O 26: Focus Session Ultrafast Electron Microscopy at the Space-Time Limit III

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: Tuesday 10:30–13:00 Location: H2

Invited Talk O 26.1 Tue 10:30 H2

Attosecond Electron Microscopy — •Peter Baum — Universität Konstanz, Germany

All processes in materials, nanostructures and devices are on a fundamental level defined by electronic and atomic motion from initial to final conformations. Our approach for a direct, real-space visualization is pump-probe electron diffraction and microscopy with single-electron wavepackets under the control of laser light. The resulting few-femtosecond and attosecond time resolution allows to see almost any light-matter interaction or structural dynamics on fundamental scales in space and time. We report selected results on strongly correlated materials, rotational phonons, electronic circuitry, free-electron quantum phenomena and attosecond dynamics in nanomaterials.

O 26.2 Tue 11:00 H2

Steady-State and Time-Resolved Cathodoluminescence of III-Nitride Semiconductors — •Kagiso Loeto, Aidan Flynn Campbell, Domenik Spallelk, and Jonas Lähnemann — Paul-Drude-Institut für Festkörperelektronik, Berlin, Deutschland

Cathodoluminescence (CL), in steady-state and time-resolved modes, has advanced the study of semiconductor optical properties, crucial for microelectronics and III-nitride optoelectronics. A new state-of-the-art time-resolved CL (TRCL) microscope at the Paul-Drude-Institut features a high-performance SEM with a stable electron source and advanced light collection system, enabling optimized imaging and high spatial resolution at acceleration voltages as low as 0.35 kV. The system features a UV-optimized CCD camera for studying UV-emitting materials like III-nitrides. Time-resolved operation is enabled by an ultrafast beam blanker paired with detectors achieving temporal resolutions of tens of picoseconds, offering new insights into the dynamic optical properties of advanced semiconductors. It will be employed in three focus areas, highlighting its distinct capabilities. First, very-low acceleration voltage operation will enable high-resolution mapping of individual point defects in AlGaN quantum well structures, revealing their impact on AlGaN-based UV LEDs. Second, ultraviolet-optimized photon detectors will study temperature-stable excitonic bands in AlN with high spectral resolution, providing insights into their origins. Lastly, the instrument's time-resolved capabilities combined with spatial mapping will explore the interplay between carrier dynamics and localization in InGaN pseudosubstrates.

O 26.3 Tue 11:15 H2

Spin Resonance Spectroscopy meets Transmission Electron Microscopy — •Philipp Haslinger — Atominstitut, USTEM, Technische Universität Wien, Austria

Coherent spin resonance methods such as nuclear magnetic resonance and electron spin resonance spectroscopy have led to spectrally highly sensitive, non-invasive quantum imaging techniques. Here, we will present a spin resonance spectroscopy approach developed for transmission electron microscopy [1,2] and will explain different techniques to sense with electrons for microwave manipulated spin states of the sample. This could enable state-selective observation of spin dynamics on the nanoscale and indirect measurement of the environment of the spin systems, providing information on, for example, atomic structure, local chemical composition and neighbouring spins.

[1] P. Haslinger, S. Nimmrichter, and D. Rätzel, Spin Resonance Spectroscopy with an Electron Microscope, Quantum Sci. Technol. 9, 035051 (2024). [2] A. Jaroš, J. Toyfl, A. Pupić, B. Czasch, G. Boero, I. C. Bicket, and P. Haslinger, Electron Spin Resonance Spectroscopy in a Transmission Electron Microscope, arXiv:2408.16492 (2024).

O 26.4 Tue 11:30 H2

Observation of Kapitza-Dirac effect with fast electrons — •Kamila Moriová[1], Petr Koutenský[1], Marius Constantin Chirita Mihaila[1], Zbyněk Šobáň[2], Andreas Schertel[3], Jaromír Kopeček[2], and Martin Kozák[1] — [1]Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic — [2]Institute of Physics, Czech Academy of Sciences, Prague, Czech Republic — [3]Carl Zeiss AG, Oberkochen, Germany

Advancing ultrafast electron microscopy relies on coherent control of free electron wavefunctions. While most research focuses on electron interactions with optical near-fields, an all-optical approach using ponderomotive forces offers a promising alternative for manipulating pulsed electron beams. The Kapitza-Dirac effect [1], where free electrons diffract coherently from a standing light wave, enables momentum transfer via stimulated Compton scattering. However, its application has been limited to low-energy electrons due to challenges in resolving small deflection angles of electron beams caused by photon absorption and emission.

