O 79: Ultrafast Electron Dynamics III

Time: Thursday 10:30–12:30 Location: H2

O 79.1 Thu 10:30 H2

Hybrid Exciton Orbital Tomography in 2D-organic interfaces — •Christian Simon Kern[1], Michele Capra[2], Marco Gruenewald[3], Torsten Fritz[3], Mirko Cinchetti[2], Peter Puschnig[1], and Giovanni Zamborlini[1] — [1]Institute of Physics, University of Graz, Austria — [2]Department of Physics, TU Dortmund University, Germany — [3]Institute of Solid State Physics, Friedrich Schiller University Jena, Germany

Organic molecules adsorbed on semiconducting 2D substrates pose an interesting class of interfaces for optoelectronic applications. Here we investigate the interface of pentacene and a transition metal dichalcogenide layer with many-body perturbation theory. In particular, we compare those results to optical spectroscopy and demonstrate the connection of excitons to their experimentally observable photoemission signatures in pump-probe spectroscopy. The latter approach— exciton photoemission orbital tomography [1]—especially allows for the unambiguous characterization of the excitons' hole and electron configuration, and is extended to the case of hybrid interlayer excitons.

[1] C. S. Kern, A. Windischbacher, and P. Puschnig, Photoemission orbital tomography for excitons in organic molecules, PRB 108, 085132 (2023)

O 79.2 Thu 10:45 H2

Ultrafast table-top three-dimensional photoemission orbital tomography — •G. S. Matthijs Jansen[1], Wiebke Bennecke[1], Thi Lan Dinh[2], Jan Philipp Bange[1], David Schmitt[1], Marco Merboldt[1], Lennart Weinhagen[1], Bent van Wingerden[1], Fabio Frassetto[3], Luca Poletto[3], Marcel Reutzel[1], Daniel Steil[1], D. Russell Luke[2], and Stefan Mathias[1] — [1]University of Göttingen, I. Physikalisches Institut, Göttingen, Germany — [2]University of Göttingen, Institute of Numerical and Applied Mathematics, Göttingen, Germany — [3]Institute for Photonics and Nanotechnologies CNR-IFN, 35131 Padova, Italy

In photoemission orbital tomography (POT), molecular orbitals can be imaged with femtosecond resolution. Also, when combined with photon-energy-dependent measurements, POT is, so far, the only method that can probe the orbitals of adsorbed molecules in 3D. However, the study of, e.g., hybridization in organic/inorganic heterostructures [Bennecke *et al.*, arXiv:2411.14993 (2024)] by 3D-POT is extremely challenging due to the demanding nature of the experiment. Here, we present a table-top approach for 3D POT: By combining a photoelectron momentum microscope with a pulse-preserving monochromator for laser-generated extreme ultraviolet light, we speed up data acquisition. Moreover, we developed a new reconstruction algorithm that reduces the sampling requirements by about an order of magnitude [Dinh *et al.*, New J. Phys. 26 043024 (2024)]. Our first results achieved on PTCDA/Ag(110) highlight the potential for ultrafast femtosecond time-resolved 3D-POT.

O 79.3 Thu 11:00 H2

Exciton wave function signatures from time-resolved photoemission tomography of an organic molecular layer — •Siegfried Kaidisch[1], Marcel Theilen[2], Monja Stettner[3], Eric Fackelman[3], Galit Cohen[4], Amir Kleiner[4], Christian Simon Kern[1], Andreas Windischbacher[1], Sivan Refaely-Abramson[4], Frank Stefan Tautz[3], Ulrich Höfer[2], and Peter Puschnig[1] — [1]Institute of Physics, University of Graz, Austria — [2]Fachbereich Physik, Philipps-Universität Marburg — [3]PGI-3, FZ Jülich & RWTH Aachen University — [4]Weizmann Institute of Science, Israel

