O 7: Focus Session Ultrafast Electron Microscopy at the Space-Time Limit I

Shaping functionalities on the nanoscale is one of the most essential challenges in modern condensed matter research. It requires a comprehensive understanding of the complex interplay of the electronic, spin, and lattice degrees of freedom in materials and requires tailoring energy transfer and dissipation pathways on the smallest length and fastest timescales. Recent instrumentation breakthroughs in different varieties of pump-probe ultrafast electron microscopy have opened the way for accessing electronic and structural dynamics at surfaces, interfaces, and nanostructures with down-to-attosecond resolution in time. While ultrafast photoemission electron microscopy techniques provide supreme sensitivity to spin and electron dynamics in real momentum space, bright ultrashort electron pulses in the ultrafast implementation of more traditional electron microscopes can probe optical states, local magnetization, and lattice dynamics with a nanometer spatial resolution.

This focus session highlights recent advances in ultrafast high-resolution electron probing. These include new instrumentation and techniques, excitations from the THz to X-ray regime, and studying novel phenomena and materials systems. At the same time, it will bring together researchers from the different areas of ultrafast condensed matter physics to foster discussions and new collaborations to explore emergent scientific questions in this field.

Organized by
Armin Feist (MPI Göttingen) and Benjamin Stadtmüller (University Augsburg).

Time: Monday 10:30–12:45 Location: H11

O 7.1 Mon 10:30 H11

Ultrafast Low-Energy Electron Microscopy — •Johannes Otto[1,2,3], Leon Brauns[1,2], Benjamin Schröder[1,2], and Claus Ropers[1,2,3] — [1]Department of Ultrafast Dynamics, Max Planck Institute for Multidisciplinary Sciences, Göttingen, Germany — [2]4th Physical Institute, University of Göttingen, Göttingen, Germany — [3]Max Planck School of Photonics

Low-Energy Electron Microscopy (LEEM) allows for imaging the first atomic layers of a surface with nanometer resolution by reflecting a low-energy electron beam [1]. This contribution reports on the first implementation and initial results of Ultrafast LEEM. We replaced the electron source of a conventional instrument with a laser-triggered tip-shaped photoemitter enabling imaging with nanometer spatial and picosecond temporal resolution [2]. We show first real-space dynamics including a thermally-induced intensity suppression (transient Debye-Waller effect) and strain-wave propagation. Additionally, we report on stimulated inelastic electron-light scattering (IELS) at beam energies below 100 eV, as recently proposed theoretically [3]. The demonstrated capabilities of the instrument open up new possibilities to investigate a wide range of dynamical phenomena at surfaces.

[1] W. Telieps and E. Bauer, Surface Science 162, 163 (1985).
[2] A. Feist et al., Ultramicroscopy 176, 63 (2017).
[3] A. P. Synanidis et al., Sci. Adv. 10, eadp4096 (2024).

O 7.2 Mon 10:45 H11

Ultrafast Electron Dynamics in Surface Plasmon Polariton Nanofoci — •Pascal Dreher, Alexander Neuhaus, Michael Horn-von Hoegen, and Frank Meyer zu Heringdorf — Faculty of Physics and Center for Nanointegration, Duisburg-Essen (CENIDE), University of Duisburg-Essen, Germany

Surface plasmon polaritons (SPPs) are collective wave-like excitations of the electron system of a metal surface that hold great potential for enhancing light-based energy conversion processes. Such enhancement is based on the efficient generation of highly-excited hot electrons via the decay of SPPs on a femtosecond timescale. To understand the corresponding microscopic dynamics, it is essential to gain a direct view into the plasmonically-generated hot electron distributions and to distinguish them from optically-generated hot electrons.

We achieve these goals by combining topologically-robust SPP nanofoci with time- and angle-resolved photoelectron spectroscopy in a photoemission microscope. This approach allows us to investigate the ultrafast non-equilibrium electron dynamics driven by SPPs on the native length-, momentum-, time-, and energy scales. We observe plasmonically-driven above-threshold electron emission, and ponderomotive shifts of the electron emission spectra provide us with a direct measure of the local SPP field strength. For different exemplary metal surfaces with distinct surface band structures we gain a direct view into the hot electron distributions generated via the coherent and incoherent decay of SPPs.