We report the observation of the Kapitza-Dirac effect in a scanning electron microscope using high-energy (20 keV) electrons. Photon sidebands in the electron transverse momentum spectrum are detected in a convergent beam diffraction geometry using spatial filtering. This effect can serve as a coherent electron beam splitter or phase plate in various types of electron microscopes and paves the way for exploring fundamental electron-light interactions.

[1] Freimund, D. L. et al. Nature 413, 142 (2001)

O 26.5 Tue 11:45 H2

Recent results of the ultrafast scanning electron microscope in Erlangen — •Stefanie Kraus[1], Tomas Chlouba[1], Roy Shiloh[1], Leon Brückner[1], Julian Litzel[1], Zhexin Zhao[1], Viacheslav Korolev[1], Manuel Konrad[1], Tatsunori Shibuya[1,3], and Peter Hommelhoff[1,2] — [1]Department Physik, Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU), 91058 Erlangen — [2]Department Physik, Ludwig-Maximilians-Universität München (LMU), 80799 München — [3]AIST, Tsukuba, Japan

Ultrafast electron microscopy is revolutionizing the capabilities of electron microscopes, allowing for unprecedented spatial and temporal

resolution. Closely related is the coupling of electrons and optical nearfields, which is based on the excellent electron pulse properties an ultrafast scanning electron microscope (USEM) can provide. We have leveraged the interaction of electrons with the near field of a periodic structure to achieve sub-femtosecond electron pulse compression as well as laser acceleration of electrons. This advancement enhances temporal resolution and facilitates large beam energy variations, enabling detailed investigations of ultrafast dynamics. In this talk, we will provide an overview of our recent progress, including electron energy modulation in larger structures illuminated with 10 micrometer light as to enhance the current throughput, electron bunch compression, and the latest experimental results.

O 26.6 Tue 12:00 H2

FEL-based core-cum-conduction momentum microscopy of ultrafast charge-density-wave dynamics — •N. Wind[1,2,3], M. Heber[1,3], D. Kutnyakhov[1], L. Wenthaus[1], J. Dilling[2], L. Bruckmeier[2], S. Chernov[1], O. Tkach[4], A. Mehta[5], J. Koralek[5], G. Dakovski[5], J.A. Sobota[6], P.E. Majchrzak[6], D. Puntel[6], D. Liu[6], G. Schönhense[4], H.J. Elmers[4], Z.X. Shen[6], M. Scholz[1], and K. Rossnagel[1,2] — [1]Deutsches Elektronen Synchrotron DESY, 22607 Hamburg, Germany — [2]Christian-Albrechts-Universität zu Kiel IEAP, 24098 Kiel, Germany — [3]Universität Hamburg, IExP, 22761 Hamburg, Germany — [4]Johannes Gutenberg-Universität, Institut für Physik, 55128 Mainz, Germany — [5]SLAC National Accelerator Laboratory, Menlo Park, CA 94205, USA — [6]Stanford University, Institute for Materials and Energy Science, CA 94305, USA

Transition-metal dichalcogenides (TMDCs) offer a rich platform for studying novel forms of quantum and nanoelectronics in layered structures approaching the 2D limit. Among them, 1-T TaS_2 has been extensively studied due to its various charge-density-wave (CDW) phases. Here, we provide novel insights into the CDW melting in 1-T TaS_2, using time- and angle-resolved photoemission spectroscopy with a momentum microscope at the free-electron laser FLASH in Hamburg. Our near-infrared pump–FEL probe experiment uncovers momentum-dependent conduction-band dynamics and core-level responses, advancing our understanding of ultrafast coupled electronic and structural dynamics.

