O 10: Focus Session Ultrafast Electron Microscopy at the Space-Time Limit II

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 15:00–17:45 Location: H2

Invited Talk O 10.1 Mon 15:00 H2

Probing coherent optical emission processes with ultrafast scanning electron microscopy — •Albert Polman — NWO Institute AMOLF, Amsterdam, the Netherlands

High-energy electron beams are unique probes of optical materials properties as their time-varying electric field can create strong materials polarizations. The subsequent light emission (cathodoluminescence, CL) provides a fingerprint of the local optical density of states at the nanoscale.

CL from plasmonic and dielectric nanostructures has a coherent phase relation with the excitation process, which enables self-referenced measurements to perform holography and metrology. Electron excitation of semiconductors creates a sequence of fs-ps-ns materials excitations, that lead to bunched CL photon emission.

New developments in ultrafast electron microscopy enable the creation of picosecond electron pulses and pump-probe spectroscopy where light and electrons serve as pump and probe or vice versa. The interaction of pulsed electrons with optical metasurfaces enables novel ways to shape the quantum mechanical electron wavepackets in space and time and may eventually create entirely new forms of ultrafast materials spectroscopy.

O 10.2 Mon 15:30 H2

Spectrally resolved free electron-light coupling strength in a transition metal dichalcogenide — •Soufiane El kabil[1], David Lerchenberger[1], Niklas Müller[1], Jonathan Weber[1], Alexander Schröder[1], and Sascha Schäfer[1,2] — [1]University of Regensburg, Regensburg, Germany — [2]Regensburg Center for Ultrafast Nanoscopy, Regensburg, Germany

In ultrafast transmission electron microscopy (UTEM), combining precisely controlled free-electron beams with localized light fields enables the creation of intricate electronic states and the visualization of transient optical near-fields via PINEM [B. Barwick, et al. Nature 462.7275 (2009): 902-906]. However, optical nearfields in photonic structures typically exhibit a strong wavelength dependence, which has so far only been partially captured by PINEM approaches.

To address this, we use strongly chirped broadband light pulses to explore the spectrally resolved interaction between free electrons and light at the edge of a MoS2 thin film [N. Müller, et al. arXiv preprint arXiv:2405.12017(2024).]. As a fast electron traverses the optical field near the MoS2 flake, it absorbs or emits multiple photons, producing photon sidebands in its energy spectrum. By varying the electron-light delay at the sample, different spectral components of the near-field can be investigated. Numerical simulations reveal that the observed spectral and spatial modulations stem from interactions between incident and reflected light fields, as well as guided thin-film optical modes. Our results highlight the ability of PINEM to resolve the optical properties of semiconductors spatially and spectrally.

O 10.3 Mon 15:45 H2

Simulating Quantum Spin Dynamics in Transmission Electron Microscopy — •Santiago Beltrán Romero[1,2], Dennis Rätzel[3], Stefan Löffler[2], and Philipp Haslinger[1,2] — [1]VCQ, Atominstitut, TU Wien, Stadionallee 2, 1020 Vienna, Austria — [2]University Service Centre for Transmission Electron Microscopy,TU Wien, Wiedner Hauptstraße 8-10/E057-02, 1040 Wien, Austria — [3]ZARM, Unversität Bremen, Am Fallturm 2, 28359 Bremen, Germany

Transmission Electron Microscopy (TEM) has revolutionized nanoscale research by enabling unprecedented simultaneous spatial and temporal resolutions, thanks to advancements such as aberration correction, cryogenic techniques, and ultra-fast probing. However, the capabilities of TEM to probe spin dynamics – critical for understanding quantum materials – are, to date, quite limited and could be significantly improved by novel microwave spectroscopic tools [1, 2]. Building on recent innovations in that direction, we present a framework that integrates scattering theory and multislice simulations to describe the probing of spin samples on the nanoscale with time-resolved TEM. Our simulations offer insights into both elastic and inelastic processes - including the electrons backaction on the spin. They reveal how the choice of set-up parameters influence the precision of spin detection, identifying optimized conditions for enhancing the signal-to-noise ratio (SNR) and contrast. This work sets the stage for combining spin resonance tools with cutting-edge TEM capabilities, paving the way for breakthroughs in spin imaging and manipulation at the atomic level.

