>1</sup>, ●NICO BASSLER<sup>2,3</sup>, RAPHAEL HOLZINGER<sup>4</sup>, ÁGNES VIBÓK<sup>5,6</sup>, GÁBOR HALÁSZ<sup>5</sup>, and CLAUDIU GENES<sup>3,2</sup> — <sup>1</sup>Department of Chemistry and Biochemistry, University of California San Diego, La Jolla, California 92093, USA — <sup>2</sup>Department of Physics, Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU), D-91058 Erlangen, Germany — <sup>3</sup>Max Planck Institute for the Science of Light, D-91058 Erlangen, Germany — <sup>4</sup>Institut für Theoretische Physik, Universität Innsbruck, A-6020 Innsbruck, Austria — <sup>5</sup>Department of Information Technology, University of Debrecen, H-4002 Debrecen, Hungary — <sup>6</sup>ELI-ALPS, ELI-HU Non-Profit Ltd, H-6720 Szeged, Hungary

Non-adiabatic molecular phenomena, i.e., processes due to the breakdown of the Born-Oppenheimer approximation, govern the fate of most photophysical and photochemical processes. We propose here an open quantum system approach based on quantum Langevin equations to non-adiabatic molecular physics, with relevance during or after the process of photoexcitation and in the presence of a dissipative, thermal environment. Based on a linear vibronic coupling model, we derive analytical expressions for the non-radiative transition rates of processes occurring at the intersection between two potential energy surfaces such as internal conversion and conical intersections. Our work allows for an intuitive understanding of these processes in terms of multiphonon sidebands and establishes a connection between open quantum system dynamics, molecular quantum optics, and quantum chemistry.

MO 20.7 Thu 16:00 HS 3042

**Molecular motion enhanced excitation transport in molecular aggregates despite internal molecular vibrations** — ●RITESH PANT<sup>1</sup>, VARADHARAJAN SRINIVASAN<sup>2</sup>, ALEXANDER EISFELD<sup>1</sup>, and SEBASTIAN WÜSTER<sup>2</sup> — <sup>1</sup>Max Planck Institute for the Physics of Com-

plex Systems, Dresden, Germany — <sup>2</sup>Indian Institute of Science Education and Research, Bhopal, India

Molecular aggregates can under certain conditions transport electronic excitation energy over large distances due to the long range dipole-dipole interactions [1]. It was shown earlier that the thermal centre-of-mass motion of molecules can enhance the efficiency of transport compared to the static case in the presence of diagonal disorder, when neglecting molecular vibration [2]. Our current research extends this understanding by examining the impact of molecular vibrations, with a particular focus on adiabatic excitation transport. To simulate quantum dynamics of the electronic excitation coupled to vibrations we use non-Markovian quantum state diffusion, solved through the hierarchy of pure states, combined with classical molecular dynamics for centre-of-mass motion of molecules [2, 3]. Using a specific model of torsional molecular motion, we can identify parameter regimes in which the motion aids excitation transfer even in the presence of vibrations, although adiabatic transport appears disrupted by vibrations.

[1] T. Brixner et. al., *Adv. Energy Mater.* 7, 1700236 (2017). [2] R. Pant et. al., *Phys. Chem. Chem. Phys.* 22, 21169 (2020). [3] D. Suess et. al., *Phys. Rev. Lett.* 113, 150403 (2014).

MO 20.8 Thu 16:15 HS 3042

**Gas-phase sugar synthesis: The formation of protonated glycolaldehyde** — ●WEIQI WANG<sup>1</sup>, HUNARPREET KAUR<sup>2</sup>, SANDRA BRÜNKEN<sup>2</sup>, and JESÚS PÉREZ RÍOS<sup>3,4</sup> — <sup>1</sup>Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany — <sup>2</sup>FELIX Laboratory, Faculty of Science, Radboud University, Toernooiveld 7c, 6525 ED Nijmegen, The Netherlands. — <sup>3</sup>Department of Physics and Astronomy, Stony Brook University, Stony Brook 11794, New York, USA — <sup>4</sup>Institute for Advanced Computational Science, Stony Brook University, Stony Brook 11794-3800, New York, USA

The investigation into the origins and processes of prebiotic synthesis in our universe is pivotal to unraveling the mystery of life’s beginnings. Currently, the understanding of the chemical pathways leading to complexity is incomplete and mostly experiential.

A crucial initial step in understanding prebiotic synthesis involves exploring pathways that give rise to simple sugars. This study delves into the intricate reaction networks governing the formation of protonated glycolaldehyde, a key component in sugar synthesis. Through the *ab initio* molecular dynamics method, we systematically explore the entire relevant phase space. To identify the species observed in infrared (IR) experiments, we calculate IR spectra from simulations at finite ensemble temperatures or under specific kinetic temperature conditions. Furthermore, we determine the thermodynamic conditions within the experimental chamber.

MO 20.9 Thu 16:30 HS 3042

**A Time-dependent Perspective on Resonant Inelastic X-ray Scattering of Pyrazine** — ●ANTONIA FREIBERT<sup>1</sup>, DAVID MENDIVETAPIA<sup>2</sup>, NILS HUSE<sup>1</sup>, and ORIOL VENDRELL<sup>2</sup> — <sup>1</sup>University of Hamburg — <sup>2</sup>Heidelberg University

The developments of advanced x-ray sources have enabled the study of physical phenomena occurring on the intrinsic timescale of nuclear and electronic motion. One technique that gained considerable attention is resonant inelastic x-ray scattering (RIXS) and its extension into the ultrafast time domain. RIXS involves a coherent scattering process where the system is resonantly excited into short-lived core-hole states and subsequently decays back to the ground and valence excited states. This technique combines the element specificity of core-level spectroscopy with the ability to reach valence excited states across a wide spectral range and a spectral resolution that is not limited by the large core-hole lifetime broadening, making it a versatile and promising tool to study the local electronic structure in complex molecular systems.

I will present RIXS simulations of pyrazine at the nitrogen K-edge including wavepacket dynamics in both the valence- and core-excited state manifold. This allows to accurately depict dynamic processes occurring within the ultrashort core-hole lifetime and their manipulation through changes in excitation frequency. Additionally, I will discuss the impact of the spectral distribution of the incoming X-ray pulse and how it manifests in the resulting spectra striving for an optimal interplay between theory and experiment.

## MO 21: Ultrafast Dynamics II

Time: Thursday 14:30–16:45

Location: HS 3044

MO 21.1 Thu 14:30 HS 3044

**Reversible Switching based on intramolecular long-range Proton Transfer** — CHRIS REHHAGEN<sup>1</sup>, MIGUEL ARGÜELLO CORDERO<sup>1</sup>, FADHIL KAMOUNAH<sup>2</sup>, VERA DENEVA<sup>3</sup>, IVAN ANGELOV<sup>3</sup>, MARVIN KRUPP<sup>1</sup>, SØREN SVENNINGSEN<sup>2</sup>, MICHAEL PITTELKOW<sup>2</sup>, STEFAN LOCHBRUNNER<sup>1</sup>, and LIUDMIL ANTONOV<sup>3</sup> — <sup>1</sup>Institute for Physics and Department of Life, Light and Matter, University of Rostock, 18059 Rostock, Germany — <sup>2</sup>Department of Chemistry, University of Copenhagen, DK-2100 Copenhagen, Denmark — <sup>3</sup>Institute of Electronics, Bulgarian Academy of Sciences

A molecular switch is one of the essential elements in molecular electronics. The main requirement in the design of molecular switches is to provide a fast and clean interconversion between structurally different molecular (on- and off-) states. Currently existing molecular switches are either chemically or light driven. The proton transfer could be a new and attractive elementary switching process, because the change in the tautomeric state is always accomplished by a fast proton exchange between the reaction centers in the same molecule. The energy required for proton transfer is fairly low, which provides the opportunity for fueling with visible and near infrared light. Therefore, absorption and emission spectra of a new compound HQBT are investigated and further characterized with femtosecond transient absorption spectroscopy. The system consists of a benzothiazole rotor attached to a 7-hydroxy quinoline stator. A clean and ultrafast off-/on- switching, based on intramolecular long-range proton transfer, is observed in solution.

MO 21.2 Thu 14:45 HS 3044

**Unraveling the photochemistry of Ti<sup>IV</sup>Cp<sub>2</sub>(NCS)<sub>2</sub>** — JONAS SCHMIDT, LUIS IGNACIO DOMENIANNI, MARCEL LEUCHNER, ANDREAS GANSÄUER, and PETER VÖHRINGER — Rheinische Friedrich-Wilhelms-Universität, Bonn, Deutschland

Recently, we reported the observation of the entry event of Ti<sup>IV</sup>Cp<sub>2</sub>(NCS)<sub>2</sub> into a photocatalytic cycle in real-time.<sup>[1]</sup> In this study we were able to show the thermally activated delayed fluorescence of Ti<sup>IV</sup>Cp<sub>2</sub>(NCS)<sub>2</sub> after excitation with 450 nm light. Furthermore, we were able to observe the reductive quenching of the reactive triplet state with NPH<sub>3</sub> as an electron donor using time-resolved spectroscopy.

In a further step, we now want to investigate the following steps in the catalytic cycle. Therefore, we employ a substrate that acts as the amine electron donor to reduce the titanocene as well as the epoxide for radical arylation. To achieve this goal, we will utilize the long pump-probe delays of up to 300 μs of our synchronized double Ti:Sapphire regenerative amplifier setup for fs-UV/Vis-pump/mIR-probe spectroscopy. To aid the analysis of the spectroscopic findings, we will employ density functional theory as well as cooperate with synthetic chemists. From these experiments, we hope to gain further insight into the later mechanism of the photocatalytic cycle as it is still unclear if one or two equivalents of the substrate are involved.

**Literature:**

[1] J. Schmidt et al., *Angew. Chem. Int. Ed.* **2023**, *62*, e202307178.

MO 21.3 Thu 15:00 HS 3044

**Ultrafast Formation of Metallo-Nitrenes** — MARKUS BAUER, LUIS DOMENIANNI, and PETER VÖHRINGER — Clausius Institut für physikalische Chemie, Rheinische Friedrich-Wilhelms-Universität Bonn, Deutschland

Nitrenes have shown great potential as nitrogen-transfer reagents, owing to their high reactivity. For the same reason they are often found only as transient species and have to be prepared in-situ.<sup>[1]</sup> Therefore, to utilize their synthetic potential to the fullest, a firm understanding of the primary processes leading up to their formation is crucial.

Herein, a set of square-planar diazide complexes<sup>[2]</sup>, [M(N<sub>3</sub>)<sub>2</sub>(dppe)] (dppe=1,2-Bis(diphenylphosphino)ethane, M=Ni, Pd, Pt) was studied using ultrafast UV-pump-mIR-probe and time-resolved FTIR-spectroscopy. For all complexes nitrene formation is observed, with an increase of the quantum yield dependent on the mass of the metal. Additionally an intermediate triplet state preceding the nitrene formation could be identified.

Literature: [1] T. Schmidt-Räntsch, H. Verplanck, J. N. Lienert, S.

Demeshko, M. Otte, G. P. Van Trieste, K. A. Reid, J. H. Reibenspies, D. C. Powers, M. C. Holthausen, S. Schneider, *Angew. Chem. Int. Ed.* **2022**, *61*, e202115626. [2] Hennig, H., Hofbauer, K., Handke, K., Stich, R., 1997. *Angew. Chem. Int. Ed. Engl.* **36**, 408\*410.

