O 98: Ultrafast electron dynamics at surface and interfaces IV

Time: Friday 9:30–12:15 Location: TRE/MATH

Invited Talk O 98.1 Fri 9:30 TRE/MATH

Electron-phonon interactions in the time domain: from non-equilibrium phonon dynamics to theoretical spectroscopy — •Fabio Caruso — University of Kiel, Germany

First-principles simulations of electron-phonon interactions (EPI) are key to clarifying the microscopic origin of the ultrafast electron and phonon dynamics revealed by pump-probe experiments in crystalline solids [1]. In this talk, I will discuss recent advances in ab-initio simulations of the EPI and their application to light-driven electron and lattice dynamics. Building on recent progress in many-body theory and software, predictive atomistic simulations of complex non-equilibrium phenomena have now become feasible. I will highlight, in particular, non-thermal phonon populations [2], coherent phonons [3], and ultrafast polaron formation as representative examples. This progress forms the basis for a strong synergy between ab-initio theory and experimental ultrafast science.

[1] F. Caruso et al., J. Phys. Mater. **9**, 012501 (2025).
[2] Y. Pan et al., ACS Nano **19**, 11381 (2025).
[3] C. Emeis et al., Phys. Rev. X **15**, 021039 (2025).

O 98.2 Fri 10:00 TRE/MATH

Coherent phonon control beyond amplitude saturation in a sliding ferroelectric — •Jan Gerrit Horstmann[1,2], Christoph Emeis[3], Andrin Caviezel[1], Quintin Meier[4], Nicolas Wyler[1], Thomas Lottermoser[1], Fabio Caruso[3], and Manfred Fiebig[1] — [1]Department of Materials, ETH Zurich, Vladimir-Prelog-Weg 4, Zurich, 8093, Switzerland — [2]University of Würzburg, Institute of Physical and Theoretical Chemistry, Am Hubland, 97074 Würzburg, Germany — [3]Institute of Theoretical Physics and Astrophysics, Kiel University, Leibnizstraße 15, Kiel, 24118, Germany — [4]Institut Néel, C.N.R.S-Université Grenoble Alpes, BP 166, Grenoble, 38042, France

Intense optical excitation can drive quantum materials into regimes where the usual linear relation between photo-excited carriers and lattice-driving forces no longer applies. Using time-resolved second-harmonic generation, we observe that the interlayer sliding phonon governing sliding ferroelectricity in WTe_2 saturates and even decreases at high single-pulse fluence. Density functional theory traces this nonlinearity to band-specific electron-phonon coupling that produces competing lattice forces. By dividing the same total fluence into two well-timed pulses, we avoid populating counteracting electronic states and achieve significantly larger coherent phonon amplitudes. The resulting enhanced sliding motion reveals anharmonic phonon coupling far from equilibrium and shows that nonlinear limits in driven solids can be overcome through temporal shaping of the optical excitation.

O 98.3 Fri 10:15 TRE/MATH

Ultrafast x-ray sonography: A novel technique to spatially resolve phase heterogeneity on picosecond timescales — •Maximilian Mattern[1], Angel Rodriguez-Fernandez[2], Roman Shayduk[2], Wonhyuk Jo[2], Vojtech Uhlir[3], Jon Ander Arregi[3], Anders Madsen[2], Daniel Schick[1], and Jan-Etienne Pudell[2] — [1]Max-Born-Institut, Germany — [2]European XFEL, Germany — [3]CEITEC BUT, Czech Republic

Laser-induced heterogeneities play an important role for ultrafast dynamics especially of first-order phase transitions due to the phase coexistence during nucleation. However, their probing on picosecond time and nanometer length scales is challenging.

To address this challenge, we introduce ultrafast X-ray sonography. It combines well-established ultrafast hard-X-ray diffraction with a propagating strain pulse as a universal and non-invasive structural marker. The signatures of the strain pulse in the phase-specific lattice dynamics localizes the different phases in both space and time. This non-invasive pump-probe technique can follow the coherent growth and stochastic nucleation of structural domains on picosecond timescales with almost no requirements for sample preparation and is broadly applicable to a broad class of materials, provided that the coexisting phases exhibit distinguishable diffraction signatures. As a benchmark, we apply this approach to the antiferromagnetic-to-ferromagnetic magneto-structural phase transition in FeRh and identify the ferromagnetic phase to nucleate at the surface as columnar domains of approximately 30nm diameter.