O 7.3 Mon 11:00 H11

Mechanisms and Dynamics of Electron Emission from Graphitic Surfaces: Insights from Correlated and Time-Resolved Spectroscopies — •Alessandra Bellissimo[1], Florian Simperl[1], Felix Blödorn[1], Wolfgang S.M. Werner[1], Gyula Halasi[2], László Óvári[2], Csaba Vass[2], Nikolett Oláh[2], Zoltán Filus[2], Tímea Grósz[2], Chinmoy Biswas[2], Balázs Major[2], Imre Seres[2], Aref Imani[1], Paolo A. Carpeggiani[1], Maosheng Hao[1], and Florian Libisch[1] — [1]TU WIEN, Vienna, Austria — [2]ELI-ALPS, Szeged, Hungary

The electron emission behaviour of graphitic surfaces was investigated using advanced spectroscopic methods alongside static & time-resolved Photo-Electron Emission Microscopy (PEEM). At TU WIEN, electron-pair coincidence spectroscopy on pyrolytic graphite detected *correlated* electron pairs from single scattering events, directly linking energy-loss processes to the secondary electron (SE) spectrum. The $(\pi+\sigma)$-plasmon was resolved in terms of the involved interband transitions revealing strong final-state resonances in the SE spectrum. Static photoemission from graphite(0001) was studied using PEEM at the NanoESCA end station at ELI-ALPS, employing extreme ultraviolet linearly polarised photons generated via High-Harmonic Generation in Argon. The k-space-resolved photoelectron (PE) signal associated to the above-mentioned final-state resonances in the SE-spectrum reflects the symmetry of the conduction bands involved in the PE-emission process. Attosecond time- and k-space-resolved RABBITT measurements provided insights into PE-emission dynamics across the Brillouin zone.

Invited Talk O 7.4 Mon 11:15 H11

Nanoimaging the electronic, plasmonic, and phononic structure and dynamics of 2D materials — •Sarah King — University of Chicago, Chicago, IL, United States

Heterogeneity plays a critical role in chemistry and physics, from the role of defect states in the carrier dynamics of semiconductors to interfaces and surfaces in catalysis. However, our ability to visualize nano-scale domains and properties in materials and their effect on material dynamics has been hampered by the simple challenge of our inability to meet the necessary nanometer and femtosecond timescales. I will discuss recent efforts by my group to determine the interplay of heterogeneity and morphology on the intrinsic optoelectronic and thermoelectric properties of materials. Using polarization-dependent photoemission electron microscopy (PD-PEEM) we have imaged the spatially dependent optical selection rules of black phosphorus, distinguishing edge-specific modes, and antiferroelectric domains of β'-In2Se3, with spatial resolution as good as 25 nm. Through ultrafast transmission electron microscopy, we've been able to determine how the bond anisotropy and structural morphology of few-layer black phosphorus impacts phonon dynamics. Ultimately my group seeks to identify ways to modify the impact of structural heterogeneity in materials and rationally design energy efficient interfaces on the nanoscale.

O 7.5 Mon 11:45 H11

Time-resolved momentum microscopy with fs-XUV photons at high repetition rates with flexible energy and time resolution — •Karl Schiller[1], Lasse Sternemann[1], Matija Stupar[1], Alan Omar[2], Martin Hoffmann[2], Jonah Nitschke[1], Valentin Mischke[1], David Janas[1], Stefano Ponzoni[1,3], Giovanni Zamborlini[1,4], Clara Saraceno[2], and Mirko Cinchetti[1] — [1]TU Dortmund University, Dortmund, Germany — [2]Ruhr University Bochum, Germany — [3]Ecole Polytechnique, Paris, France — [4]Karl-Franzens-University Graz, Austria

We present a versatile setup for time-resolved ARPES that combines an energy-filtered momentum microscope with a custom-designed high-harmonic generation (HHG) photon source [1]. The HHG source is powered by a commercial multi-100 kHz Yb-based ultrafast laser system delivering femtosecond pulses in the extreme ultraviolet range. A nonlinear pulse compression stage, utilizing spectral broadening in a Herriott-type bulk multi-pass cell, enables flexible control of the driving pulse duration. This adaptability allows two distinct operational modes, optimized for either energy or time resolution, making the setup highly suitable for ultrafast photoelectron microscopy at the space-time limit. We demonstrate the system's capabilities by tracking conduction band dynamics in the valleys of a bulk WS_2 crystal. Using uncompressed laser pulses, we achieve an energy resolution better than (107 ± 2) meV. Compressed pulses, in contrast, yield a time resolution of (48.8 ± 17) fs.