MO 21.4 Thu 15:15 HS 3044

**Time-resolved insights in the fs-range on novel Fe(III)-complexes with functional modifications** — SAMIRA DABELSTEIN<sup>1</sup>, JAKOB STEUBE<sup>2</sup>, MIGUEL ANDRE ARGÜELLO CORDERO<sup>1</sup>, FRANZISKA FENNEL<sup>1</sup>, LENNART SCHMITZ<sup>2</sup>, MARVIN KRUPP<sup>1</sup>, CHRISTOPH VON DER OELSNITZ<sup>1</sup>, MATTHIAS BAUER<sup>2</sup>, and STEFAN LOCHBRUNNER<sup>1</sup> — <sup>1</sup>University of Rostock, Germany — <sup>2</sup>Paderborn University, Paderborn, Germany

Precious metal-based photosensitizers in photochemistry face scarcity and extraction challenges. There's a growing shift towards Earth-abundant metal alternatives, with iron being a promising, widely available, and cost-efficient candidate. The filled t<sub>2g</sub> orbitals in Fe(II)-complexes facilitate metal-to-ligand charge transfer (MLCT) upon optical excitation, while the ligand-to-metal charge transfer (LMCT) state is optimally achieved in Fe(III)-complexes owing to the partially filled t<sub>2g</sub> orbitals. In this study, we present a series of emitting Fe(III)-complexes modified with functional groups. These modified variants exhibit emission from LMCT states. The lifetime and energy of the LMCT state appear to be modulated by the attachment of diverse functional groups to the ligand's backbones. Our findings, obtained through time-resolved methods, specifically femtosecond transient absorption UV-Vis spectroscopy and streak camera measurements, are presented herein. These results are thoroughly examined in the context of electronic relaxation dynamics, shedding light on the intricate processes governing the observed phenomena.

MO 21.5 Thu 15:30 HS 3044

**Photophysical and photochemical investigations on a series of group VI carbonyl complexes with a meso-ionic carbene ligand** — DANIEL MARHÖFER<sup>1</sup>, PIT BODEN<sup>1</sup>, TOBIAS BENZ<sup>2</sup>, SOPHIE STEIGER<sup>1</sup>, BIPRAJIT SARKAR<sup>2</sup>, and GEREON NIEDNER-SCHATTEBURG<sup>1</sup> — <sup>1</sup>Department of Chemistry and Research Center Optimas, RPTU Kaiserslautern-Landau, Erwin-Schrödinger-Straße 52, 67663 Kaiserslautern, Germany. — <sup>2</sup>University of Stuttgart, Chair of Inorganic Coordination Chemistry, Institute of Inorganic Chemistry, Pfaffenwaldring 55, 70569 Stuttgart, Germany.

Photo-catalysis is a well-explored field in contemporary research. It enables reactions under comparably mild conditions, that wouldn't otherwise take place. A drawback of most photo-catalysts is the requirement for rare metals as late transition metals or even rare earth metals. In order to make this field of research more sustainable and economical, it is of particular interest to develop photo-catalysts, that contain only earth-abundant metals.

In this work we present a series of Cr(0), Mo(0) and W(0) carbonyl complexes that are able to stereo-selectively cleave a CO ligand under irradiation and recombine in the dark thereafter both in the solid phase and in solution. The photophysical behavior regarding emission and excited state vibrations as well as the respective excited state lifetimes was investigated on using luminescence spectroscopy and step-scan spectroscopy. The results were compared to previously reported, isomeric complexes and backed up by DFT calculations.

MO 21.6 Thu 15:45 HS 3044

**Symmetry-breaking charge transfer and intersystem crossing in copper phthalocyanine thin films** — ESTHER DEL PINO ROSENDO<sup>1</sup>, OKAN YILDIZ<sup>2</sup>, TOMASZ MARZSALEK<sup>2</sup>, CHARUSHEELA RAMANAN<sup>3</sup>, and PAUL W. M. BLOM<sup>2</sup> — <sup>1</sup>Johannes Gutenberg-Universität, Staudingerweg 7, 55128 Mainz — <sup>2</sup>Max Planck Institut für Polymerforschung, Ackermannweg 10 55128 Mainz — <sup>3</sup>Vrije Universiteit Amsterdam, De Boelelaan 1105, 1081 HV Amsterdam

Intermolecular interactions in π-stacked chromophores strongly influence their photophysical properties, and thereby also their function in photonic applications. Mixed electronic and vibrational coupling interactions lead to complex potential energy landscapes with competitive photophysical pathways. The photoexcited dynamics of the small molecule semiconductor CuPc are characterized in solution as well as in thin film. In the thin film case, the material can organize in two

different  $\pi$ -stacked architectures,  $\alpha$ -CuPc and  $\beta$ -CuPc. In solution, CuPc undergoes ultrafast ISC to the triplet excited state. In the solid state, both  $\alpha$ -CuPc and  $\beta$ -CuPc morphologies exhibit a mixing between Frenkel and charge-transfer excitons. We find that this mixing influences the photophysical properties differently, based on morphology.  $\alpha$ -CuPc demonstrates symmetry-breaking charge transfer, which furthermore depends on excitation wavelength. This mechanism is not observed in  $\beta$ -CuPc. These results elucidate how molecular organization mediates the balance of competitive photoexcited decay mechanisms in organic semiconductors.

MO 21.7 Thu 16:00 HS 3044

**Insights into exciton coupling of RNA-templated merocyanine dimer through higher-order transient absorption spectroscopy** — ●AJAY JAYACHANDRAN<sup>1</sup>, JULIA DIETZSCH<sup>2</sup>, STEFAN MÜLLER<sup>1</sup>, CLAUDIA HÖBARTNER<sup>2</sup>, and TOBIAS BRIXNER<sup>1</sup> — <sup>1</sup>Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — <sup>2</sup>Institut für Organische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany

The synthesis of oligonucleotide scaffolds to arrange chromophores into molecular aggregates with control over the formation of their molecular excitonic states offers an exciting prospect for achieving programmable photophysics. Our study exploits the Watson-Crick base pairing interactions in a RNA double helix by incorporating a new barbituric acid merocyanine as a nucleobase surrogate via solid-phase synthesis [1]. We observe the formation of a non-fluorescent, short-lived H-aggregate.

In order to study the single- and multi-excitonic manifold of this aggregate and its associated dynamics, we use a technique that we recently developed which employs systemic intensity variation of the pump pulse in pump-probe spectroscopy to separate pure third-order and fifth-order nonlinear signals [2]. We apply this higher-order transient absorption spectroscopy for the first time on oligonucleotide scaffolds and find indications for ultrafast exciton-exciton annihilation in the H-type dimer system [1].

- [1] J. Dietzsch et al., *Chem. Commun.* **59**, 7395–7398 (2023).  
[2] P. Malý et al., *Nature* **616**, 280–287 (2023).

MO 21.8 Thu 16:15 HS 3044

**Exciton Dynamics Pathways in a Merocyanine Dye-based Artificial Light-Harvesting Antenna** — ●REBECCA FRÖHLICH<sup>1</sup>, AJAY JAYACHANDRAN<sup>1</sup>, ALEXANDER SCHULZ<sup>2</sup>, MATTHIAS STOLTE<sup>3</sup>, FRANK WÜRTHNER<sup>2,3</sup>, and TOBIAS BRIXNER<sup>1</sup> — <sup>1</sup>Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg — <sup>2</sup>Institut für Organische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg — <sup>3</sup>Center for

Nanosystems Chemistry (CNC), Universität Würzburg, Theodor-Boveri-Weg, 97074 Würzburg

Natural photosynthesis relies on light-harvesting systems with precisely arranged chromophore structures. We investigate the exciton dynamics in an artificial light-harvesting antenna comprised of four dipolar merocyanine chromophores, covering a broad range of the visible spectrum. The molecule shows a solvent-dependent folding-induced fluorescence enhancement previously described on a trimer structure [1]. Irrespective of excitation wavelength, the molecule fluoresces only from the lowest excitonic state, which is a sign for efficient energy transfer in the tetramer. To investigate the exciton transfer pathways we performed transient absorption measurements with selective excitation of the exciton bands and coherent 2D electronic spectroscopy. The data show downwards energy transfer via parallel energy transfer pathways. A solvent comparison reveals that the energy transfer times depend on the folding degree of the supramolecular structure.

- [1] A. Schulz and F. Würthner, *Angew. Chem. Int. Ed.*, **61**, e202114667 (2022)

MO 21.9 Thu 16:30 HS 3044

**Following the Chlorophyll Coupled Dynamics with High-Level Multireference X-ray Absorption Spectra** — ●LENA BÄUML and REGINA DE VIVIE-RIEDLE — Department of Chemistry, LMU Munich, Germany

The omnipresent natural pigment chlorophyll is crucial for photosynthetic light-harvesting. Its nonradiative relaxation from high-energy excited states to the ground state is still not fully understood.

In an already published study we could show that the  $Q_x$  and  $Q_y$  band are strongly coupled via internal vibrations by applying grid-based wavepacket quantum dynamics on representative XMS-CASPT2 potential energy surfaces<sup>[1]</sup>. We accounted for the coupled nuclear and electron dynamics using the NEMol ansatz developed in our group. Due to the strong coupling of the states we found the electronic coherence to be present during the entire simulation time. Transient X-ray absorption spectra (XAS) offer the possibility to resolve the ultrafast coupled dynamics experimentally. To predict the observed features we simulated XAS for the magnesium and nitrogen K-edge of chlorophyll *a* at the XMS-CASPT2 level of theory. We derived time-resolved XAS using a workflow to combine the static XAS with the coupled NEMol dynamics<sup>[2]</sup>. Explicitly accounting for the pump pulse we could follow the nuclear wavepacket dynamics as well as predict the influence of the electronic coherence on the XAS.

- [1] L. Bäuml et al., *Phys. Chem. Chem. Phys.* **24**, 27212 (2022).  
[2] L. Bäuml et al., *J. Phys. Chem. A*, **127**, 9787 (2023).

## MO 22: Poster: Molecules in Strong Fields

Time: Thursday 17:00–19:00

Location: Tent C

MO 22.1 Thu 17:00 Tent C

**Dissociation Dynamics of Diiodomethane following XUV-induced inner-shell ionisation** — ●F. TROST<sup>1</sup>, H. LINDENBLATT<sup>1</sup>, S. MEISTER<sup>1</sup>, K. SCHNORR<sup>1</sup>, S. AUGUSTIN<sup>1</sup>, G. SCHMID<sup>1</sup>, Y. LIU<sup>1</sup>, P. SCHOCH<sup>1</sup>, F. HOSSEINI<sup>2</sup>, M. ZMERLI<sup>2</sup>, M. BRAUNE<sup>5</sup>, M. KUHLMANN<sup>5</sup>, S. DÍAZ-TENDERO<sup>4</sup>, F. MARTÍN<sup>4</sup>, R. GUILLEMIN<sup>2</sup>, M.-N. PIANCASTELLI<sup>3</sup>, M. SIMON<sup>2</sup>, T. PFEIFER<sup>1</sup>, C. D. SCHRÖTER<sup>1</sup>, and R. MOSHAMMER<sup>1</sup> — <sup>1</sup>Max-Planck-Institut für Kernphysik, Heidelberg — <sup>2</sup>Sorbonne Université, Paris — <sup>3</sup>Uppsala Universitet — <sup>4</sup>Universidad Autónoma de Madrid — <sup>5</sup>DESY, Hamburg

Knowledge of de-excitation, charge redistribution and fragmentation of molecules upon XUV irradiation is essential for our understanding of light-matter interaction. Here, the sequential three-body fragmentation of diiodomethane (CH<sub>2</sub>I<sub>2</sub>) following 4d inner-shell ionisation of one iodine atom is presented. The data was obtained by a time-resolved XUV-XUV pump-probe measurement using the reaction microscope endstation at the free-electron laser FLASH2 at DESY. In the two-step dissociation process of the CH<sub>2</sub>I<sub>2</sub> molecule a rotating intermediary state is identified through time-resolved 3D momentum correlation of the fragments. These results are supported by classical as well as quantum-mechanical simulations.