O 10.4 Mon 16:00 H2

Ultra-Nonlinear Subcycle Photoemission of Few-Electron States from Sharp Gold Nanotapers — •Germann Hergert, Rasmus Lampe, Andreas Wöste, and Christoph Lienau — Institut für Physik, Carl-von-Ossietzky Universität, 26129 Oldenburg, Germany

Generating attosecond electron pulse trains by coherent modulation of swift electrons enabled attosecond resolution in ultrafast transmission electron microscopy [1,2]. The possibility to transfer photon statistics to the electron number statistics in multiphoton photoemission (MPP) from nanotapers [3], opens up a window to increase photoemission nonlinearities of few-electron states and generating subcycle electron pulses. This provides an alternative road to reach subcycle resolution in electron microscopy.

Here, we present MPP of few-electron wavepackets triggered by near-infrared pulses from gold nanotapers, demonstrating 20th-order nonlinearities for electron triplets. Event-based interferometric autocorrelations of the photoemission yield are quenched to single-peak traces with 0.8 fs duration. We observe a modulation of the electron yield by the carrier-envelope phase, indicating the emission of subcycle isolated electron beams, with prospects to improve the temporal resolution in ultrafast point-projection electron microscopy.

[1] D. Nabben, Nature, 619, 63 (2023)
[2] J. Gaida, Nat. Photon., 18, 509 (2024)

[3] J. Heimerl, Nat. Phys., 20, 945 (2024)
[4] G. Hergert, Nano Lett., 24, 11067 (2024)

Invited Talk O 10.5 Mon 16:15 H2

Ultrafast exciton dynamics in momentum space — Alexander Neef[1], Tommaso Pincelli[1,2], Lawson Lloyd[1], Shuo Dong[1], Samuel Beaulieu[1], Tania Mukherjee[1,2], Sebastian Hammer[3], Malte Selig[2], Dominik Christiansen[2], Andreas Knorr[2], Martin Wolf[1], Jens Pflaum[3], Laurenz Rettig[1], and •Ralph Ernstorfer[1,2] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, 14195 Berlin, Germany — [2]Technische Universität Berlin, 10623 Berlin, Germany — [3]Julius-Maximilian-Universität Würzburg, 97070 Würzburg, Germany

Time- and angle-resolved photoemission spectroscopy (trARPES) provides a quantum-state-resolved picture of the ultrafast dynamics of many-body states like excitons in non-equilibrium states of matter. Following the formation and scattering of excitons in momentum space in real time reveals all key properties of the excitons like binding energy, exciton-phonon coupling, and the real-space distribution of the many-body wave functions. Additionally, information about the orbital properties and Berry curvature is encoded in the multidimensional trARPES signals. Applied to heterostructures, the ultrafast exciton and charge dynamics across interfaces reveal the mechanism of charge and energy transfer processes. We will exemplify this approach for transition metal dichalcogenides heterostructures, molecular crystals, and layered semiconducting antiferromagnets.
References: S. Dong et al., Nature Commu. 14, 5057 (2023); T. Pincelli et al., Adv. Mater. 2209100 (2023), A. Neef et al., Nature 616, 275 (2023), S. Beaulieu et al., Sci. Adv. 10, eadk3897 (2024).

O 10.6 Mon 16:45 H2

Subcycle band-structure videography of quantum materials — •Vincent Eggers[1], Manuel Meierhofer[1], Jakob Helml[1], Lasse Münster[1], Robert Wallauer[2], Giacomo Inzani[1], Sarah Zajusch[2], Suguru Ito[2], Leon Machtl[1], Yin Hao[3], François C. Posseik[3], Changhua Bao[1], Jens Güdde[2], F. Stefan Tautz[3], Rupert Huber[1], and Ulrich Höfer[1,2] — [1]Department of Physics and Regensburg Center for Ultrafast Nanoscopy, University of Regensburg, 93040 Regensburg, Germany — [2]Department of Physics, Philipps-Universität Marburg, 35037 Marburg, Germany — [3]Peter Grünberg Institut (PGI-6), Forschungszentrum Jülich GmbH, 52428 Jülich, Germany

We introduce the next generation of subcycle band-structure videography. By combining atomically strong few-cycle mid-infrared lightfields with sub-10-femtosecond XUV pulses in a momentum microscope, lightwave-driven dynamics can now be investigated throughout the entire Brillouin zone. Here, we observe electrons driven by carrier fields of light reaching amplitudes as high as MV/cm in graphene. Subcylce analysis of the timing of these lightwave-driven currents reveals femtosecond scattering times. Our novel setup provides a new platform to explore strong-field phenomena ranging from inter- and intraband dynamics to Bloch oscillations and the emergence of Floquet-Bloch states directly in subcycle videos covering the full band structure.

