O 14: Ultrafast electron dynamics at surface and interfaces I

Time: Monday 15:00–18:00 Location: TRE/MATH

Invited Talk O 14.1 Mon 15:00 TRE/MATH

Spin and time-resolved ARPES and the orbital angular momentum of light — Pierre Nonnon[1,2], Romain Géneaux[1], Karol Hricovini[1,2], Christine Richter[1,2], David Bresteau[1], and •Mauro Fanciulli[1,2] — [1]Cergy Paris University, France — [2]LIDYL CEA Saclay, France

I will present the recently developed Panoramix beamline at Attolab, open to users, which allows for spin, time and angle-resolved photoemission spectroscopy (STARPES) experiments. The combination of a high harmonic generation (HHG) setup with tunable repetition rate and pulse duration, and a hemispherical analyzer with a 3D spin detector, permits to study charge and spin ultrafast dynamics in material surfaces. As an example, I will compare the observation of chiroptical resonant control of spin polarized excitons in bulk 2H-WSe2, to how the spin polarization changes for electrons pumped beyond bandgap resonance in the unoccupied conduction band. In the search of ever-increasing multidimensional spectroscopic techniques, I will conclude on the possibility to further include exotic light pulses with azimuthally varying phase or polarization, and how they might affect the photoemission process.

O 14.2 Mon 15:30 TRE/MATH

Disentangling exciton resonances in 2D materials by time-resolved XUV-ARPES — •Gregor Zinke[1,2], Antonius Naujok[1], Franz Spartz[1], Sebastian Hedwig[1], Tobias Eul[2], Martin Aeschlimann[1], and Benjamin Stadtmüller[2] — [1]Department of Physics and Research Center OPTIMAS, RPTU University Kaiserslautern - Landau, 67663 Kaiserslautern, Germany — [2]Experimental Physics II, Institute for Physics, University of Augsburg, 86159 Augsburg

Ultrafast and coherent electron dynamics in 2D materials and their heterostructures are governed by the complex energy landscape of various excitons. These quasi-particles are often separated only by tens of meV, making them difficult to distinguish in conventional time-resolved experiments due to the spectral linewidth of ultrashort pulses. Although interferometric photoemission experiments have shown potential in resolving (coherent) excitation pathways and absorption resonances, they depend mostly on optical pulses, limiting the observable momentum space, leaving high-symmetry points of 2D materials inaccessible. To address these limitations, we present a modified version of interferometric trARPES that employs two active, phase-stabilized IR pump pulses and a fs-XUV probe pulse for photoionization. We demonstrate the capability of our approach by examining the well-known A-exciton transition energies in WSe_2 at different frequencies of our pump spectrum, showing how temporal Fourier analysis of the entire ARPES spectrum allows linking distinct transition energies to congruent excitation paths within the WSe_2 electronic band structure.

O 14.3 Mon 15:45 TRE/MATH

Multi-mode momentum microscopy for studying light-induced metastable states — •Junde Liu[1], Bent van Wingerden[1], Hashima Marukara[1], Paul Werner[1], Daniel Steil[1], G. S. Matthijs Jansen[1], Wiebke Bennecke[1], Jan Philipp Bange[1], Olena Tkach[2], Gerd Schönhense[2], and Stefan Mathias[1] — [1]Georg-August-Universität Göttingen, Germany — [2]Johannes Gutenberg-Universität Mainz, Germany

Light-induced ultrafast phase transitions characterized by domain competition involving interlayer ordering, chirality, and topological defects are attracting growing interest [1]. This calls for time-resolved momentum microscopy, which offers simultaneous spatiotemporal resolution and direct access to band structures [2]. However, conventional high-field setups near the sample are prone to field-emission and space-charge effects, particularly in the case of cleaved crystal surfaces.

Here, using cleaved bulk TaS_2 as an exemplary system, we employ a momentum microscope with a newly designed lens system [3] that minimizes the electric field at the sample in the gap-lens mode to suppress field emission, or applies a retarding field in the repeller mode to remove slow electrons and thereby mitigates space-charge effects. This development paves the way to study cleaved crystals and enables dark-field imaging capabilities of domain-resolved electronic structure, nonequilibrium pathways, and the microscopic mechanisms of light-driven phase transitions. [1] Liu et al., *arxiv:2405.02831* (2024); [2] Reutzel, Jansen, Mathias, *Adv. in Phys. X* 9, 2378722 (2024); [3] Tkach & Schönhense, *Ultramicroscopy* 276, 114167 (2025).