MO 22.2 Thu 17:00 Tent C

**Energy and Charge Transfer in Xenon Dimers and**

**Trimers after XUV-photoionization at FLASH2** — ●HANNES LINDENBLATT<sup>1</sup>, KIRSTEN SCHNORR<sup>2</sup>, SVEN AUGUSTIN<sup>2</sup>, SEVERIN MEISTER<sup>1</sup>, FLORIAN TROST<sup>1</sup>, PATRIZIA SCHOCH<sup>1</sup>, GEORG SCHMID<sup>1</sup>, YIFAN LIU<sup>1</sup>, MARKUS BRAUNE<sup>3</sup>, MARION KUHLMANN<sup>3</sup>, ROLF TREUSCH<sup>3</sup>, CLAUS DIETER SCHRÖTER<sup>1</sup>, THOMAS PFEIFER<sup>1</sup>, and ROBERT MOSHAMMER<sup>1</sup> — <sup>1</sup>Max-Planck-Institut für Kernphysik, Heidelberg — <sup>2</sup>Paul Scherrer Institut, Villigen, Schweiz — <sup>3</sup>DESY, Hamburg

We investigated the relaxation dynamics of xenon dimers and trimers employing momentum coincidence spectroscopy at the reaction microscope beamline FL26 at FLASH2. Using the grazing incidence split, delay and focusing optics, an XUV-XUV pump-probe scheme was utilized to track dissociation dynamics. On the poster, we present the measurement scheme and time-resolved results for different fragmentation channels. For example, in dimer-fragmentation we identified the role of radiative electron-transfer, and for trimers the so-called electron transfer-mediated decay (ETMD3), where all three atoms are involved, was observed as function of time.

MO 22.3 Thu 17:00 Tent C

**Time-resolved Imaging of CH<sub>4</sub> Fragmentation in Strong Laser Fields** — ●WEIYU ZHANG, DAVID VACAS CHICHARRO, NIKOLAS RAPP, THOMAS PFEIFER, and ROBERT MOSHAMMER — Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg,

Deutschland

With a Reaction Microscope (ReMi) [1] the ionization and dissociation dynamics of methane in strong laser fields were studied in a series of pump-probe experiments. For the creation of temporally separated laser pulses and pulse shaping a spatial light modulator (SLM) was used. It allows control of laser pulses in terms of amplitude, polarization, and phase [2, 3]. By employing the SLM technique we successfully compressed the laser pulses to below 10 fs and achieved a precise control over the time-delay between the two pulses. Upon strong-field ionization the molecule undergoes fragmentation and Coulomb explosion (CE) [4] and the corresponding ionic fragments are collected with the ReMi. For example, in the case of CE the initial inter-nuclear distances can be determined via the measurement of final kinetic energies, and in pump-probe measurements the evolution of the molecular geometry is visualized as function of time. Selected results will be presented and discussed.

References:

- [1] J. Ullrich et al., 2003, Rep. Prog. Phys. 66, 1463-1545
- [2] Stefanie Kerbstadt, 2016, MA thesis. Universität Oldenburg
- [3] T Brixner and G Gerber, 2001, Opt. Lett. 26,557-559
- [4] Larsen, J et al., 1998, J. Chem. Phys. 109, 8857-8863

MO 22.4 Thu 17:00 Tent C

**Dynamics of Molecules in Intense Laser Fields Studied with a Reaction Microscope** — ●MARTÍN GARRO, ARNE SENFTLEBEN, and JOCHEN MIKOSCH — Institut für Physik, Universität Kassel, Heinrich-Plett-Straße 40, 34132 Kassel, Germany

Intense laser fields can drive coherent electronic dynamics in a molecule on a timescale faster than the optical cycle of the light field. Electron wavepackets released by field-assisted tunneling ionization can be accelerated and driven back to the ion core with substantial energy by the action of the oscillating laser field. Different attosecond processes may ensue upon return of the continuum wavepacket. Here we are particularly interested in the aspect of a laser-driven scattering experiment which leads to the diffraction of the wavepacket encoding structural information on the molecule.

A Reaction Microscope allows us to experimentally investigate the electron dynamics of molecules exposed to strong ionizing fields with coincidence detection of electron and ion momenta. On the one hand side, we will present our previous work on laser-driven rescattering with two different, simultaneously created electron wavepackets in the 1,3-butadiene molecule. We show that the return probability of the electron depends on the molecular frame and contains structural information of the ionized orbital. On the other hand we will discuss our progress towards recollision and diffraction experiments on chiral molecules at the University of Kassel.

MO 22.5 Thu 17:00 Tent C

**Electron-nuclear energy sharing through low-energy inelastic recollisions in dissociative multiphoton ionization of D<sub>2</sub>** — ●SEBASTIAN HELL<sup>1</sup>, GERHARD G. PAULUS<sup>1,2</sup>, and MATTHIAS KÜBEL<sup>1,2</sup> — <sup>1</sup>Institute for Optics and Quantum Electronics, Universität Jena, D-07743 Jena, Germany — <sup>2</sup>Helmholtz Institute Jena, D-07743 Jena, Germany

Inelastic electron recollisions are known to contribute to high (*i.e.* several eV) kinetic energy release (KER) ion emission from D<sub>2</sub> in strong laser fields. Low-KER ion emission (*i.e.* up to few eV), however, is typically understood to result from single or multiphoton absorption from the laser field. Using coincidence detection of the correlated final states of photoelectron and ionic fragments, we investigate the dissociative ionization of D<sub>2</sub> in an intense 515 nm laser field with a half-frequency perturbation. Our experimental results show unambiguous fingerprints of low-energy inelastic electron recollisions contributing to dissociation of D<sub>2</sub><sup>+</sup>, even at KER below 1 eV. Our observations provide evidence for vibrational excitation and dissociative electron capture following laser-driven electron collisions.

MO 22.6 Thu 17:00 Tent C

**Time-resolved measurement of laser-induced dissociation of the argon dimer cation with a two-color pump-probe scheme** — ●BO YING<sup>1,2,3</sup>, GIORGIO VISENTIN<sup>2,3</sup>, MATTHIAS KÜBEL<sup>1,2</sup>, STEPHAN FRITZSCHE<sup>2,3,4</sup>, and GERHARD G PAULUS<sup>1,2,3</sup> — <sup>1</sup>Institute of Optics and Quantum Electronics, Friedrich Schiller University Jena, Max-Wien-Platz 1, 07743 Jena, Germany — <sup>2</sup>Helmholtz Institute Jena, Fröbelstieg 3, 07743 Jena, Germany — <sup>3</sup>GSI Helmholtzzentrum für Schwerionenforschung GmbH, Planckstraße 1, 64291 Darmstadt, Germany — <sup>4</sup>Institute for Theoretical Physics, Friedrich Schiller Uni-

versity Jena, Max-Wien-Platz 1, 07743 Jena, Germany

We present the time-resolved measurements of the photodissociation dynamics in the argon dimer cation Ar<sub>2</sub><sup>+</sup>, utilizing a pump-probe technique involving two different colors. In the experiment, the first laser pulse dissociates the generated Ar<sub>2</sub><sup>+</sup> ions, and the consequent dynamics are probed by a time-delayed second pulse. In contrast to the previous measurements of the simplest molecular ion H<sub>2</sub><sup>+</sup>, the delay-dependent kinetic energy distribution of the fragments not only shows the breakage of Ar<sub>2</sub><sup>+</sup> on the femtosecond time scale, but also reveals some intriguing features. Calculations of the potential energy curves of Ar<sub>2</sub><sup>+</sup> and Ar<sub>2</sub> help to understand the origin of such features, which results from the inherent multielectron nature of these heavy ions. This capability paves the way to investigate the temporal dynamics of hitherto unexplored molecular ion targets.

MO 22.7 Thu 17:00 Tent C

**Studies on the construction of a neon recycling system for High Harmonic Generation** — ●RON DUCKE, MAXIMILIAN POL-LANKA, MAXIMILIAN FORSTER, and REINHARD KIENBERGER — Chair for Laser and X-Ray Physics E11, Technical University Munich, Germany

The poster deals with a current project in which a neon recycling system is being implemented in our attosecond laboratory. In that, high harmonics, which are high-energy ultrashort light pulses, are generated in a vacuum chamber. For this purpose, neon gas is fed into this chamber and a pulsed near-infrared laser is focused on it. This produces the aforementioned high harmonics. By superimposing these pulses with the near-infrared laser pulse, physical effects can be measured in the attosecond range. This measurement technique is known as "attosecond streaking". The consumption of the required neon gas is one of the main costs of operating the laboratory (ca. 40.000 € in 2022). Previously, the gas was released into the exhaust air after passing through the chamber. The current attempt is to implement a recirculation of the neon gas. My efforts are aimed at reducing neon consumption and making the laboratory more sustainable on the one hand, but also massively reducing the costs of running the laboratory on the other. The first step is to collect the gas from the chamber and analyze its composition. The required set-up includes three pumps, a gas sampling bag and a measuring chamber with a mass spectrometer.

MO 22.8 Thu 17:00 Tent C

**Attosecond Electron Dynamics of Surface-Oriented Iodomethane on Pt(111)** — ●SVEN-JOACHIM PAUL<sup>1</sup>, PASCAL SCIGALLA<sup>1</sup>, CHRISTIAN SCHRÖDER<sup>1</sup>, KONSTANTIN SEIDENFUS<sup>1</sup>, PETER FEULNER<sup>2</sup>, and REINHARD KIENBERGER<sup>1</sup> — <sup>1</sup>Chair for laser and x-ray physics, E11, Technische Universität München, Germany — <sup>2</sup>Surface and Interface Physics, E20, Technische Universität München, Germany

We report on attosecond streaking measurements of the electron photoemission process from Pt(111) surfaces covered in well-oriented iodomethane molecules. Attosecond streaking allows for the measurement of relative time delays in photoemission from two energetically different bound electronic states. For this experiment, the photoemission from the platinum valence band has been timed against the photoemission of the iodine 4d orbital in iodomethane. A routine has been developed to control the self-assembly surface orientation process of the adsorbed iodomethane molecules by varying the coverage of the Pt(111) crystal. Depending on the surface coverage, iodomethane aligns vertically or horizontally on the surface. This enables the study of photoemission dynamics through a well-oriented potential. Attosecond streaking has then been systematically performed for nine different surface coverages, most of them in the sub-monolayer regime. With that, first-ever insights into the influence of adsorbate potential orientation and density on photoemission delay were possible.

MO 22.9 Thu 17:00 Tent C

**Attosecond Chronoscopy of CO-Structures on Pt(111)** — ●KONSTANTIN SEIDENFUS, PASCAL SCIGALLA, SVEN-JOACHIM PAUL, and REINHARD KIENBERGER — Chair for Laser and X-Ray Physics, E11, Technische Universität München, Germany

This work investigates time delays occurring in the electron photoemission process of a Carbon-monoxide-covered Pt(111)-Surface, for which the attosecond streaking technique is used. The relative time delay in photoemission of multiple energetic differently bound electronic states can be measured with this technique. In this case, the platinum valence band is measured against the carbon valence band, at different

coverages. A routine for sub-monolayer accurate, reproducible surface coverages of CO is developed and tested with TPD. Carbon monoxide has previously been investigated in the gas phase, where it was measured against a helium reference. CO orients itself vertically on the

surface of the platinum crystal, whereas in gas the orientation is incoherent. This enables one to gain additional insight into the physical effect of an oriented potential on the photoemission process.

## MO 23: Poster: Chirality

Time: Thursday 17:00–19:00

Location: Tent C

MO 23.1 Thu 17:00 Tent C

**Towards perfect enantiomer-specific state transfer of chiral molecules** — ●ELAHE ABDIHA, JUHYEON LEE, JOHANNES BISCHOFF, DANIEL FONTOURA BARROSO, BORIS SARTAKOV, GERARD MEIJER, and SANDRA EIBENBERGER-ARIAS — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany

Chiral molecules are important in many chemical and biological processes and are also at the heart of some fundamental physics questions. Recently, enantiomer-specific state transfer (ESST) was experimentally demonstrated [1,2]. Here, the application of three mutually orthogonally polarized microwave fields yields enantiomer-specific population control in a chosen quantum state that is part of a triad of rotational states. I will present our recent work on ESST, where we largely overcome the previous limitation due to initial thermal population by combining ESST with optical methods [3]. By depleting the target state using resonant UV light prior to ESST we achieve state-specific enantiomeric enrichment in the order of 50%. Importantly, we quantitatively study ESST, explicitly including the role of spatial degeneracy. I will also discuss our recent study on the influence of microwave pulse conditions on ESST [4]. Extensions to our scheme will allow to create a molecular beam with an enantiomer-pure rotational level, holding great prospects for future spectroscopic and scattering studies.