[1] Optica Open Preprint 115282 (2024)

O 7.6 Mon 12:00 H11

Dark field photoelectron momentum microscopy of electric field gated 2D semiconductors — •Jan Philipp Bange[1], Bent van Wingerden[1], Jonas Pöhls[1], Wiebke Bennecke[1], Paul Werner[1], David Schmitt[1], AbdulAziz AlMutairi[3], Daniel Steil[1], R. Thomas Weitz[1], G. S. Matthijs Jansen[1], Stephan Hofmann[3], Giuseppe Meneghini[2], Samuel Brem[2], Ermin Malic[2], Marcel Reutzel[1], and Stefan Mathias[1] — [1]Georg-August-Universität Göttingen, Germany — [2]Philipps-Universität Marburg, Germany — [3]University of Cambridge, U.K.

A possibility to tune many-body interactions in two-dimensional semiconductors is in-situ electric field gating, which allows precise and reversible control of the filling of states in a moiré potential. In combination with ARPES for static band structure measurements, this approach has been shown to be a powerful experimental probe [1]. However, the study of excited states in gated 2D material structures, such as interlayer excitons [2] and trions, has so far remained elusive.

Here we combine time-resolved momentum microscopy with dark field imaging techniques to gain access to many-body interactions on femtosecond time and nanometer lengthscales [3]. We use this method to study electric field gated homobilayer WSe_2 and report the ultrafast formation of quasiparticles as a function of applied gate voltage.

[1] Nguyen *et al.*, Nature **572**, 220 (2019).
[2] Bange *et al.*, Science Advances **10**, eadi1323 (2024).
[3] Schmitt *et al.*, Nature Photonics, in press, arXiv.2305.18908.

O 7.7 Mon 12:15 H11

Plasmonic spin meron pair: Spatio-temporal topology revealed by time resolved polarimetric photo-emission microscopy — Pascal Dreher[1], •Alexander Neuhaus[1], David Janoschka[1], Alexandra Roedl[1], Tim Meiler[2], Bettina Frank[2], Timothy J. Davis[1,2,3], Harald Giessen[2], and Frank Meyer zu Heringdorf[1] — [1]Faculty of Physics and Center for Nanointegration, Duisburg-Essen (CENIDE), University of Duisburg-Essen, 47048 Duisburg, Germany — [2]4th Physics Institute and Research Center SCoPE, University of Stuttgart, 70569 Stuttgart, Germany — [3]School of Physics, University of Melbourne, Parkville Victoria 3010, Australia

We have developed a novel method, polarimetric photo-emission electron microscopy (polarimetric PEEM), which combines an optical pump-probe polarimetry with photo-emission electron microscopy. This method enables the accurate generation and measurement of surface plasmon polariton fields at deep sub-wavelength spatial resolution and sub-cycle temporal resolution. Using polarimetric PEEM, we extend the study of electromagnetic fields on surfaces to a spin quasiparticle with the topology of a meron pair and analyze its topology by calculating the Chern number. We find the Chern number to be C=1 and constant over time, demonstrating the stability of the plasmonic meron pair on a femtosecond time scale. Additionally, we show that the in-plane vectors of the three-dimensional field are constrained by the embedding topology of the space as dictated by the Poincare-Hopf theorem.

O 7.8 Mon 12:30 H11

Momentum microscopy with attosecond time resolution at ELI ALPS to map the full Brillouin zone — Gyula Halasi[1], Csaba Vass[1], Nikolett Oláh[1], Zoltán Filus[1], Tímea Grósz[1], Chinmoy Biswas[1], Tamás Csizmadia[1], Lénárd Gulyás Oldal[1], Balázs Major[1], Péter Jójárt[1], Felix Blödorn[2], Florian Simperl[2], Aref Imani[2], Paolo Carpeggiani[2], Péter Dombi[1], Wolfgang S.M. Werner[2], Alessandra Bellissimo[2], and •László Óvári[1] — [1]ELI ALPS, Szeged, Hungary — [2]Vienna University of Technology, Austria

Time-resolved photoemission is a highly efficient tool for unraveling surface electron dynamics. The coupling of a photoemission electron microscope (PEEM) with an imaging hemispherical analyzer in our NanoESCA end station allows for the combination of spectroscopy and microscopy in real or momentum space. The NanoESCA end station is attached to a high harmonic generation (HHG) beamline, hence time-resolved momentum microscopy studies can be performed in a pump-probe scheme. To illustrate the performance of our user-ready system, the first results obtained by the RABBITT (Reconstruction of Attosecond Beating By Interference of Two-photon Transitions) scheme are presented. In this experiment, a graphite(0001) single crystal was studied, and RABBITT oscillations have been identified throughout the whole Brillouin zone, with attosecond precision, in the form of a time-resolved momentum space movie.