O 98.4 Fri 10:30 TRE/MATH

Ultrafast energy flow among electrons and phonons in a Pb/Si nanoscale heterosystem — •Christian Brand[1], Mohammad Tajik[1], Tobias Witte[1], Laurenz Rettig[2], Birk Finke[1], Björn Sothmann[1], Uwe Bovensiepen[1], and Michael Horn-von Hoegen[1] — [1]Department of Physics, University of Duisburg-Essen, Germany — [2]Department of Physical Chemistry, Fritz Haber Institute, Germany

Microscopic excitations in condensed matter interact on ultrafast time scales. In this study, we employed time-resolved photoelectron spectroscopy and ultrafast electron diffraction to probe the spatially confined dynamics of electrons and phonons in ultrathin epitaxial Pb films on Si(111) subsequent to fs-laser excitation. Spatial confinement is crucial here, as without it, the energy flow into the unexcited bulk would obscure the transient dynamics and prevent targeted investigation. Electrons dissipate their excess energy within 400 fs, while lattice vibrations gradually build up over 3-8 ps. During this time gap, energy is transiently stored in high-frequency phonon modes, as captured by a three-temperature model. The transient electron temperature provides direct experimental access to the occupation of these phonons after equilibration at $\approx$ 3 ps. Subsequent excitation of low-frequency phonons and full lattice thermalization at $\approx$ 20 ps are governed by anharmonic phonon-phonon coupling within the Pb film.

O 98.5 Fri 10:45 TRE/MATH

Ultrafast manipulation of topological transport properties in Td-MoTe2 via coherent phonon excitation — •Huimin Wang[1], Matthew Day[2,3], Juan Aranzadi[1], James McIver[2,3], and Michael Sentef[1] — [1]Institute for Theoretical Physics and Bremen Center for Computational Materials Science, University of Bremen, Bremen, Germany — [2]Department of Physics, Columbia University, New York, NY, USA. — [3]Max Planck Institute for the Structure and Dynamics of Matter, CFEL, Hamburg, Germany

Ultrafast control of topological properties in quantum materials has emerged as a promising route toward dissipative-free and fast optoelectronic functionalities. In type-II Weyl semimetals such as Td-MoTe*, selective photoexcitation of coherent phonons offers an efficient non-thermal pathway to drive topological phase transitions by modifying lattice symmetry. Yet the microscopic origin of the laser-induced interlayer shear mode*central to this process*remains unclear, with both electron*phonon coupling (EPC) and strong phonon anharmonicity expected to contribute. Here, using real-time TDDFT combined with non-adiabatic molecular dynamics, we map out the excitation pathways of this shear mode under varying photon energies, polarizations, and field strengths. We reveal cooperative EPC and phonon*phonon mechanisms that enable control of the phonon phase and amplitude. The resulting lattice dynamics induce shifts of Weyl node positions and associated nonlinear photocurrents, providing insight into observed ultrafast Hall responses.[1]Nature 565, 61*66 (2019). [2]Nat. Commun., vol. 12, p. 1885, 2021.[3]Phys. Rev. X, vol. 9, no. 2, p. 021036, 2019.

O 98.6 Fri 11:00 TRE/MATH

Competition of electron-electron and electron-phonon scattering in thermalization of laser-excited electrons — •Christopher Seibel, Tobias Held, Markus Uehlein, Sebastian T. Weber, and Baerbel Rethfeld — Department of Physics and Research Center OPTIMAS, RPTU University Kaiserslautern-Landau, 67663 Kaiserslautern, Germany

When an ultrashort laser pulse irradiates a metal, electrons are excited to a state of strong nonequilibrium. According to the common picture, they thermalize subsequently towards a hot Fermi distribution on a femtosecond timescale and transfer energy to the still cold lattice on a picosecond timescale. Often these two processes are considered to be independet and driven by different interactions. Electron-electron scattering is considered responsible for the thermalization, whereas electron-phonon scattering is considered responsible for the energy transfer to the lattice. However, there are indications that these two processes influence each other and can act on similar timescales.

We use a kinetic description based on full Boltzmann collision integrals to trace the dynamics of the electron distribution during the thermalization after ultrashort laser excitation. We show that electron-electron and electron-phonon scattering have different effects on the

distribution, which both lead to thermalization. Comparing various excitation fluences, we find that the thermalization times of the two processes exhibit an opposite fluence dependence and can become comparable for weak excitations.

O 98.7 Fri 11:15 TRE/MATH

Interfacial charge dynamics in hybrid plasmonic-excitonic nanostructures — •Tommaso Pincelli[1], Alessandro De Vita[1], Gian Marco Pierantozzi[2], Manuel Izquierdo[3], Pavel Trofimov[4], Trideep Kadwe[4], Hélène Seiler[4], Niclas S. Müller[4], Caterina Cocchi[5], Laurenz Rettig[6], Martin Wolf[6], Ralph Ernstorfer[1], and Giancarlo Panaccione[2] — [1]Technische Univ. Berlin — [2]IOM - CNR — [3]EuXFEL — [4]Freie Univ. Berlin — [5]Fredrich Schiller Univ. Jena — [6]Fritz Haber Inst. - MPG

