O 75: Focus Session: Ultrafast Dynamics in Nanostructures I

In nanostructures, the response of electrons and atoms to external stimuli often differs significantly from that of their bulk counterparts due to spatial confinement and greatly increased surface-to-volume ratios. Whereas pump-probe techniques provide real time access to the dynamics of nanostructures in excited states, investigating the response of single nanostructures and probing their dynamics locally requires ultrafast imaging techniques that provide simultaneously a high spatial and temporal resolution.

Several ultrafast nanoscale imaging methodologies have made a tremendous progress in recent years. Crucial instrumental developments have enabled time-resolved variants of scanning probe techniques, such as ultrafast scanning near-field optical microscopy and ultrafast scanning tunneling microscopy. Furthermore, bright sources for femtosecond electron as well as extreme ultraviolet and x-ray pulses now allow for the ultrafast far-field probing with nanometer resolution.

To highlight these recent developments, this focus session brings together researchers interested in ultrafast nanoscale science and in the development of ultrafast high-resolution imaging approaches.

Organizers: Melanie Müller (FHI Berlin) and Sascha Schäfer (U Regensburg)

Time: Thursday 10:30–13:15 Location: GER 38

Topical Talk O 75.1 Thu 10:30 GER 38

Ultrafast nano-imaging: probing quantum dynamics in space and time — •Markus Raschke — Department of Physics and JILA, University of Colorado, Boulder, USA

Understanding and ultimately controlling the properties of matter, from molecular to quantum systems, requires imaging the elementary excitations on their natural time and length scales. To achieve this goal, we developed scanning probe microscopy with ultrafast and shaped laser pulse excitation for multiscale spatio-temporal optical nano-imaging. In corresponding ultrafast movies we resolve the fundamental quantum dynamics from the few-femtosecond coherent to the thermal transport regime. I will discuss specific examples visualizing in space and time the nanoscale heterogeneity in competing structural and electronic dynamic processes that define the performance in perovskite photovoltaics or energy dissipation in 2D heterostructures. I will extend the discussion to new forms of photon-matter hybrid states that emerge from confining light on the nano- to atomic scale, with imaging in tip-enhanced strong coupling of single emitters, to new regimes of nonlocal and quantum nonlinear nano-optics.

O 75.2 Thu 11:00 GER 38

Towards a nanoscale ultrafast optical oscilloscope — •Abbas Chimeh, Sam Nochowitz, Tom Jehle, Juanmei Duan, Sven Stephan, and Christoph Lienau — Universität Oldenburg

Measuring the ultrafast time response on the nanoscale gives insights into the complex structure and dynamics of nanomaterials. A variety of all-optical and electron-based spectroscopy techniques are currently under development to probe such ultrafast dynamics [1].

Here, we propose and demonstrate a broadband, interferometric scattering-type scanning near-field optical spectroscopy technique to measure ultrafast response function on the nanoscale. The idea is to mix the weak optical near field that is scattered out from the gap of a laser-illuminated tip-sample junction with a broadband reference pulse in a single spatial mode. From the resulting phase-stable spectral interferogram, we can directly retrieve the linear optical response function of the coupled tip-sample system and, thus, the time structure of re-emitted electric field with sub-cycle precision. By recording scattering spectra at a rate much higher than typical tip-sample modulation frequencies, we can separate the near-field response from unmodulated signal backgrounds and directly probe the time structure of optical near-fields on the nanoscale. We demonstrate the technique by probing local light scattering from single TMDC monolayers. Our technique provides a general approach for measuring the response functions of nanostructures in the visible and near-infrared spectral range with femtosecond temporal and nanometer spatial resolution.

[1] P. Dombi et al., Rev. Mod. Phys., 92, 025003 (2020).

O 75.3 Thu 11:15 GER 38

Ultrafast electron dynamics in semiconductor nanowires — •Jan Vogelsang[1,2], Lukas Wittenbecher[1], Cord L. Arnold[1], Anne L'Huillier[1], and Anders Mikkelsen[1] — [1]Department of Physics, Lund University, 221 00 Lund, Sweden — [2]present address: Institut für Physik, Universität Oldenburg, 26129 Oldenburg, Germany

The dynamics and, in particular, the transport of charge carriers in semiconductor nanostructures after optical excitation are influenced by different processes whose interaction is not yet fully understood. Here, we employ ultrashort laser pulses in combination with a photoemission electron microscope (PEEM) to spatiotemporally resolve these processes. We observe the radial transport of charge carriers in semiconductor nanowires after impulsive optical excitation and investigate the influence of electron-hole scattering on the relaxation times of hot electrons. We conclude with a glimpse on an attosecond streaking experiment in a PEEM promising an even higher temporal resolution.