[1] Eibenberger et al, PRL 118, 123002 (2017) [2] Pérez et al, Angew. Chem. Int. Ed. 56, 12512 (2017) [3] Lee et al, PRL 128, 173001 (2022) [4] Lee et al, arxiv: 2310.11120 (2023)

MO 23.2 Thu 17:00 Tent C

**Intensity dependence of PECD using near-ultraviolet femtosecond laser pulses** — ●SUDHEENDRAN VASUDEVAN, HAN-GYEOL LEE, ERIC KUTSCHER, SIMON T. RANECKY, NICOLAS LADDA, TONIO ROSEN, SAGNIK DAS, TILL STEHLING, JAYANTA GHOSH, ANTON N. ARTEMYEV, HENDRIKE BRAUN, ARNE SENFTLEBEN, PHILIPP V. DEMEKHIN, and THOMAS BAUMERT — Institut für Physik, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel

Exploiting an electric dipole effect in ionization, photoelectron circular dichroism (PECD), i.e., an asymmetry in the photoelectron angular distribution along the light propagation, is a highly sensitive enantioselective spectroscopy for studying chiral molecules in the gas phase using either single-photon [1] or multiphoton ionization [2]. In the latter case, resonance-enhanced multiphoton ionization (REMPI) gives access to intermediate electronic states [3]. The PECD sensitivity opens the door to study control of the coupled electron and nuclear motion in enantiomers. A prerequisite is the detailed understanding of PECD in REMPI schemes. In this contribution, we demonstrate the intensity-dependence of PECD on fenchone via 2+1 REMPI using 30 fs near-ultraviolet laser pulses centered at 396 nm. We find that the magnitude of LPECD decreases with the linear increase in the FWHM of the photoelectron spectra.

### References

- [1] Böwering, N. et al. Phys. Rev. Lett. 86, 1187 (2001).  
 [2] Lux, C. et al. Angew. Chem. Int. Ed. 51, 5001\*5005 (2012).  
 [3] Lee, H.-g. et al. PCCP 24, 27483\*27494 (2022).

MO 23.3 Thu 17:00 Tent C

**Photoelectron circular dichroism after O1s ionization in sec-butyl trimethylsilylether** — ●CHRISTINA ZINDEL, EMILIA HEIKURA, CATMARNA KÜSTNER-WETEKAM, LUTZ MARDER, JOHANNES VIEHMANN, DENIS KARGIN, RUDOLF PIETSCHNIG, ANDREAS HANS, and ARNO EHRESMANN — Institut für Physik and CINSaT, University of Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany

Following ionization with circularly polarized radiation, randomly oriented enantiomers show a forward-backwards asymmetry in their photoelectron angular distribution (PAD) with respect to the light's propagation direction. This effect is described as photoelectron circular

dichroism (PECD) — a powerful tool to examine the molecular chirality of gas-phase samples. In our studies, we use a velocity map imaging spectrometer (VMI) along with synchrotron radiation to investigate the chiral molecule sec-butyl trimethylsilylether ( $C_4H_9-O-Si_3(CH_3)_3$ ). When varying the distance between the stereocenter and the silicon atom by subsequently inserting  $CH_2$  groups in between, it is of interest whether the PECD magnitude changes after ionization of the O1s orbital, because although the length of the molecule varies in this case, for oxygen, its geometry with respect to the stereocenter remains the same.

MO 23.4 Thu 17:00 Tent C

**Coherent Control of Photoelectron Circular Dichroism using Two-color Laser Pulses** — ●TILL STEHLING, HANGYEOL LEE, SUDHEENDRAN VASUDEVAN, SIMON RANECKY, NICOLAS LADDA, TONIO ROSEN, SAGNIK DAS, JAYANTA GHOSH, HENDRIKE BRAUN, ARNE SENFTLEBEN, and THOMAS BAUMERT — Universität Kassel Institut für Physik Heinrich-Plett-Str. 40 34132 Kassel/Germany

We study two-pathway interference in multiphoton ionization of fenchone. By employing two pulses at 380 nm and 440 nm, two distinct (2+1) resonance-enhanced multiphoton ionization (REMPI) pathways via the 3s and the 3p band of fenchone are simultaneously driven. The ionization out of these states can result in the same final continuum state, such that the which-way-information is unknown, and interference can be observed. Photoelectron energy-spectra (PES) show three distinct peaks attributed to different ionization pathways. One of which exhibits a dependence on the temporal delay between the two pulses. These preliminary results may indicate a successful implementation of two-pathway interference during the ionization of fenchone. Future experiments will be concerned with the influence of such interference effects on the photoelectron-circular-dichroism (PECD) of fenchone as they present a possible method to implement control schemes in chiral photophysics.

MO 23.5 Thu 17:00 Tent C

**Experimental setup to study enhancement of circular dichroism in ion yield of 3-methyl cyclopentanone via tailored femtosecond laser pulses** — ●SAGNIK DAS, JAYANTA GHOSH, SUDHEENDRAN VASUDEVAN, HANGYEOL LEE, NICOLAS LADDA, SIMON RANECKY, TONIO ROSEN, TILL STEHLING, FABIAN WESTMEIER, ARNE SENFTLEBEN, THOMAS BAUMERT, and HENDRIKE BRAUN — Institut für Physik, Universität Kassel

Femtosecond laser pulses were employed to ionize 3-methyl cyclopentanone (3-MCP) in the home-built time-of-flight mass spectrometer. A 'twin peak' measurement technique [1] was utilized, to obtain ion yields from left and right circularly polarised laser pulses in the same laser shot. Such a technique reduces the statistical error of measurement and allows for simultaneous chirp compensation in both left and right-handed light. Circular dichroism in ion yield (CDIY) is the difference in ion yield from the left and right circularly polarised light for a given enantiomer [2,3]. Switching the enantiomer, the sign of CDIY is flipped. At the wavelengths where we performed the experiments, a 1+1+1 resonance-enhanced multiphoton ionization in 3-MCP takes place via the  $\pi^* \leftarrow n$  band, which exhibits a strong circular dichroism. It was found that linear chirp enhances the CDIY. We systematically investigated the reason behind the enhancement of CDIY for chirped pulses.

- [1] T. Ring et al., Rev. Sci. Instrum., 92, 033001, 2021  
 [2] U. Boesl and A. Bornschlegl, ChemPhysChem, 7, 2085, 2006  
 [3] H. G. Breunig et al., ChemPhysChem, 10, 1199, 2009

MO 23.6 Thu 17:00 Tent C

**Time-Resolved Circular Dichroism Spectroscopy with Ultrafast Broadband Circularly Polarized Laser Pulses** — ●KARINA HEILMEIER<sup>1</sup>, EMELY FREYTAG<sup>2</sup>, CHRISTINA KAUFMANN<sup>2</sup>, FRANK WÜRTHNER<sup>2</sup>, CHRISTOPH LAMBERT<sup>2</sup>, and TOBIAS BRIXNER<sup>1</sup> —

<sup>1</sup>Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — <sup>2</sup>Institut für Organische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany

Chiral exciton states that have a small dipole strength have only weak contributions in linear absorption spectroscopy. However, such states can be well resolved with circular dichroism spectroscopy. Analogously, time-resolved circular dichroism (TRCD) spectroscopy may give information on the ultrafast dynamics of these exciton states which cannot be resolved with non-chiral transient absorption spectroscopy.[1]

Here, we report a TRCD setup based on the pump-probe approach employing ultrafast broadband white-light probe pulses with left- and right-handed circular polarization. The circular polarization is induced by a polarization grating. In TRCD one measures the difference in absorption of left- and right-circularly polarized light of the excited and non-excited states. To investigate the dynamics of chiral excitons, measurements were carried out on a chiral squaraine dimer and a chiral perylene bisimide dimer, both exhibiting excitonic coupling.

[1] L. Röss, P. Malý, J. B. Landgraf, D. Lindorfer, M. Hofer, J. Selby, C. Lambert, T. Renger, T. Brixner, *Chem. Sci.* **14**, 9328 (2023).

MO 23.7 Thu 17:00 Tent C

**Electron correlation in circular dichroism and chirality-induced spin selectivity** — ●RAOUL M. M. EBELING<sup>1</sup>, MAURICE BÉRINGUIER<sup>1</sup>, VLADIMIRO MUJICA<sup>2</sup>, DANIEL M. REICH<sup>1</sup>, and CHRISTIANE P. KOCH<sup>1</sup> — <sup>1</sup>Freie Universität Berlin, Berlin, Germany — <sup>2</sup>Arizona State University, Arizona, United States of America

We study two phenomena related to the interaction of chiral molecules with circularly polarized light, absorption circular dichroism (CD), and chirality-induced spin selectivity (CISS). We investigate both phenomena in chiral hydrogen and chiral helium, two model systems into which we introduce chirality via an artificial chiral potential. The chiral potential is constructed from a superposition of spherical harmonics and it can be interpreted as a way to mimic the chiral environment of a real molecule. Alternatively, our chiral hydrogen and chiral helium models could even be experimentally realized by placing the atoms in a setup involving several electric fields. Quantifying the chirality of the potential with a suitable measure, we find that it can be tuned via the coefficients in the spherical harmonics expansion. We investigate the influence of the strength of the chiral potential, the strength of the spin-orbit coupling, and the strength of the electron-electron interaction on both CD and CISS.

MO 23.8 Thu 17:00 Tent C

**Towards Comparing the PECD Effect in Closed-Shell and Open-Shell Chiral Molecules** — ●VIKTORIA K. BRANDT, ANDRÉ FIELICKE, GERARD MEIJER, and MALLORY GREEN — Department of Molecular Physics, Fritz Haber Institute of the MPG, Berlin

Photoelectron Circular Dichroism (PECD) is a chiral optical effect that manifests in the angle-dependent photoemission of an electron upon irradiation of a chiral molecule by circularly polarized light. A PECD effect in anions was observed for the first time in 2021, showing that forces other than long-range coulombic interactions can yield a measurable PECD effect. To further explore the effect of short-range forces, as well as the universal electron dynamics that govern PECD, 1-phenylethanol was investigated as a closed-shell deprotonated anion, a dehydrogenated, neutral radical and a closed-shell neutral. A photoelectron spectrum of the anion showed the presence of only two deprotonated tautomers. However, the landscape of radical isomers, formed by reaction with fluorine atoms, is calculated to be complicated and includes a large number of achiral species, making a comparison to the closed-shell species difficult. In the energy region that overlaps with predicted spectra of chiral isomers, photoionization of the radical enantiomers shows a reversal of a small PECD effect. However, statistics of this effect are low and confirmation of this effect is still inconclusive. At this point, only the closed-shell neutral has a confirmed PECD measured at around 5%. Further work will focus on obtaining a PECD of the closed-shell anion and identifying new molecular targets with less complicated isomeric landscapes.

MO 23.9 Thu 17:00 Tent C

**Raman Optical Activity Of Glucose** — ●KLAUS HOFMANN, LUISA MARTIN, and INGO FISCHER — Universität Würzburg, 97074 Würzburg, Germany

Raman Optical Activity (ROA) is a type of vibrational circular dichroism: chiral samples show different Raman intensities when utilizing circular polarized light. The ROA signal is very sensitive to the molecular geometry and environment of the sample, but also exhibits high levels of noise and false signals, since the intensity difference is roughly 0.1% of the corresponding Raman peak.