Hybrid interfaces that merge plasmonic surfaces with 2D semiconductors are pivotal in energy materials research, allowing to tailor local fields and functionalize charge-transfer excitations. In this work, we exfoliate few-layer WSe_2 on a plasmonic lattice of nanostructured Au in UHV. Angle-resolved reflectivity reveals a rich photonic band structure matching finite-element calculations. Using time and angle resolved photoemission spectroscopy (trARPES), we observe an ultrafast quench of excitonic signatures and hints of rapid interfacial charge transfer. Angle-integrated trPES at higher photon energies allows access to the buried interface by tracking shallow core levels. Notably, the plasmonic lattice induces a pronounced slowing of Au 4f peak shifting dynamics compared with unpatterned Au, suggesting a bottleneck in the interfacial charge transfer. These results highlight the potential of hybrid interfaces as flexible platforms for engineering optoelectronic responses at ultrafast timescales. We acknowledge the Energy Materials community proposal collaboration, the teams of SXP@EuXFEL and HEXTOF@FLASH.

O 98.8 Fri 11:30 TRE/MATH

Non-equilibrium electron dynamics in antenna-reactor plasmonic photocatalysts — •Henry T. Snowden[1] and Reinhard J. Maurer[1,2] — [1]University of Warwick, UK — [2]University of Vienna, AT

Antenna-reactor nanoparticles are a promising form of plasmonic photocatalyst, combining strong light absorption and high catalytic activity. A key materials design question is the correct nanoparticle geometry with proposals ranging from core-shell structures to nanoalloys. Optimal photocatalysts combine strong light absorption, effective carrier transport, and long carrier lifetime. Here, we study the non-equilibrium electron dynamics after pulsed laser excitation using the Athermal Electron Model (AthEM), which explicitly considers thermal and non-thermal electrons in contact with a phonon bath, and a laser source term. Electron-electron scattering effects are considered at the level of the relaxation time approximation. We investigate how athermal electron-hole pair distributions evolve in different classes of promising materials according to the interplay of electron-electron and electron-phonon scattering. Based on our findings, we propose optimal materials compositions and photocatalyst geometries.

O 98.9 Fri 11:45 TRE/MATH

Influence of sulphur vacancies on ultrafast charge separation in WS2-graphene heterostructures — •Johannes Gradl[1], Niklas Hofmann[1], Leonard Weigl[1], Camilla Coletti[2], Raül Perea-Causín[3], Ermin Malic[4], and Isabella Gierz[1] — [1]University of Regensburg, Regensburg, Germany — [2]Istituto Italiano di Tecnologia, Pisa, Italy — [3]AlbaNova University Center, Stockholm, Sweden — [4]University of Marburg, Marburg, Germany

Ultrafast charge separation following photoexcitation, commonly observed in van der Waals heterostructures, is a key process for future optoelectronic devices that rely on the conversion of light into electricity. While electron and hole transfer rates are largely determined by band alignment and interlayer hybridization, recent experiments have revealed that defects critically affect the lifetime of the charge-separated state [1][2]. A microscopic understanding of defect-assisted charge transfer, however, remains elusive. We disentangle the role of sulphur vacancies on ultrafast charge separation and recombination in WS2-graphene heterostructures by (i) systematically increasing the defect density and (ii) switching the defect-assisted charge transfer channel on and off. Using time-resolved ARPES, we probe the resulting charge-transfer dynamics on ultrafast time scales directly in the band structure.

[1] Phys. Rev. Lett. 127, 276401, (2021)
[2] Sci. Adv. 7, eabd9061, (2021)

O 98.10 Fri 12:00 TRE/MATH

Formation and thermalization of non-equilibrium excitonic occupations — •Paul Werner[1], Wiebke Bennecke[1], Jan Philipp Bange[1], Giuseppe Meneghini[2], David Schmitt[1], Marco Merboldt[1], Anna Seiler[1], AbdulAziz AlMutairi[3], G. S. Matthijs Jansen[1], Junde Liu[1], Daniel Steil[1], Stephan Hofmann[3], R. Thomas Weitz[1], Ermin Malic[2], Stefan Mathias[1], and Marcel Reutzel[3] — [1]I. Physikalisches Institut, Georg-August Universität Göttingen, Germany — [2]Fachbereich Physik, Philipps-Universität Marburg, Germany — [3]Department of Engineering, University of Cambridge, United Kingdom

Upon optical excitation, bright excitons can scatter into lower energy, potentially dark, exciton states. In these formation and relaxation processes, non-equillibrium exciton populations play a major role, but remain largely elusive because they are inaccessible by most spectroscopic methods. In this work we provide direct access to the formation and thermalization of non-equilibrium exciton states in 2H-homobilayer MoS_2. Using time-resolved momentum microscopy and microscopic many-particle calculations, we reveal the fingerprint of non-equillibrium exciton distributions and quantify their formation and relaxation timescale [1].

[1] Werner *et al.*, arXiv:2505.06074 (2025).