O 26.7 Tue 12:15 H2

Developing a versatile fiber-based cathodoluminescence detection system for an ultrafast scanning electron microscope — •Paul H. Bittorf[1], Filip Majstorovic[1], and Nahid Talebi[1,2] — [1]Institute for Experimental and Applied Physics IEAP, Kiel University, 24118 Kiel, Germany — [2]Kiel Nano, Surface and Interface Science KiNSIS, Kiel University, 24118 Kiel, Germany

Cathodoluminescence (CL) is emitted when a high-energy electron beam interacts with materials like minerals, semiconductors and plasmonic nanoparticles. Depending on the underlying interaction mechanisms of electrons with the sample this radiation can be coherent or incoherent, where both spectral and temporal statistics can be unraveled for material characterization. Thanks to the high spatial resolution and large spectral excitation bandwidth of the electron beams, we could resolve the spatial far-field distribution of locally probed photonic modes by CL microscopy. Moreover, we combined a commercial scanning electron microscope (SEM) with an ultrafast laser system to obtain a pulsed electron beam via the photoemission process. In addition to the excitation by the pulsed electron beam, a time-delayed laser pulse is focused onto the sample to induce an optical near-field and achieve a time-resolved pump-probe measurement. The interaction properties of electrons with nanostructured matter are analyzed through the emitted CL. Here, we report on technical aspects and the implementation of a multimode fiber-based CL detection system inside an ultrafast SEM and highlight its functionality by performing CL spectroscopy and time correlated single-photon counting.

O 26.8 Tue 12:30 H2

Ultrafast Electron Diffraction and Microscopy of Structural Phase Transitions at Megahertz Rates — •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

Control over laser-induced structural phase transformations promises tuning of macroscopic materials properties on femtosecond timescales. Ultrafast electron diffraction (UED) elucidates the spatially averaged evolution of lattice symmetries and phonon populations during the transitions. However, resolving nanoscale structural heterogeneity in these measurements remains challenging due to the reduced brightness of pulsed electron beams. Here, we overcome fundamental limitations in the stroboscopic investigation of structural dynamics in thin material films by UED and ultrafast electron microscopy. A high-coherence electron source offers enhanced momentum resolution in collimated electron nanobeams, while thermally-optimized sample supports enable reversible driving of structural transitions at high duty cycles [1]. Utilizing the associated gain in coherent electron current, we conduct nano-UED investigations of charge-density wave dynamics in layered materials, tomographically reconstructing three-dimensional phase formation kinetics in 1T-TaS_2 [2], and revealing a femtosecond structural quench in 1T'-$TaTe_2$ cycled at a repetition rate of 2 MHz [3].

[1] T. Domröse, *et al.*, arXiv:2410.02310 (2024)
[2] T. Domröse, *et al.*, Nature Materials **22(11)** (2023)
[3] T. Domröse, C. Ropers, Physical Review B **110(8)** (2024)

O 26.9 Tue 12:45 H2

From Electron-Photon Ghost Imaging Towards Entanglement Certification — •Alexander Preimesberger[1,2], Sergei Bogdanov[1,2], Phila Rembold[1], Santiago Beltrán-Romero[1,2], Dominik Hornof[1,2], Isobel C Bicket[1,2], Nicolai Friis[1], Elizabeth Agudelo[1], Dennis Rätzel[3], and Philipp Haslinger[1,2] — [1]VCQ, Atominstitut, TU Wien, Vienna, Austria — [2]USTEM, TU Wien, Vienna, Austria — [3]ZARM, University of Bremen, Bremen, Germany

Time-resolved detection of single electrons and their associated cathodoluminescence (CL) photons enables the identification of coincident electron-photon pairs. We recently employed this technique to study the tight momentum correlations generated by coherent CL within a transmission electron microscope [1]. In this contribution, we demonstrate ghost imaging using electron-photon pairs in both near-field and far-field configurations. In photonic quantum optics, the ability to produce such images is used to investigate quantum entanglement in photon pairs [2]. We discuss how to translate this concept to electron-photon states and introduce a robust method to certify and quantify their entanglement using measurements in mutually unbiased bases.

[1] A. Preimesberger et al., arXiv:2409.12216 (2024). [2] R. S. Bennink et al., Phys. Rev. Lett. 92 (2004).