O 75.4 Thu 11:30 GER 38

SPP induced coherent electronic excitations investigated by Time- and Angle-Resolved Plasmoemission-Spectroscopy on Cs/Au(111) — •Alexander Neuhaus, Pascal Dreher, David Janoschka, Michael Horn-von Hoegen, and Frank-J. Meyer zu Heringdorf — Faculty of Physics, University of Duisburg-Essen, 47048 Duisburg, Germany

Non-perturbative interactions of intense light fields with the electronic band structure in a solid can result in transient electronic properties. The experimental conditions required to realize the necessary field strength can be realized in nano-optical systems, as these can be designed to provide tremendous enhancements of the local field amplitude. Ultimately, observing the non-equilibrium electron dynamics in such systems requires a combination of precise control over the local driving field, state resolution, and spatial selectivity. Here, we explore electron emission from nano-focused femtosecond surface plasmon polariton (SPP) pulses, providing us with deep-subwavelength spatial selectivity. Two-dimensional time- and angle-resolved photoelectron spectroscopy using a birefringent delay line provides us with direct access to the coherent and incoherent dynamics of the electronic excitations with attosecond precision. The technique is applied to the system Cs/Au(111), where we observe SPP induced coherent electronic excitations of the first image potential state.

Topical Talk O 75.5 Thu 11:45 GER 38

Lightwave-driven scanning tunneling microscopy and spectroscopy at the atomic scale — •Vedran Jelic[1], Spencer E. Ammerman[1], Yajing Wei[1], Vivian N. Breslin[1], Mohammed Hassan[1], Nathan Everett[1], Sheng Lee[1], Stephanie Adams[1], Trevor Hickle[1], Kaedon Cleland-Host[1], Qiang Sun[2], Carlo A. Pignedoli[2], Pascal Ruffieux[2], Roman Fasel[2,3], and Tyler L. Cocker[1] — [1]Department of Physics and Astronomy, Michigan State University, East Lansing, MI 48824, USA — [2]Empa Swiss Federal Laboratories for Materials Science and Technology, 8600 Dübendorf, Switzerland — [3]Department of Chemistry, Biochemistry and Pharmaceutical Sciences, University of Bern, 3012 Bern, Switzerland

Lightwave-driven scanning tunneling microscopy is a new technique for studying atomic-scale surfaces that exhibit sub-picosecond dynamics. By coupling free-space-propagating single-cycle terahertz pulses to an atomically sharp metal tip, lightwave-driven microscopy can achieve simultaneous sub-angstrom and sub-picosecond spatio-temporal resolution. We utilize terahertz scanning tunneling microscopy (THz-STM) to investigate seven-atom-wide graphene nanoribbons on an Au(111) surface and unveil highly localized wavefunctions that are inaccessi-

ble with conventional STM. Three-dimensional tomographic THz-STM imaging of the electron densities reveals a faster spatial decay of the valance band compared to the conduction band. Additionally, an algorithm is introduced for extracting the underlying differential conductance from THz-STS measurements in both steady-state and pump-probe scenarios.

O 75.6 Thu 12:15 GER 38

Probing hot electron dynamics in a metal nanotip with THz-STM — •Vivien Sleziona[1], Faruk Krecinic[1], Natalia Martín Sabanés[1,2], Fabian Schulz[1,3], Takashi Kumagai[1,4], Martin Wolf[1], and Melanie Müller[1] — [1]Fritz-Haber Institute, Berlin — [2]IMDEA Nanoscience, 28049 Madrid, Spain — [3]CIC NanoGUNE, San Sebastian, Spain — [4]Institute of Molecular Science, 444-8585 Okazaki, Japan

In metal nanostructures, such as the tip of a scanning tunneling microscope (STM), the excitation and dynamics of hot electrons can differ significantly from their behavior in bulk solids. However, how exactly the electrons in an STM tip respond to femtosecond (fs) laser excitation is still poorly understood, although this is important for optimizing and understanding fs-laser-excited STM operation. Here we use a THz-gated STM to study the ultrafast dynamics of photoexcited electrons in a tungsten STM tip [1]. Specifically, we investigate the role of thermalized hot electrons for the generation of ultrafast photocurrents and tunneling in STM. Knowledge of the THz voltage transient combined with simulation of the electron dynamics inside the confined volume of the STM tip allows us to extract the electronic temperature and its ultrafast decay. We evaluate the relevance for fs laser-excited STM and discuss how the tip geometry affects electronic heating on ultrafast time scales. [1] N. Martín Sabanés et al., ACS Nano 16, 9, 14479-14489 (2022)