O 14.4 Mon 16:00 TRE/MATH

Tracking ultrafast exciton-polariton propagation and coherent energy exchange in thin WSe_2 films — •Tobias Eul[1,2], Kai Rossnagel[2,3,4], and Michael Bauer[2,3] — [1]Institute of Physics, University of Augsburg — [2]Kiel Nano, Surface and Interface Science KiNSIS, Kiel University — [3]Institute of Experimental and Applied Physics, Kiel University — [4]Deutsches Elektronen-Synchrotron DESY, Hamburg

Efficient charge transfer of electrons and holes after photogeneration is fundamental for light-induced energy conversion. The propagation of bound electron-hole pairs, i.e., excitons, can enhance this efficiency by reducing non-radiative losses before charge separation and extraction. A promising approach to improve exciton propagation is via exciton-polaritons, quasiparticles formed by the strong coupling of excitons and photons. In these hybrid states, the excitation coherently oscillates between exciton and photon character, enabling the photonic component to transport energy over extended distances. Using photoemission electron microscopy, we visualize the propagation of exciton-polaritons in a thin WSe2 film by observing the ultrafast energy exchange between excitons and photons.

O 14.5 Mon 16:15 TRE/MATH

Probing On-Chip THz Generation by Momentum Streaking in Photoelectron Emission Microscopy — •David Huber, Wolfgang Hoppe, Georg Woltersdorf, and Wolf Widdra — Martin-Luther-Universität Halle-Wittenberg

THz electromagnetic radiation provides ultrafast electric fields on the femtosecond timescale. Using THz radiation as a pump in time-resolved photoemission experiments enables sub-cycle probing of induced dynamics e.g. charge and spin currents. The momentum and energy streaking of electrons in vacuum – i.e., their acceleration due to the Coulomb force – has long been shown to reliably probe electric fields and THz radiation.

In a spintronic THz emitter, we generate a spin current through the ultrafast demagnetization of a 3 nm ferromagnetic thin film, which results in a surface charge current in an adjacent 3 nm platinum film due to the inverse spin Hall effect. Similar to dipole radiation, the charge current emits THz radiation into the vacuum. By time-resolved in-plane momentum streaking of photoemitted electrons, we probe the single-cycle THz electric field in front of the surface and deduce the electric surface current. We verify our findings using simulated THz radiation and streaking.

O 14.6 Mon 16:30 TRE/MATH

Interplay between intraband acceleration and interband excitation in Bi_2Te_3 by subcycle time-resolved THz-ARPES — •Tim Bergmeier[1,2], Suguru Ito[1], Jens Güdde[1], and Ulrich Höfer[2] — [1]Fachbereich Physik, Philipps-Universität Marburg, Germany — [2]Fachbereich Physik, Universität Regensburg, Germany

Angle-resolved photoemission spectroscopy (ARPES) using THz driving fields with subcycle temporal resolution offers unique capabilities to investigate the dynamics of lateral Dirac currents in the surface state of topological insulators. Whereas low THz-frequencies result in pure intraband accelerations within the surface state [1], additional interband excitations become possible at higher frequencies [2,3].

Here, we present subcycle-resolved THz-ARPES measurements with 2D momentum imaging resolving the full Dirac cone of Bi_2Te_3. For this purpose, we generate frequency-tunable two-cycle MIR pulses (20-40 THz) with electric field strengths up to MV/cm at the sample surface, combined with an ultrashort 400 nm two-photon probe (<15 fs), at a repetition rate of 200 kHz. For a frequency of 25 THz, we show that the momentum distribution of the field-driven intraband acceleration is strongly influenced by the Dirac cones hexagonal warping, while being superimposed with resonant interband excitations that are only possible in the materials $\overline{\Gamma M}$ direction.

[1] J. Reimann et al., Nature 562, 396 (2018).
[2] S. Ito et al., Nature 616, 696 (2023).
[3] C.P. Schmid et al., Nature 593, 385 (2021).

O 14.7 Mon 16:45 TRE/MATH

Realistic nuclear ensembles for electronic excitation dynamics — •Hannah Bertschi, George Trenins, and Mariana Rossi — MPI for the Structure and Dynamics of Matter, Hamburg, Germany

Understanding how large-amplitude anharmonic nuclear motion influences electronic excitations is essential for explaining related phenomena in weakly-bound systems. To model charge transfer and vibronic spectra, we employ real-time time-dependent density functional theory coupled to multitrajectory Ehrenfest dynamics. In this approach, nuclear anharmonicity is incorporated through the sampling of initial conditions. In contrast to more conventional methods, we generate nuclear configurations and momenta using quantum thermostat molecular dynamics [1]. The resulting distributions of nuclear positions and momenta agree well with exact quantum references obtained from path-integral molecular dynamics, even for systems with large-amplitude motion such as the water dimer. However, the quantum thermostat does not perfectly capture the anharmonic zero-point energy of every degree of freedom, which can be mitigated by carefully choosing the parametrization of the thermostat. We show that the initial nuclear conditions of a water dimer on phenanthrene have a strong impact on both the direction and magnitude of charge transfer. Moreover, the nonadiabatic dynamics can even reverse the direction of the transfer. [1] M. Ceriotti et al., Phys. Rev. Lett. 103, 030603 (2009).