Oriented layers of organic molecules adsorbed on passivated metal surfaces are a promising class of interfaces for studying electron dynamics at femtosecond timescales. In particular, time-resolved photoemission tomography promises to reveal information about the electron distribution in optically excited states. Using the example of sexithiophene (6T) multilayers adsorbed on Cu(110)-p(2x1)O, we observe photoemission patterns for the low-energy excited states. These measured momentum maps exhibit signatures that cannot be explained by a mere population of the lowest unoccupied molecular orbital of 6T. To analyze the source of these features, we perform GW/BSE (Bethe-Salpeter equation) calculations on multiple levels of theory (gas-phase, cluster, embedded and periodic calculations). Using the framework of photoemission orbital tomography for excited states, we also simulate the photoemission patterns and thereby shed light on the nature of the exciton wave function of the lowest optically allowed state.

O 79.4 Thu 11:15 H2

Quenching Strong-Field Recollisions at Nanotapers with Strong Bias Fields — •Rasmus Lampe, Germann Hergert, and Christoph Lienau — Institut für Physik, Carl-von-Ossietzky Universität, 26129 Oldenburg, Germany

Recollisions of intensively accelerated electrons with the parent are a well-known effect in strong-field photoemission of electrons, leading to photoemission spectra showing a recollision plateau with a cut-off energy of ten times the ponderomotive energy. It has recently been demonstrated that such a recollision plateau also arises in multiphoton photoemission from metallic nanostructures, showing that recollisions are much more fundamental to photoemission than anticipated [1].

We analyze electron recollisions from strongly biased tungsten nanotapers illuminated by few-cycle near-infrared laser pulses. We demonstrate significant changes in the photoelectron spectra in the strong-field regime by applying static electric fields on the order of the optical near-field driving the photoemission. A continuous transition of a horizontal recollision plateau into triangularly shaped spectra is observed.

Stronger static fields increase the acceleration of the electrons away from the apex, such that the ponderomotive acceleration by the near-field is not sufficient to drive the electrons back to the surface, ultimately suppressing recollisions. This enables the control of the electron motion with bias fields, which has an immediate importance for ultrafast low-energy electron microscopy allowing to maintain few fs time resolution of the electron beam over mesoscopic distances.

[1] B. Bánhegyi et al., Phys. Rev. Lett. 133, 033801 (2024)

O 79.5 Thu 11:30 H2

Time-resolved photoemission orbital tomography of 6T on Cu(110)-(2×1)O — •Marcel Theilen[1], Monja Stettner[2], Eric Fackelman[2], Francois C. Bocquet[2], Alexa Adamkiewicz[1], Sarah Zajusch[1], Siegfried Kaidisch[3], Christian Kern[3], Robert Wallauer[1], Peter Puschnig[3], F. Stefan Tautz[2], and Ulrich Höfer[1,4] — [1]Fachbereich Physik, Philipps-Universität Marburg — [2]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich — [3]Institut für Physik, Universität Graz — [4]Fachbereich Physik, Universität Regensburg

For ordered molecular layers, photoemission orbital tomography (POT) is a powerful technique for imaging the electron distribution of molecular orbitals in momentum space. When combined with laser pump-probe techniques, time-resolved photoemission orbital tomography (tr-POT) offers the ability to track the population dynamics of the excited molecular states on a femtosecond time scale[1].

In this talk, I will discuss our recent results obtained through tr-POT for three distinct, well-ordered layers of sexithiophene (6T) on a Cu(110)-(2×1)O surface: a monolayer, a bilayer and a multilayer. The focus will primarily be on the dynamics of the populated 6T LUMO via an optically HOMO-LUMO transition. For instance, a comparison between the individual layers reveals a significant extension of the LUMO lifetime with increasing layer thickness. Specifically, we find that the lifetime for the monolayer is less than 50 fs, while it increases to more than 600 fs for the multilayer.

[1] R. Wallauer *et al.*, Science **371**, 1056 (2021).