O 75.7 Thu 12:30 GER 38

Spin current control by hybrid pulses — •Jyoti Krishna[1], Sangeeta Sharma[2], Julen Ibañez-Azpiroz[1,3], and Sam Shallcross[2] — [1]Centro de Fisica de Materiales (CSIC-UPV/EHU), 20018, San Sebastian, Spain — [2]Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie, Max-Born-Straße 2A,12489 Berlin — [3]Ikerbasque Foundation, 48013 Bilbao, Spain

Spin and valley indices represent the key quantum labels of quasiparticles in a wide class of two dimensional materials, and control over these degrees of freedom, the creation of valley and spin states as well as the generation of pure spin and valley currents, remains a central challenge in these fields. Here we show that hybrid femtoscond laser pulses combining optical frequency circularly polarized pulse and a terahertz frequency linearly polarized pulse, a so-called "hencomb" pulse, can generate currents, whose magnitude and direction can be precisely controlled by the THz envelope. We explore this effect both in bilayer graphene and the dichalocogenide WSe_2, finding control over substantial currents that are nearly 100% pure valley and spin currents respectively. Employing a Wannier tight-binding method we then explore the creation of coherent photocurrents in the 2d magnets CrI_3 and FGT, generalizing the role of the THz and optical pulses to "momentum guiding" and "excitation" components of hybrid laser pulses.

Funding was provided by the European Union's Horizon 2020 research and innovation programme under the European Research Council (ERC) grant agreement No 946629.

O 75.8 Thu 12:45 GER 38

Mode-selective ballistic pathway to a metastable electronic phase — •Hannes Böckmann[1,2], Jan Gerrit Horstmann[3], Felix Kurtz[1,2], Abdus Samad Razzaq[4], Stefan Wippermann[4], and Claus Ropers[1,2] — [1]Max Planck Institute for Multidisciplinary Sciences, Göttingen 37077, Germany — [2]4th Physical Institute, Solids and Nanostructures, University of Göttingen — [3]Department of Materials, ETH Zurich, Zurich — [4]Max-Planck-Institut für Eisenforschung GmbH, Düsseldorf

Exploiting vibrational excitation for the dynamic control of material properties is an attractive goal with wide-ranging technological potential. Most metal-to-insulator transitions are mediated by few structural modes and are, thus, ideal candidates for selective driving toward a desired electronic phase. Such targeted navigation within a generally multi-dimensional potential energy landscape requires microscopic insight into the non-equilibrium pathway. However, the exact role of coherent inertial motion across the transition state has remained elusive. Here, we demonstrate mode-selective control over the metal-to-insulator phase transition of atomic indium wires on the Si(111) surface, monitored by ultrafast low-energy electron diffraction. We use tailored pulse sequences and spectrally selective excitation to individually enhance or suppress key phonon modes and thereby steer the collective atomic motion within the potential energy surface underlying the structural transformation. Our work illustrates that coherent excitation of collective modes via exciton-phonon interactions evades entropic barriers and enables the dynamic control of functionality.

O 75.9 Thu 13:00 GER 38

Nanoscale coherent phonon excitation via plasmon-matter interaction in ultrathin ZnO films — •H. Wiedenhaupt[1], S. Liu[1,2], L. Parra López[1], A. Hammud[1], I. Hamada[3], F. Schulz[1,4], M. Wolf[1], T. Kumagai[1,2], and M. Müller[1] — [1]Fritz Haber Institute, Berlin, Germany — [2]Institute for Molecular Science, Okazaki 444-8585, Japan — [3]Hunan University, Hunan 410082, P. R. China — [4]CIC NanoGUNE, San Sebastian, Spain

In crystalline nanostructures, nanoscale variations of the lattice and/or electronic structure can alter their microscopic response and its interaction with light. Using light-coupled scanning tunneling microscopy (STM) with plasmonic junctions, we demonstrated recently that the local electronic structure of ultrathin ZnO on Ag(111) can strongly affect light-matter interaction on the nanoscale. Specifically, resonant laser excitation of these films seems crucial for tip-enhanced Raman spectroscopy (TERS) [1] and for the nanoscale excitation and observation of coherent phonon dynamics [2]. CPs are excited by femtosecond excitation of localized surface plasmons (LSP), and modulate the femtosecond photocurrent in the STM on ultrafast time scales. To better understand the optical excitation mechanism in these films, we measure plasmon-enhanced STM-induced luminescence, which allows us to correlate the local electronic structure of the films with their light coupling efficiency on the nanoscale. [1] S. Liu, Nano Lett. 19, 5725 (2019), [2] S. Liu et al., Sci. Adv. 8, eabq5682 (2022)