O 10.7 Mon 17:00 H2

Approaching Atomic Resolution in Ultrafast Transmission Electron Microscopy — •Sophie Schaible[1,2], Till Domröse[1,2], and Claus Ropers[1,2] — [1]Max Planck Institute for Multidisciplinary Sciences, Göttingen, Germany — [2]4th Physical Institute, University of Göttingen, Germany

Ultrafast transmission electron microscopy (UTEM) [1] extends the study of structural heterogeneity in conventional TEM by introducing femtosecond temporal resolution, providing the means to map structural phase transitions at the nanoscale. However, access to atomic-scale ultrafast dynamics remains a major challenge due to the limited brightness of pulsed photoelectron beams. In this contribution, we explore approaches to atomic-resolution imaging of a structural transformation in a UTEM employing a high-coherence photoelectron source. Highly dose-efficient imaging is crucial to make optimum use of the available electron signal. We further gauge the impact of experimental parameters on the achievable spatiotemporal resolution such as sample drift, acquisition time, repetition rate and electron pulse length with and without optical excitation of the specimen.
[1] Feist *et al.* Ultramicroscopy **176** (2017)

O 10.8 Mon 17:15 H2

Towards Detection of Spin Resonance Excitations with TEM — •Antonín Jaroš, Johann Toyfl, Benjamin Czasch, Michael Stanislaus Seifner, Isobel Claire Bicket, Santiago Belrán-Romero, and Philipp Haslinger — VCQ, Atominstitut, TU Wien, USTEM, Stadionallee 2, 1020 Vienna, Austria

Microwave (MW) excitations of spin systems induce precessional spin motion at GHz frequencies. Traditional spin resonance spectroscopy techniques, such as Electron Spin Resonance (ESR) and Ferromagnetic Resonance (FMR), are employed to determine key parameters like gyromagnetic ratios and damping constants in magnetic materials. However, these methods often lack the spin sensitivity and spatial resolution required for spin studies at the atomic level. We present a novel approach that synergistically combines spin resonance techniques with Transmission Electron Microscopy (TEM). Spin state polarization is induced by the magnetic field of the TEM pole piece, while spin system excitation is achieved through an impedance-matched micro-resonator integrated into a custom-designed sample holder. The detection of spin resonance excitations in TEM might represent an important step towards MW driven spin studies with highly controlled electron probe at the nanoscale.

O 10.9 Mon 17:30 H2

Laser-driven cold-field emission source for ultrafast transmission electron microscopy — Alexander Schröder[1], •Andreas Wendeln[1,2], Jonathan Weber[1,2], Masaki Mukai[3], Yuji Kohno[3], and Sascha Schäfer[1,4] — [1]Department of Physics, University of Regensburg, Regensburg, Germany — [2]Insitute of Physics, Carl-von-Ossietzky Universität Oldenburg, Oldenburg, Germany — [3]JEOL Ltd., Tokio, Japan — [4]Regensburg Center for Ultrafast Nanoscopy (RUN), Regensburg, Germany

In recent years ultrafast transmission electron microscopy (UTEM), which combines the nanometer spatial resolution of a TEM with the femtosecond temporal resolution of a pump-probe approach, has become an increasingly important tool for investigating nanoscale dynamics. Further improving the spatio-temporal resolution in time-resolved electron imaging experiments requires femtosecond photoelectron sources with a higher degree-of-coherence. Here, we present the development of a laser-driven cold field electron source integrated in a UTEM instrument [1]. This approach yields 220-fs electron pulses with electron energy widths down to 360 meV, photoelectron spot sizes of 2 Å, and a peak normalized beam brightness exceeding $6.5 \cdot 10^{13}$ A/m^{2}sr, providing a new level of spatial and spectral precision in observing ultrafast nanoscale dynamics for UTEM applications. Lastly, we discuss the implementation of laser-driven cold-field emitters in a probe-aberration-corrected electron microscope potentially leading to smaller spot sizes with less coherent beams and significantly increased electron currents. [1] Schröder et al., arXiv:2410.23961, (2024).