O 14.8 Mon 17:00 TRE/MATH

Laser-induced anisotropic electronic response and inverse Faraday effect in hexagonal boron nitride from real-time time-dependent density functional theory — •Cheng Wang, Markus E. Gruner, and Rossitza Pentcheva — Department of Physics, University of Duisburg-Essen

The ultrafast electronic response of hexagonal boron nitride (h-BN) is investigated using real-time time-dependent density functional theory (RT-TDDFT) within an all-electron LAPW framework of the Elk code. We vary the light polarization (in-plane vs out-of-plane), dimensionality (monolayer, bilayer, bulk) and photon energy, and resolve the time-dependent projected density of states into orbital channels and identify the microscopic pathways of carrier injection. For out-of-plane linearly polarized light, the charge transfer is suppressed in the monolayer, weak in the bilayer, and strong in the bulk. Under in-plane polarization and a photon energy slightly above the DFT band gap, we identify a direct interband excitation pathway from the N p_z to B p_z orbitals. At higher photon energy, the bulk response is dominated by a local charge redistribution within N p_x-orbitals. Circularly polarized light induces a notable orbital angular momentum, while spin contributions remain negligible.

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O 14.9 Mon 17:15 TRE/MATH

Theory of resonantly enhanced light-induced superconducting pairing in K_3C_{60} — •Juan I. Aranzadi[1], Joseph Tindall[2], Paul Fadler[1], and Michael A. Sentef[1,3] — [1]Institute for Theoretical Physics, University of Bremen/ BCCMS — [2]Center for Computational Quantum Physics, Flatiron Institute, New York, New York 10010, USA — [3]MPSD, CFEL, Hamburg

K_3C_{60} exhibits unconventional s-wave superconductivity with a critical temperature of approximately 20K, where the superconducting state emerges from a balance between strong electronic correlations and Jahn-Teller phonon-mediated pairing [1].

Recent pump-probe measurements have shown that optical excitation induces optical signatures reminiscent of the equilibrium superconducting state at temperatures higher than Tc [2, 3]. Notably, a 10 THz resonance was observed to trigger metastable superconducting-like properties that persist for temperatures up to 300 K [4].

The mechanism underlying this non-equilibrium superconducting state remains poorly understood. To address this question, we investigate a driven three-orbital Hubbard-Kanamori model which serves as a minimal model for equilibrium superconductivity in K_3C_{60} using numerical techniques including exact diagonalization and tensor networks.

[1] Nomura, Y., et al. (2015). *Science Advances*, **1**
[2] Mitrano, M. et al. (2016). *Nature*, **530**, 461–464.
[3] Budden, M. et al. (2021). *Nature Physics*, **17**, 611–618.
[4] Rowe, E. et al. (2023). *Nature Physics*, **19**, 1821–1826.

O 14.10 Mon 17:30 TRE/MATH

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 14.11 Mon 17:45 TRE/MATH

Temporal Advancement of Photoelectrons emitted via Evanescent Waves — •Luis Maschmann, Andreas Gebauer, Tillmann Schabbehard, and Walter Pfeiffer — Bielefeld University, Universitätsstr. 25, 33615 Bielefeld

Photoelectron emission dynamics in solids is governed both by propagating and evanescent wave components. Here we employ a one-dimensional Kronig-Penney model to investigate the role of evanescent waves on the photoelectron wave packet dynamics based on time-reversed LEED states within one-step theory and time-dependent Schrödinger equation simulations. Using a Kronig-Penney potential with a large bandgap, we can clearly distinguish the dynamics for evanescent and propagating photoelectron emission channels. For emission via evanescent states, i.e., within the final state bandgap, the photoelectron appears temporally advanced and seemingly originates from the bulk-vacuum interface, although the emitter well is located well below the interface. Our results establish a direct analogy to the temporal evolution of a wave packet undergoing quantum mechanical tunneling through a potential barrier and significantly influence the interpretation of attosecond time-resolved photoemission delays.