Using a custom built cost-efficient ROA spectrometer, the signal of the two glucose enantiomers in aqueous solution was recorded, with exposure times of one week each. Python was used to automate the setup modulation, data acquisition and post-processing. The result agrees with literature and matches spectra acquired on commercially available spectrometer.

## MO 24: Poster: Experimental Techniques

Time: Thursday 17:00–19:00

Location: Tent C

MO 24.1 Thu 17:00 Tent C

**Ionic Liquids with Three-Valent Cations: Spectroscopic Studies Towards Their Use as Reaction Media** — ●MAX SCHAHDENFROH, CARINA ALLACHER, SELINA REIGL, MANUEL ROTHE, WERNER KUNZ, and PATRICK NÜRNBERGER — Institut für Physikalische und Theoretische Chemie, Universität Regensburg, 93040 Regensburg

Ionic liquids (ILs) are defined as salts with a melting point below 100 °C [1]. ILs are multifaceted systems with highly customizable physical and chemical properties, and thus offer a plethora of applications. While classical ILs are often toxic and environmentally harmful, a new class of ILs was introduced in 2018 [2], which better satisfies the principles of Green Chemistry. These compounds rely on the **CO**ncept of **M**elting **P**oint **L**owering due to **E**thoxylation (**COMPLET**). More recently, this concept could be extended to three-valent cations [3].

Here, we present spectroscopic studies of ionic liquids containing  $\text{Eu}^{3+}$ ,  $\text{Y}^{3+}$ , and  $\text{La}^{3+}$  cations. Combining excitation and emission spectroscopy, an energy transfer from the anionic ligand to the metal upon photoexcitation can be corroborated. This ligand-metal interaction may be exploited in photochemical organic synthesis by utilization of these ILs as solvent environment. The approach to combine catalyst and solvent in one compound could be beneficial for confinement-controlled synthesis and an alternative to conventional photocatalysts.

[1] H. Weingärtner, *Angew. Chem. Int. Ed.* **47**, 654–670 (2008).

[2] E. Müller *et al.*, *J. Mol. Liq.* **251**, 61–69 (2018).

[3] M. Rothe *et al.*, *Chem. Eur. J.*, **27**, 13052–13058 (2021).

MO 24.2 Thu 17:00 Tent C

**Ultrafast dynamics of Metanil Yellow studied by time-resolved transient absorption and XUV photoelectron spectroscopies in solution** — ●ALINA KHODKO<sup>1,4</sup>, MATTHEW MGBUKWU<sup>3</sup>, CAMILO GRANADOS<sup>1,3</sup>, EVGENII TITOV<sup>2</sup>, NATALIA KACHALOVA<sup>4,5</sup>, VALERII VOITSEKHOVYCH<sup>4</sup>, IGOR DMYTRUK<sup>4,6</sup>, STEFAN HAACKE<sup>3</sup>, OLEG KORNILOV<sup>1</sup>, and JÉRÉMIE LÉONARD<sup>3</sup> — <sup>1</sup>Max Born Institute, Berlin, Germany — <sup>2</sup>Institute of Chemistry, University of Potsdam, Germany — <sup>3</sup>Institut de Physique et Chimie des Matériaux de Strasbourg, Université de Strasbourg, France — <sup>4</sup>Institute of Physics, Kyiv, Ukraine — <sup>5</sup>L.M. Litvinenko Institute of Physical and Organic Chemistry and Coal Chemistry, Kyiv, Ukraine — <sup>6</sup>Taras Shevchenko National University of Kyiv, Ukraine

The excited-state dynamics of the Metanil Yellow (MY) were studied by ultrafast transient absorption (TA) spectroscopy and state-of-the-art XUV time-resolved photoelectron spectroscopy (TRPES). Here the TA experiments were carried out with two excitation wavelengths,  $\lambda=370$  nm and  $\lambda=490$  nm, to investigate the non-hydrated and hydrated forms of the molecule and reveal differences in their dynamics in two solvents: water and ethanol. In TRPES experiments the dynamics were studied in water solution, using a  $\lambda=400$  nm pump, thus exciting both forms. In general, the timescales from the TRPES experiments are in good agreement with the results from the TA measurements. Based on quantum chemical calculations, the dynamics are tentatively

assigned to the S2\*S1 conversion followed by relaxation to a long-lived state, the nature of which remains to be confirmed.

MO 24.3 Thu 17:00 Tent C

**A through-flow cell for highly-resolved Stark effect measurements of Rydberg states in thermal nitric oxide** —

•FLORIAN ANSCHÜTZ<sup>1</sup>, ETTORE EDER<sup>1</sup>, FABIAN MUNKES<sup>1</sup>, ALEXANDER TRACHTMANN<sup>1</sup>, PHILIPP HENGEL<sup>2</sup>, YANNICK SCHELLANDER<sup>3</sup>, PATRICK SCHALBERGER<sup>3</sup>, MATTHEW RAYMENT<sup>4</sup>, STEPHEN HOGAN<sup>4</sup>, NORBERT FRUEHAUF<sup>3</sup>, JENS ANDERS<sup>2</sup>, ROBERT LÖW<sup>1</sup>, TILMAN PFAU<sup>1</sup>, and HARALD KÜBLER<sup>1</sup> — <sup>1</sup>5. Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart — <sup>2</sup>Institut für Intelligente Sensorik und Theoretische Elektrotechnik, Universität Stuttgart, Pfaffenwaldring 47, 70569 Stuttgart — <sup>3</sup>Institut für Großflächige Mikroelektronik, Universität Stuttgart, Allmandring 3b, 70569 Stuttgart — <sup>4</sup>Department of Physics and Astronomy, University College London, Gower Street, London WC1E 6BT, UK

We show the setup of a glass cell equipped for the electric readout of Rydberg states in nitric oxide (NO) at room temperature. The field distribution is discussed and our results on both the Stark effect and the collisional shift and broadening of Rydberg states in NO is presented.

MO 24.4 Thu 17:00 Tent C

**Near field spectroscopy of molecular aggregates with topological phases** —

•SIDHARTHA NAYAK, ARITRA MISHRA, and ALEXANDER EISFELD — Max Planck Institute for the Physics of Complex Systems, Dresden, Germany

In this theoretical study, we focus on delocalized electronic excitonic states in molecular aggregates, particularly those exhibiting topological phases. It has been shown that a two-dimensional molecular aggregate, composed of two different sublattices and complex transition dipole moments, possesses topological edge states [1]. However, these states are predominantly 'dark' in traditional far-field absorption spectra. We consider a typical scattering scanning optical near-field microscopy (s-SNOM) setup, where the aggregate interacts with the near field stemming from a metallic tip [2, 3]. With the help of s-SNOM, we can not only excite these dark states but also record spatially resolved absorption spectra, revealing clear signatures of both excitonic edge states and bulk states.

[1] J.Y. Zhou, S. K. Saikin, N.Y. Yao and A. Aspuru-Guzik, *Nature materials* 13, 1026-1032 (2014)

[2] X. Gao and A. Einfeld, *J. Phys. Chem. Lett.* 9, 6003 (2018)

[3] S. Nayak, F. Zheng and A. Einfeld, *J. Chem. Phys.* 155, 134701 (2021)

MO 24.5 Thu 17:00 Tent C

**Installation of a hollow cathode molecular ion source** —

•JULIAN RIMATZKI, SIMON REINWARDT, and MICHAEL MARTINS —

Universität Hamburg, Hamburg, Deutschland

A research plan has been devised for studying inner-shell photoionization of molecular ions with a small thermal energy at the photon-ion spectrometer at PETRA III (PIPE)[1] of DESY in Hamburg. First result to characterise the plasma of a hollow cathode ion source will be shown. Such a characterisation can be obtained by studying COH<sup>+</sup> and HCO<sup>+</sup> isomers in the gas phase using soft X-Ray spectroscopy. The challenge of forming ions in a higher geometrical state like the COH<sup>+</sup> can be solved by using a ion source with a plasma temperature below 18.000 K [2].

We are undergoing the construction and parametrization of our new Ion source at the PIPE-setup. We will present first results to form homogeneous ion beams with only one constitutional isomer.

[1] Schippers, S. et al., X-ray Spec., 2020, **49**, 11.

[2] Nobes, R. H., Radom, L., Chem. Phys., 1981, **60**, 1.

MO 24.6 Thu 17:00 Tent C

**Characterization of a simple supersonic expansion source for**

**small molecular ions** — •LUKAS BERGER<sup>1</sup>, AIGARS ZNOTINS<sup>1</sup>, FLORIAN GRUSSIE<sup>1</sup>, DAMIAN MÜLL<sup>1</sup>, FELIX NÜSSLEIN<sup>1</sup>, ARNAUD DOCHAIN<sup>2</sup>, JOFFREY FRÉREUX<sup>2</sup>, XAVIER URBAIN<sup>2</sup>, and HOLGER KRECKEL<sup>1</sup> — <sup>1</sup>Max-Planck-Institut für Kernphysik, 69117 Heidelberg, Germany — <sup>2</sup>Institute of Condensed Matter and Nanosciences, Université Catholique de Louvain, Louvain-la-Neuve, B-1248 Belgium

The Cryogenic Storage Ring (CSR) at the Max Planck Institute for Nuclear Physics in Heidelberg is an electrostatic storage facility with a circumference of approx. 35 m that can be cooled to cryogenic temperatures (~4 K) by a closed-cycle liquid helium unit. The blackbody radiation field is strongly reduced compared to room-temperature experiments, and small infrared-active molecular ions will cool to their lowest rotational states within minutes in this environment, allowing for experiments with ions in defined quantum states. However, some astrophysically relevant molecular ions do not cool on accessible timescales, owing to the lack of a permanent dipole moment. To address this issue, we have developed a simple supersonic expansion ion source, based on a commercial pulsed valve and static discharge voltages. We have characterized the source performance with N<sub>2</sub>O<sup>+</sup> ions, conducting experiments at the STARGATE setup at UCLouvain (Louvain-la-Neuve, Belgium), which resulted in internal temperatures between 40 K and 200 K. The ion source was then integrated into the ion source platform of the CSR to deliver cold ions for merged beams experiments. The design and performance will be presented.

MO 24.7 Thu 17:00 Tent C

**Resonance Energy Transfer Involving Chiral Molecules and Macroscopic Environment** —

•JANINE C. FRANZ<sup>1,2</sup>, STEFAN YOSHI BUHMANN<sup>1</sup>, and A. SALAM<sup>3</sup> — <sup>1</sup>University Kassel, Germany — <sup>2</sup>University Freiburg, Germany — <sup>3</sup>Wake Forest University, Winston-Salem, USA

Resonance energy transfer between chiral molecules can be used to discriminate between different enantiomers. The transfer rate between chiral molecules consists of nondiscriminatory and discriminatory parts. We show that their ratio is usually larger in the far zone regime and that the degree of discrimination can be modified when considering a surrounding medium. We highlight the importance of local field effects on the degree of discrimination, predict the optimum dielectric medium for general identical chiral molecules for discrimination, and show that exotic media can even invert the discriminatory effect. When considering a chiral medium, the environment can actively participate in the discrimination, but the local-field corrections become more involved. We show that the local-field corrections in a chiral medium then lead to a surprising effect in the discrimination.