O 79.6 Thu 11:45 H2

Understanding the ultrafast electron dynamics and CDW transition in $LaTe_3$ using machine learning — •Gesa Siemann[1], Davide Curcio[1], Paulina Majchrzak[1], Charlotte Sanders[2], Jenny Rigden[2], Yu Zhang[2], Deepnarayan Biswas[3], Leslie Schoop[4], Emma Springate[2], and Philip Hofmann[1] — [1]Department of Physics and Astronomy, Aarhus University, DK — [2]Central Laser Facility, Harwell, UK — [3]Diamond Light Source, UK — [4]Department of Chemistry, Princeton University, USA

The rare-earth tritelluride $LaTe_3$ hosts a unidirectional charge density wave (CDW) with a high transition temperature of 670 K. Recently, it has been suggested that exposing the system to a short light pulse not only suppresses this primary CDW but also induces a second CDW in the perpendicular direction[1]. An open question is, how these struc-

tural dynamics affect the electronic structure, and if fingerprints of the second CDW can be found in corresponding data obtained by time- and angle-resolved photoemission spectroscopy. Here, we explore this question, studying the frequency-dependent coherent response of the system, and the time-dependent evolution of the Fermi surface topology, which we compare to predictions by a simple tight-binding model. We support our analysis using k-means clustering, a machine learning technique, in order to identify different dynamics throughout the Brillouin zone. This reveals varying relaxation times across the Fermi surface, as well as multiple frequencies that can be ascribed to coherent excitations. [1]A. Kogar *et al.*, *Nat. Phys.* 16, 159*163 (2020).

O 79.7 Thu 12:00 H2

Following charge-transfer between plasmonic NPs and adsorbed molecules by time-resolved IR spectroscopy — •Daniel Sandner[1], Katrin Schulz[1], Andrei Stefancu[2], Reinhard Kienberger[1], Emiliano Cortes[2], and Hristo Iglev[1] — [1]Lehrstuhl für Laser- und Röntgenphysik E11, TUM, James-Franck Str 1, 85748 Garching — [2]Fakultät für Physik, LMU München

Plasmonic Nanoparticles can efficiently convert light in a broad range into hot charge carriers, which can be subsequently transferred to molecules or semiconductors for photocatalytic processes. The role of charge-carrier-mediated or thermal reaction pathways is still under debate. Here, we use time-resolved IR spectroscopy between 1200-3000 cm-1 as a sensitive probe for free charges and study the dynamics of different molecules attached to silver NPs. Charge transfer is only observed for resonant excitation of the plasmon resonance and in the presence of attached molecules. Furthermore, we find a correlation between the lifetime of transferred charges and the chemical reactivity.

O 79.8 Thu 12:15 H2

Ultrafast Photoexcitation of Semiconducting Photocathode Materials: An Ab Initio Study — •Hilde Bellersen, Michele Guerrini, and Caterina Cocchi — Carl von Ossietzky Universität Oldenburg, Institute of Physics, 26129 Oldenburg, Germany

Cs-based semiconductors like Cs_3Sb and Cs_2Te are currently used as photocathode materials in particle accelerators. Their performance as electron sources critically depends on their response to the intense laser radiation impinging them. In this work, we investigate from first principles the time-dependent response of Cs_3Sb and Cs_2Te to ultrafast laser pulses of varying intensities, ranging from 1 GW/cm^2 to 100 TW/cm^2. Nonlinear effects, including high harmonic generation and multiphoton absorption, emerge at thresholds of 400 GW/cm^2 for Cs_3Sb and 500 GW/cm^2 for Cs_2Te. Beyond these intensities, the energy uptake and number of excited electrons saturate, with renewed increases observed beyond 10 TW/cm^2. These findings provide new insights into the nonlinear optical properties of Cs_3Sb and Cs_2Te, contributing to the optimization of these materials for the development of next-generation photoinjectors.