MO 24.8 Thu 17:00 Tent C

**Relayed hyperpolarization for zero- to ultralow-field nuclear magnetic resonance** —

•ERIK VAN DYKE<sup>1,2,3</sup>, JAMES EILLS<sup>1,2,3,4</sup>, ROMAN PICAZO-FRUTOS<sup>1,2,3</sup>, KIRILL SHEBERSTOV<sup>1,2,3,5</sup>, YINAN HU<sup>1,2,3,6</sup>, DMITRY BUDKER<sup>1,2,3,7</sup>, and DANILA BARSKIY<sup>1,2,3</sup> — <sup>1</sup>Helmholtz Institute Mainz, Mainz, Germany — <sup>2</sup>Johannes Gutenberg University, Mainz, Germany — <sup>3</sup>GSI Helmholtz Center for Heavy Ion Research, Darmstadt, Germany — <sup>4</sup>Institute for Bioengineering of Catalonia, Barcelona, Spain — <sup>5</sup>Ecole normale supérieure, Paris, France — <sup>6</sup>Chinese Academy of Sciences, Beijing, China — <sup>7</sup>University of California at Berkeley, Berkeley, USA

Zero- to ultralow-field nuclear magnetic resonance (ZULF NMR) provides rich spectroscopic information in the absence of large magnetic fields. Still, signal acquisition requires a bulk magnetic moment for detection. We demonstrate that the parahydrogen-based Signal Amplification by Reversible exchange-Relay method (SABRE-Relay) can be used to generate hyperpolarized analytes for ZULF-NMR by observing J-spectra of methanol and ethanol (from vodka) at natural <sup>13</sup>C isotopic abundance. The magnetic-field dependence of SABRE efficiency is also shown.

## MO 25: Novel Experimental Approaches

Time: Friday 11:00–12:45

Location: HS 3044

MO 25.1 Fri 11:00 HS 3044

**Cryo-cooled beams of "small" macromolecules** — ●JINGXUAN HE<sup>1,2,3</sup>, LENA WORBS<sup>1,2</sup>, SURYA KIRAN PERAVALI<sup>1,4</sup>, ARMANDO D. ESTILLORE<sup>1</sup>, AMIT K. SAMANTA<sup>1,3</sup>, and JOCHEN KÜPPER<sup>1,2,3</sup> — <sup>1</sup>Center for Free-Electron Laser Science (CFEL), Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany — <sup>2</sup>Department of Physics, Universität Hamburg, Germany — <sup>3</sup>Center for Ultrafast Imaging (CUI), Universität Hamburg, Germany — <sup>4</sup>Fakultät für Maschinenbau, Helmut-Schmidt-Universität, Germany

We have demonstrated the preparation of cold and controlled beams of nanoparticles and macromolecules that are desired for x-ray single particle diffractive imaging (SPI) using the buffer-gas cell (BGC) cooling and aerodynamic focusing techniques [1-2]. The cooling and control techniques we developed for SPI can be extended to experiments to study the electron dynamics in complex biomolecules on the few femtosecond timescale, such as charge and energy transfer following electronic excitation, where the details have not been revealed so far [3]. We present an approach towards investigating the time-resolved ultrafast dynamics in proteins with UV/VIS ultrashort-pulse lasers. The photoexcitation-induced energy transfer, for instance, can be studied by photofragmentation of cryogenically-cooled proteins with time-of-flight mass spectrometry and velocity-map-imaging.

- [1] A. K. Samanta, et al., *Structural dynamics* **7**, 024304 (2020)
- [2] L. Worbs, et al., *In preparation*, (2024)
- [3] H. Duan, et al., *PNAS* **114**, 8493 (2017)

MO 25.2 Fri 11:15 HS 3044

**Characterizing temperature, charging and adsorption dynamics of single nanoparticles** — ●BJÖRN BASTIAN, SOPHIA LEIPPE, KLEOPATRA PAPAGRIGORIOU, and KNUT ASMIS — Wilhelm-Ostwald-Institut, Linnéstraße 2, D-04103 Leipzig

Single nanoparticle (NP) techniques allow to probe intrinsic properties of nanoparticles, but typically rely on surface deposition. Instead, we develop the analysis of single NPs in the gas phase using a cryogenic radio-frequency ion trap and UV/Vis or IR action spectroscopy. Absorption is indirectly monitored using NP mass spectrometry (NPMS): the produced heat causes the loss of messenger atoms or molecules that are adsorbed to the particle surface. Here, we present current progress on controlling and characterizing the charge state, temperature and surface coverage of single trapped NPs that will ease the implementation and quantitative analysis of future experiments.

Inducing charge changes is crucial for absolute mass determination and facilitates control in long experiments ( $\sim$ days). Using a filament to emit electrons for electron attachment or charge transfer — mediated by different collision gases at different pressures — we demonstrate full control of the charge state of positively charged silica NPs.

Characterizing binding sites and energies is an important goal for NP characterization and essential for quantitative action spectroscopy. Extensive adsorption measurements on silica NPs are presented and we demonstrate *in situ* fluorescence thermometry for semiconductor quantum dots. We will report on the latest progress to simultaneously measure temperature and adsorption on single fluorescent nanoplatelets.

MO 25.3 Fri 11:30 HS 3044

**Laser-induced alignment of macromolecules and nanoparticles** — ●LUKAS VINCENT HAAS<sup>1,2,3</sup>, XUEMEI CHENG<sup>1</sup>, MUHAMED AMIN<sup>1</sup>, AMIT KUMAR SAMANTA<sup>1,2,3</sup>, and JOCHEN KÜPPER<sup>1,2,3</sup> — <sup>1</sup>Center for Free-Electron Laser Science (CFEL), Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany — <sup>2</sup>Department of Physics, Universität Hamburg, Germany — <sup>3</sup>Center for Ultrafast Imaging (CUI), Universität Hamburg, Germany

X-ray free-electron lasers (XFELs) promise to enable the diffractive imaging of single molecules and nanoparticles, while image reconstruction remains a major bottleneck in achieving atomic spatial resolution [1]. Laser-induced alignment of nanoparticles and macromolecules has the potential to improve the achievable resolution by reducing the complexity of the diffraction volume search space and push it toward the atomic scale [2]. Here, we will present quantitative computational modeling of nanoparticle alignment using classical mechanics and electrodynamics [3] and first experimental evidence of laser-induced alignment of tobacco mosaic virus (TMV) in a setup that is applicable to XFEL experiments [4]. Furthermore, a recently conducted XFEL ex-

periment provides first results on diffractive imaging of laser-aligned TMV. Comparing computational and experimental results, we can conclude that a high degree of alignment is achieved for TMV in our experiments.

- [1] K. Ayyer, et al., *Optica* **8**(1) (2021)
- [2] J. C. H. Spence, et al., *Phys. Rev. Lett.* **92**, 198102 (2004)
- [3] M. Amin, et al., arXiv:2306.05870 [physics], (2023)

MO 25.4 Fri 11:45 HS 3044

**Charge density model for the interaction of molecules with vortex beams** — MIKHAIL MASLOV<sup>1</sup>, GEORGIOS M. KOUTENTAKIS<sup>1</sup>, ●MATEJA HRAST<sup>1</sup>, OLIVER H. HECKL<sup>2</sup>, and MIKHAIL LEMESHKO<sup>1</sup> — <sup>1</sup>Institute of Science and Technology Austria (ISTA), Klosterneuburg, Austria — <sup>2</sup>Christian Doppler Laboratory for Mid-IR Spectroscopy and Semiconductor Optics, Faculty Center for Nano Structure Research, Faculty of Physics, University of Vienna, Austria

We present a new model for the interaction of molecules with the orbital angular momentum of light, which has long been argued to benefit structural studies and quantum control of molecular ensembles. We derive a general description of the light-matter interaction in terms of the coupling between spherical gradients of the electric field and an effective molecular charge density that exactly reproduces molecular multipole moments. Our model can accommodate for an arbitrary complexity of the molecular structure and is applicable to any electric field, with the exception of tightly focused beams. Within this framework, we derive the general mechanism of angular momentum exchange between the spin and orbital angular momenta of light, molecular rotation and its center-of-mass motion. We demonstrate that vortex beams strongly enhance certain ro-vibrational transitions that are considered forbidden in the case of a non-helical light.

MO 25.5 Fri 12:00 HS 3044

**Investigation on the dynamics of single atom catalysis in superfluid helium nanodroplets** — ●WENTAO CHEN, BRENDAN WOUTERLOOD, and FRANK STIENKEMEIER — Institute of Physics, University of Freiburg, 79104 Freiburg

We introduce a new experimental approach on the dynamics of single atom catalysis in superfluid helium nanodroplets. Single-atom catalysts have recently emerged as a new type of catalysts which are comprised of one metal atom and has different catalytic properties compared to bulk-particle catalysts. The common way to form single-atom catalysts is to isolate a single metal atom on a supporting surface, which makes it difficult to characterize the catalytic activity of the single atom and separate the influence of the surface. Superfluid helium nanodroplets can be an ideal tool to form the isolated molecule-metal atom complexes by doping the reactant molecule and the metal atom successively. Specifically, we were able to form 1,8-octanediol- Au(n=0-2) complexes without a supporting surface by sequentially doping octanediol and a gold atom in helium droplets. After ionizing the complexes by electron impact and comparing the fragment, it has been found that the complexes with Au atoms prominently produce C<sub>2</sub>H<sub>4</sub><sup>+</sup> in this dissociative reaction, while the complexes without Au atoms have more diverse fragments: C<sub>2</sub>H<sub>4</sub><sup>+</sup>, HCO<sup>+</sup> and CH<sub>2</sub>OH<sup>+</sup>. We plan to use femtosecond pump-probe spectroscopy and photoelectron-photoion coincidence methods to study the real-time dynamics of the octanediol- Au complex during the reaction.

MO 25.6 Fri 12:15 HS 3044

**Nanophotonics for precise mid-infrared molecular spectroscopy** — ●JÉRÉMIE PILAT<sup>1</sup>, LUCAS DENIEL<sup>1</sup>, MELISSA A. GUIDRY<sup>2</sup>, DANIL M. LUKIN<sup>2</sup>, BINGXIN XU<sup>1</sup>, KIYOUL YANG<sup>2</sup>, JOSHUA YANG<sup>2</sup>, JELENA VUČKOVIĆ<sup>2</sup>, THEODOR W. HÄNSCH<sup>1,3</sup>, and NATHALIE PICQUÉ<sup>1,4</sup> — <sup>1</sup>Max-Planck Institute of Quantum Optics, Garching, Germany — <sup>2</sup>E. L. Ginzton Laboratory, Stanford University, Stanford, California, USA — <sup>3</sup>Ludwig-Maximilian University of Munich, Faculty of Physics, München, Germany — <sup>4</sup>Max Born Institute, Berlin, Germany

A nanophotonic silicon-carbide waveguide on a 5x5 mm<sup>2</sup> chip dramatically simplifies comb-assisted mid-infrared spectroscopy. The emerging 4H silicon carbide (SiC) on insulator platform provides a high refractive index, strong second- and third-order optical nonlinearity, low losses, and a broad transparency range. Here, a mode-locked laser at

1560 nm excites a dispersion-engineered SiC waveguide. This simultaneously enables frequency-comb self-referencing with an integrated f-2f interferometer and mid-infrared dispersive-wave frequency-comb generation at low 120-pJ pulse energies. By stabilizing the carrier-envelope offset frequency beatnote  $f_{ceo}$  provided by the integrated f-2f interferometer and the repetition rate of the mode-locked laser, accurate tunable-laser molecular spectroscopy of methane is demonstrated at 3.6  $\mu\text{m}$ . Our new tool opens up new opportunities for precision measurements in the mid-infrared molecular fingerprint region, where most molecules exhibit characteristic intense rovibrational transitions, of interest to fundamental research and environmental sensing.

MO 25.7 Fri 12:30 HS 3044

**Azobenzene based lipids as a tool to manipulate physiochemical properties of membrane mimetic systems via light** — ●JUSTIN HORNBOGEN<sup>1</sup>, RITU RAJENDER<sup>2</sup>, STEFAN KINS<sup>2</sup>, ANNETTE MEISTER<sup>3</sup>, DAVID GLÜCK<sup>4</sup>, SANDRO KELLER<sup>4</sup>, and ROLF DILLER<sup>1</sup> — <sup>1</sup>Phys. Dept., RPTU, 67663 Kaiserslautern, GER. — <sup>2</sup>Human Biol. Dept., RPTU, 67663 Kaiserslautern, GER. — <sup>3</sup>Inst. Biochem. and

Biotech., MLU, 06120 Halle, GER. — <sup>4</sup>Inst. Molecular Bioscience (IMB), Univ. of Graz, 8010 Graz, AUT.

Regulation of physiological membrane properties is an auspicious approach towards the treatment of various illnesses, e.g. Alzheimer disease (AD). Azobenzene (AB) decorated lipids are used to manipulate membranes by photoinduced AB trans(E)/cis(Z)-isomerization. We present the photophysical switching behaviour of 18:0-azophosphatidylcholine incorporated into unilamellar phosphatidylcholine (POPC or DMPC) LUV's and glycodiisobutylene/maleic acid lipid particles (POPC- or DMPC-nanodiscs). In addition, we explore the physicochemical impact of AB isomerization by means of methods like (transient) UV/Vis spectroscopy, TEM, DLS and others. The self-assembling nanoparticles can serve as a model system to investigate biochemical functionality of membrane proteins in native-like biomembranes while altering membrane properties such as structure, thickness, lateral pressure, permeability etc. through a light stimulus. Ongoing purification and incorporation of APP and  $\gamma$ -secretase may reveal an influence of AB isomerization on the generation of pathogenic amyloid plaques and  $\gamma$ -secretase activity related to AD.

## MO 26: Cluster

Time: Friday 14:30–16:00

Location: HS 3042

MO 26.1 Fri 14:30 HS 3042

**Setup for time- and energy-resolved fluorescence measurements of collective effects in polyacene aggregates attached to rare gas clusters** — ●ALEKSANDR DEMIANENKO, MORITZ MICHELBACH, SEBASTIAN HARTWEG, and FRANK STIENKEMEIER — Institute of Physics, University of Freiburg, Germany

Collective effects in organic semiconductors affect excited state lifetimes, important for organic optoelectronic and photovoltaic applications. A complete understanding of the energy level structure, and decay mechanisms require high spectral and temporal resolution. We present a setup combining conventional laser-induced fluorescence (LIF) spectroscopy with time-correlated single photon counting (TC-SPC) and discuss advantages and implementation challenges. Using wavelength-tunable nanosecond dye laser pulses allows us to measure high-resolution LIF spectra of transitions to highly excited states of tetracene embedded in superfluid helium nanodroplets, or deposited on solid rare-gas clusters. The newly implemented TCSPC detection in combination with a femtosecond laser system is aimed at studying radiative and non-radiative decay mechanisms connected to collective effects in aggregates of polyacenes. This technique allows us to cover the sub-ns lifetime region not previously reachable in our fluorescence measurements.

MO 26.2 Fri 14:45 HS 3042

**Investigation of the homogeneous linewidth of organic molecules on solid rare-gas clusters** — ●ARNE MORLOK, ULRICH BANGERT, YILIN LI, FELIX RIEDEL, LEONIE WERNER, LUKAS BRUDER, and FRANK STIENKEMEIER — University of Freiburg, Institute of Physics, Hermann-Herder-Str. 3, 79104 Freiburg, Germany

Doped rare-gas clusters are a well-established model system to study molecular systems. In order to harness the full potential of such cluster isolation techniques, it is crucial to understand the residual system-bath interaction between cluster and dopant. We employ two-dimensional electronic spectroscopy (2DES) to study the interaction of organic molecules with solid rare-gas clusters, which allows us to resolve the ensemble inhomogeneity and retrieve information about the molecule-cluster binding configurations [1]. Previously, this approach was applied to resolve the homogeneous linewidth of phthalocyanine molecules attached to neon clusters and ultimately provided a deeper understanding of the structural configurations in a nanoconfined system [2].

We extended this investigation in varying the cluster species and improving the data acquisition scheme, since the previous measurements were limited by long acquisition times. First results are presented, which suggest differences in the homogeneous linewidth and dynamics depending on the cluster species.

[1] L. Bruder et al., J. Phys. B: At. Mol. Opt. Phys. 52 183501 (2019).

[2] U. Bangert et al., Nat. Commun. 13 3350 (2022).

MO 26.3 Fri 15:00 HS 3042

**Collision dynamics and uptake of alcohol molecules by hydrated nitric acid clusters** — YIHUI YAN<sup>1</sup>, KAROLÍNA FÁRNÍKOVÁ<sup>2</sup>, ANDRIY PYSANENKO<sup>2</sup>, EVA PLUHAŘOVÁ<sup>2</sup>, MICHAL FÁRNÍK<sup>2</sup>, and ●JOZEF LENGYEL<sup>1</sup> — <sup>1</sup>TU München, Garching, Germany — <sup>2</sup>Czech Academy of Sciences, Prague, Czechia

Aerosol particles represent one of the most important, yet perhaps the least understood, components of our atmosphere. Due to their influence on global climate, there is a need for detailed kinetic data to be used in climate prediction models. We have therefore developed a novel method for quantifying the uptake process of various molecules by hydrated HNO<sub>3</sub> clusters using a pickup technique. Our experiment combines mass spectrometry of the clusters with velocity measurements. However, the evaluation of the uptake cross sections from the experimental data is based on simplifying assumptions about the molecule-cluster collisions. We validate these assumptions through extensive MD simulations. These calculations allow evaluation of the scattering and uptake processes in the collisions, and subsequently the uptake cross sections can be derived and compared to the experimental values. Herein, we examine the uptake of different alcohol molecules by hydrated HNO<sub>3</sub> clusters. We discuss the dependence of uptake on the length of the carbon chain (i.e., size, mass, and hydrophobicity) and on steric effects. The combination of experimental data with simulations provides insight into the dynamics involved in molecule-cluster collisions, which is essential for validating our experimental approach.

MO 26.4 Fri 15:15 HS 3042

**Mass Spectrometry Analysis of Binary Formic Acid-Water Clusters upon Collision with Electrons** — ●KEVIN LI<sup>1</sup>, JOZEF ĎURANA<sup>2</sup>, MICHAL FÁRNÍK<sup>2</sup>, and JOZEF LENGYEL<sup>1</sup> — <sup>1</sup>TU München, Garching, Germany — <sup>2</sup>Czech Academy of Sciences, Prague, Czechia

A significant portion of atmospheric particles is formed through the nucleation and condensation of precursor gases in a process known as new particle formation (NPF), where organic acids play a crucial role as key precursor gases that enhance nucleation rates. It is, therefore, essential to understand the collisions of gas-phase molecules with clusters and to establish protocols for analyzing these particles using mass spectrometry. This is particularly important for hydrogen-bonded particles, as they frequently undergo extensive fragmentation upon ionization. In our experiments, mixed clusters of formic acid and water were produced in supersonic expansion and subsequently investigated by mass spectrometry using different ionization methods, namely (i) the electron ionization at 70 eV (EI) and (ii) the low energy electron attachment (EA). While for positive ionization mainly protonated clusters (H<sub>2</sub>O)<sub>n</sub>/(HCOOH)<sub>m</sub>/H<sup>+</sup> were detected, negative mass spectroscopy revealed two species, (H<sub>2</sub>O)<sub>n</sub>/(HCOOH)<sub>m</sub> and (H<sub>2</sub>O)<sub>n</sub>/(HCOOH)<sub>m-1</sub>/HCOO<sup>-</sup>. Both techniques indicate that higher water content in the solution results in clusters with a high degree of hydration and fewer formic acid molecules. Additionally, the fraction of the two anionic species is influenced by cluster size, level

of hydration, and electron energy. The detailed behavior of ionization will be discussed in the presentation.

MO 26.5 Fri 15:30 HS 3042

**Electron scattering in neutral water clusters** — ●KATINKA HORN<sup>1</sup>, SVETLANA TSIZIN<sup>1</sup>, LOREN BAN<sup>1</sup>, SEBASTIAN HARTWEG<sup>2</sup>, PETRA HOFFMANN<sup>1</sup>, EGOR CHASOVSKI<sup>1</sup>, BRUCE L. YODER<sup>1</sup>, and RUTH SIGNORELL<sup>1</sup> — <sup>1</sup>ETHZ, Laboratory of Physical Chemistry, Switzerland — <sup>2</sup>University of Freiburg, Institute of Physics, Germany

A detailed understanding of low-energy electron scattering in water (with kinetic energies below 100 eV) is crucial to modeling and controlling many processes occurring in nature, ranging from atmospheric chemistry to radiation biology. While condensed and gas phase electron scattering cross sections are known for water, analogous data for scattering in water clusters is still missing. This is the case even though clusters, often exhibiting unique/tunable properties, are of great interest for bridging the gap between the gas and condensed phases. The presented work is an extension and refinement of previous studies, providing more detailed information on electron scattering in neutral water clusters of various sizes ionized with photon energies up to ~50 eV. Electron transport scattering in water clusters was investigated by angle-resolved photoelectron spectroscopy. The scattering cross sections for the model were retrieved from cluster-size and energy resolved data contained in the photoelectron anisotropy parameter  $\beta$ . We found larger electron scattering cross sections for clusters than for the condensed phase, likely due to reduced dielectric screening in clusters. Good agreement to experiment is achieved with a condensed phase scattering model, using a kinetic energy and scattering channel

dependent scaling of bulk cross sections.

MO 26.6 Fri 15:45 HS 3042

**Electron transfer processes and the formation of solvated dielectrons by UV excitation in sodium-ammonia clusters** — ●SEBASTIAN HARTWEG<sup>1,2</sup>, JONATHAN BARNES<sup>3</sup>, BRUCE L. YODER<sup>3</sup>, GUSTAVO A. GARCIA<sup>2</sup>, LAURENT NAHON<sup>2</sup>, EVANGELOS MILIORDOS<sup>4</sup>, and RUTH SIGNORELL<sup>3</sup> — <sup>1</sup>Institute of Physics, University of Freiburg, Germany — <sup>2</sup>Synchrotron Soleil, St. Aubin, France — <sup>3</sup>DCHAB, ETH Zürich, Switzerland — <sup>4</sup>Auburn University, Alabama, USA

Solvated electrons play important roles in the origin and formation of radiation damage in biological tissue as well as for large-scale chemical synthesis. Electron solvation has first been observed in alkali ammonia solutions. These systems with their many peculiar concentration dependent properties[1-3] including the formation of stable solvated dielectrons and a transition to a metallic phase, are not well understood on a molecular level, despite the many studies conducted on them.

I will present our recent photoelectron/photoion coincidence study with support from quantum chemical calculations[4], in which we could identify different electron transfer processes occurring in sodium ammonia clusters upon interaction with UV and VUV radiation. Among these processes, the formation of transient solvated dielectrons and their subsequent decay via an electron-transfer mediated decay process constitutes the first direct observation of solvated dielectrons.

- 1.Zurek, E., et al. *Angew. Chem. Int. Ed.*, 2009. 48(44)
- 2.Buttersack, T., et al. *Science*, 2020. 368(6495)
- 3.Hartweg, S., et al. *Angew. Chem. Int. Ed.*, 2016. 55(40)
- 4.Hartweg, S., et al. *Science*, 2023. 380(6650)

## MO 27: Ultrafast Dynamics III and High-harmonic Generation (joint session MO/A)

Time: Friday 14:30–16:30

Location: HS 3044

MO 27.1 Fri 14:30 HS 3044

**Absolute photoemission timing in neon** — ●MAXIMILIAN FORSTER, MAXIMILIAN POLLANKA, CHRISTIAN SCHRÖDER, and REINHARD KIENBERGER — Chair for laser and x-ray physics, E11, Technische Universität München, Germany

We measure the relative photoemission time delay between the Ne2p, Ne2s and the Iodine 4d states in iodomethane utilizing attosecond streaking. This allows us to experimentally determine the absolute time delay of neon 2s and 2p photoelectrons for the first time. The delay of neon, being the first ever evidence of atomic delay, has received repeated attention by both experimental and theoretical investigations due to the large cross section and convenient properties of neon. While helium has been the gold standard for absolute time delay measurements, enabled by remarkable theoretical agreement, due to spectral overlap helium cannot be used to reference neon. Recent developments, namely the availability of different chronoscopes, enable measuring the absolute time delay of neon. We take the path via iodomethane and the I4d core state, which has been timed on an absolute scale, and use it to reference neon. The delay between Ne2s and Ne2p can be extracted simultaneously, allowing for a positive consistency check with previous experiments conducted only with neon. Timing neon on an absolute scale allows an assignment of absolute values to these experiments in retrospect and establishes neon as a chronoscope species.

MO 27.2 Fri 14:45 HS 3044

**Isosteric molecules in the time-domain** — ●MAXIMILIAN POLLANKA, CHRISTIAN SCHRÖDER, MAXIMILIAN FORSTER, and REINHARD KIENBERGER — Physik Department, Technische Universität München, James-Franck-Str. 1, 85748 Garching, Germany

We report on absolute photoemission timing measurements on isosteric molecules in the gas phase. Photoemission time delays are accessed via streaking spectroscopy on attosecond timescales. To be able to (directly) access absolute photoemission times of the respective outer and inner valence states of N<sub>2</sub>O and CO<sub>2</sub> we are using iodomethane (I4d) as a timing reference. In a complementary study He was used as reference to cross-check the results as well as to verify the usability of the respective chronoscope species. Due to the similarities in molecular structure (isostericity) and electronic configurations (isoelectronicity) between these investigated molecules, the pure effect of the specific molecular/orbital characteristics is expected to be probed. Additionally, N<sub>2</sub> and CO is studied in the same way on the basis of their isos-

teric behavior. The experimental data show great similar tendencies but also differences between the compared molecular orbitals, which are determined but not completely understood up to now. Nonetheless, recent theoretical calculations hint towards an additional channel coupling photoemission time delay contribution that can be assigned to electron correlations responsible for re-disturbing the excitation among different final photoionization channels.

MO 27.3 Fri 15:00 HS 3044

**Attosecond time-resolved coincidence spectroscopy of ethylene** — ●BARBARA MERZUK<sup>1</sup>, DAVID BUSTO<sup>1,2</sup>, IOANNIS MAKOS<sup>1</sup>, DOMINIK ERTEL<sup>1</sup>, MARVIN SCHMOLL<sup>1</sup>, BENJAMIN STEINER<sup>1</sup>, FABIO FRASSETTO<sup>3</sup>, LUCA POLETTI<sup>3</sup>, ROBERT MOSHAMMER<sup>4</sup>, CLAUDIUS DIETER SCHRÖTER<sup>4</sup>, THOMAS PFEIFER<sup>4</sup>, SERGUEI PATCHKOVSKI<sup>5</sup>, JAKUB BENDA<sup>6</sup>, ZDENĚK MAŠÍN<sup>6</sup>, and GIUSEPPE SANSONE<sup>1</sup> — <sup>1</sup>Albert-Ludwigs-Universität Freiburg, Germany — <sup>2</sup>Lund University, Sweden — <sup>3</sup>CNR, Padova, Italy — <sup>4</sup>MPIK, Heidelberg, Germany — <sup>5</sup>MBI Berlin, Germany — <sup>6</sup>Charles University, Prague, Czech Republic

Studying photoionization dynamics and characterising the time delays associated with the photoemission of an electron wave packet can unveil important characteristics of coupled electronic-nuclear dynamics in molecular systems. Attosecond photoelectron spectroscopy in combination with electron-ion coincidence detection is beneficial since this allows disentangling the different photoionization and dissociation channels. Additionally, it may give access to the orientation of the molecule at the instant of photoionization. Using our experimental setup that consists of an attosecond beamline, based on high-order harmonic generation operating at 50 kHz repetition rate, we investigate the photoionization dynamics in ethylene molecules by performing RABBIT (Reconstruction of Attosecond Beating By Interference of Two-photon transitions) measurements while detecting photoelectrons and photoions in coincidence. The experimental results are interpreted with the help of multi-electron R-matrix calculations of two-photon ionization.

MO 27.4 Fri 15:15 HS 3044

**Probing well aligned molecular environments on surfaces via attosecond streaking** — ●PASCAL SCIGALLA<sup>1</sup>, SVEN PAUL<sup>1</sup>, CHRISTIAN SCHRÖDER<sup>1</sup>, PETER FEULNER<sup>2</sup>, and REINHARD KIENBERGER<sup>1</sup> — <sup>1</sup>Chair for laser and x-ray physics, E11, Technische Universität München, Germany — <sup>2</sup>Surface and Interface Physics, E20, Technische Universität München, Germany

We report on the photoemission timing measurements of well-aligned iodomethane and -ethane molecules on a Pt111 surface. In this set of experiments, we clock the  $I4d$  photoemission of iodine against the Platinum valence photoemission using the attosecond streak camera technique, allowing the extraction of a relative photoemission delay. As the  $I4d$  photoemission in the selected energy range is dominated by a giant resonance in the  $I4d \rightarrow ef$  channel, its photoemission time is mostly unaffected by its chemical environment; thus, any observed change in the photoemission delay can be attributed to the traversed potential landscape of the molecule. By carefully selecting the detection angle and crystal surface coverage we can reliably choose whether only parts of the molecule or its entirety was traversed by the detected photoelectron wavepackets. It is furthermore possible to investigate the influence of slight coverage variations onto the observed photoemission delay. Planned, complementary scattering simulations will be used to gain deeper insight into the observations with the goal to establish photoemission timing experiments as an efficient and accurate means to study molecular environments on surfaces.

MO 27.5 Fri 15:30 HS 3044

**Automatic optimization of intense high-harmonic pulses** — ●JOSÉ GÓMEZ TORRES, FREDERIC USSLING, SIMON WÄCHTER, ALESSANDRO COLOMBO, LINOS HECHT, KATHARINA KOLATZKI, ALEXANDRE ROSILLO VORSIN, MARIO SAUPPE, and DANIELA RUPP — ETH Zurich, Laboratory for Solid State Physics, John-von-Neumann-Weg 9, 8093 Zurich, Switzerland

High harmonic generation (HHG) allows the production of extreme ultraviolet pulses ranging from picosecond up to attosecond timescales from intense infrared (IR) pulses, making it an invaluable tool for the study of ultrafast phenomena. It has been recently demonstrated that HHG is capable of producing pulses intense enough for diffraction experiments like coherent diffraction imaging of isolated nanoparticles [1]. Very intense pulses of short time duration in a stable delivery over hours are necessary for this, requiring a time-consuming optimization of the experimental parameters. We developed a tool for the automatic optimization of HHG parameters, sweeping different geometric parameters of the setup and measuring for each step the pulse energy achieved. Due to the complexity of simulating the specific conditions of the experiment, this trial and error approach is a necessary final step to achieve the highest pulse energy. In order to optimize the XUV peak focal intensity, we perform electron spectroscopy on a diffuse gas in the focus region. Via IR-XUV pump probe, RABBITT measurements can be carried out for the temporal characterization of pulses.

[1] D. Rupp et al., Nature Communication 8, 493 (2017)

MO 27.6 Fri 15:45 HS 3044

**Orbital interference effects in low-order harmonic generation in benzene** — ●SAMUEL SCHÖPA, FALK-ERIK WIECHMANN, FRANZISKA FENNEL, and DIETER BAUER — Universität Rostock, Rostock, Germany

We explore the impact of the driving laser's ellipticity and polarization on the low-order harmonic spectrum of benzene and find a strong interference in the 5th harmonic between emission originating from transitions between  $\pi$  orbitals and emission from  $\sigma$  orbitals. The contribution of the  $\pi$  orbitals entirely vanishes due to interference for driving with a laser polarized along a  $\sigma_v$  mirror axis. However, the

$\pi$  orbital's contribution takes over for elliptic polarization while being fundamentally different from the  $\sigma$  orbital emission, i.e., having the opposite helicity and a perpendicular major polarization axis. The resulting interference yields a complex dependence of the low-order harmonic spectrum of benzene on the ellipticity and the polarization of the driving field.

MO 27.7 Fri 16:00 HS 3044

**Observation of HHG from organic molecular crystals** — ●FALK-ERIK WIECHMANN<sup>1</sup>, SAMUEL SCHÖPA<sup>1</sup>, ALEXANDER VILLINGER<sup>2</sup>, DIETER BAUER<sup>1</sup>, and FRANZISKA FENNEL<sup>1</sup> — <sup>1</sup>Institute of Physics, Rostock, Germany — <sup>2</sup>Institute of Chemistry, Rostock, Germany

This project aims at a detailed understanding of the harmonic generation process in large organic molecules in the crystalline phase. Unlike previous studies, which were limited to small molecules in the gas phase, we introduce organic molecular crystals as a novel target for HH spectroscopy, taking advantage of the inherent molecular alignment. Unlike in gas phase experiments, neighboring molecules in organic crystals experience a weak but finite coupling, leading to 'solid like' features, e.g. a delocalization of the electronic states over several unit cells. With a fundamental 4000 nm mid-IR beam reaching 6 TW/cm<sup>2</sup> we demonstrate that HHG up to the order of 17 is possible without imposing physical damage. When the fundamental driving polarization is rotated, maxima of harmonic emission occur at polarization directions parallel to connecting axes between neighboring molecules, reflecting the crystal structure. Despite the linearly polarized driving field, the emitted harmonics exhibit elliptical polarization with a main axis different from the fundamental polarization direction.

MO 27.8 Fri 16:15 HS 3044

**High-order Harmonic Generation (HHG) in the nonadiabatic regime over a sub-mm glass chip** — ●SABINE ROCKENSTEIN<sup>1,2</sup>, AGATA AZZOLIN<sup>1,2</sup>, GAIA GIOVANETTI<sup>2</sup>, GUANGYU FAN<sup>2,3</sup>, MD SABBIR AHSAN<sup>2,4</sup>, OLIVIERO CANNELLI<sup>2</sup>, LORENZO COLAIZZI<sup>1,2,5</sup>, ERIK P MÅNSSON<sup>2</sup>, DAVIDE FACCIALÀ<sup>4</sup>, FABIO FRASSETTO<sup>4</sup>, DARIO W LODI<sup>5</sup>, CRISTIAN MANZONI<sup>4</sup>, REBECA M VÁZQUEZ<sup>4</sup>, MICHELE DEVATTA<sup>4</sup>, ROBERTO OSELLAME<sup>4</sup>, LUCA POLETTA<sup>4</sup>, SALVATORE STAGIRA<sup>4,5</sup>, CATERINA VOZZI<sup>4</sup>, VINCENT WANIE<sup>2</sup>, ANDREA TRABATTONI<sup>2,6</sup>, and FRANCESCA CALEGARI<sup>1</sup> — <sup>1</sup>UHH (DE) — <sup>2</sup>DESY (DE) — <sup>3</sup>CUI (DE) — <sup>4</sup>CNR (IT) — <sup>5</sup>Politecnico di Milano (IT) — <sup>6</sup>Uni. Hannover (DE)

HHG-based sources are nowadays operating up to the soft-x spectral region. One of the main challenges remains to extend the cut-off frequency while retaining high-photon flux. Approaches based on the so-called nonadiabatic regime have allowed to overcome phase matching limitations and achieve substantial cut-off extension [1]. We present a new HHG source, operating with high driver laser intensities (up to 1E16 W/cm<sup>2</sup>) and a laser-micromachined glass cell allowing for highly efficient gas confinement over 900  $\mu$ m, to achieve nonadiabatic phase matching. The setup was operated with both 800-nm and 1500-nm sub-35-fs driving pulses. With the 800-nm driver, the HHG energy cutoff was extended to 100 eV in Argon and 180 eV in Neon, 160 eV were reached using the 1500-nm driver in Argon. Our results highlight the potential of optimizing the nonadiabatic regime for covering the water-window spectral region. [1] Johnson et al., Sci. Adv. 4(5), 2018