HL 11: Ultra-fast Phenomena I

Time: Monday 16:00–18:45 Location: H14

HL 11.1 Mon 16:00 H14

Femtosecond Photocurrents in 2D materials — •Björn Sinz[1,2], Johannes Schmuck[1,2], Johannes Gröbmeyer[1,2], Nina Pettinger[1,2], Sergey Zherebtsov[1,2], and Alexander Holleitner[1,2] — [1]Walter Schottky Institute and Physics Department, TU Munich, Munich, Germany — [2]Munich Center of Quantum Science and Technology (MCQST), Munich, Germany

Light-field-driven currents have already been investigated in graphene by ultrashort laser pulses. In the strong-field regime, the electric field directly generates such photocurrents on a time scale of femtoseconds, whereas in the weak-field regime other photogeneration processes like the photo-thermoelectric effect dominate [1]. The different photocurrents are not limited to graphene but are also predicted for transition metal dichalcogenide (TMDC) monolayers. We report on femtosecond-pulse driven photocurrents in monolayer TMDC samples.

[1]: J. Gröbmeyer, P. Zimmermann, B. Huet, J. A. Robinson, A. W. Holleitner; Space-charge limited and ultrafast dynamics in graphene-based nano-gaps. Appl. Phys. Lett. 3 July 2023; 123 (1): 013504. https://doi.org/10.1063/5.0154152

HL 11.2 Mon 16:15 H14

Hot-Electron-Induced Substrate Response in Transient Absorption Spectroscopy of Tantalum — •Erik Willem de Vos[1,2], Sergej Neb[1], Marko Hollm[1], Florence Burri[1], Lukas Gallmann[1], and Ursula Keller[1] — [1]Department of Physics, Institute for Quantum Electronics, ETH Zurich, Switzerland — [2]Department of Materials, ETH Zurich, Zurich, Switzerland

We show that for extreme ultraviolet transient absorption spectroscopy measurements on thin-film metals, the substrate can significantly contribute to the observed change in absorption even if the substrate is transparent to the excitation wavelengths of the pump pulse and does by itself not produce a transient signal. Irradiation of a thin-film tantalum layer deposited on a silicon nitride substrate by a near-infrared femtosecond pulse is found to excite a coherent acoustic phonon in both the tantalum as well as the substrate. The response in the substrate rises on sub-picosecond timescales and is the result of direct excitation by the hot-electron distribution in the metal layer.

HL 11.3 Mon 16:30 H14

Nonequilibrium electron-phonon dynamics: dynamical control of quantum matter — •Yaxian Wang — Institute of Physics, Chinese Academy of Sciences

Electron-phonon interaction is an old yet evergreen problem in condensed matter physics. It is closely related to many quantum states we are concerned with, such as superconductivity, charge density waves, and polarons. It also profoundly affects, or often accompanies, critical phenomena such as the formation of excitonic insulators and metal-insulator transitions. When the system is driven out of equilibrium, for example pumped by an ultrafast laser pulse, the potential energy surface and thus the coupling in the excited states can be greatly reshaped, and this may open up a new avenue of ultrafast coherent control of quantum phases and topological orders. However, theoretical approaches often fail to capture the coupled dynamics of the non-thermal excited carriers and the nonequilibrium lattice order. In this talk, I will introduce how light-induced coherent phonons can cause a quasi-static lattice distortion and result in a Lifshitz transition in a nodal-line semimetal. We also demonstrate how the laser energy can shift the quasi-equilibrium lattice structure towards opposite directions, thus engineering the electronic structure via different regimes. Moreover, I will discuss our recent discovery on how nonequilibrium electron-phonon interaction can interact with the spin degree of freedom, causing ultrafast demagnetization and excitation of chiral phonons in a monolayer ferromagnet.

HL 11.4 Mon 16:45 H14

Nonequilibrium control of the ultrafast electron dynamics in semiconductors via light-driven coherent phonons — •Chenyu Wang[1,2], Yaxian Wang[1], and Sheng Meng[1,2,3] — [1]Institute of physics, Chinese Academy of Sciences — [2]School of physical sciences, University of Chinese Academy of Sciences — [3]Songshan Lake Materials Laboratory

Driving lattice vibration with a high degree of spatial and temporal coherence via strong light-matter interactions has emerged as a unique knob to control the out-of-equilibrium quantum states and the exotic ultrafast phenomena.

In this talk, I will present our recent theoretical works on the exploration and understanding of how the coherent phonon excitation can be utilized to control the electronic behaviors on ultrafast timescales, for example to engineer the carrier transport in monolayer WSe2 and to manipulate the nonrelativistic spin splitting in the prototypical altermagnetic semiconductor MnTe.

In photoexcited monolayer WSe2, we observe an unconventional 'step-like' electron intervalley scattering dynamics and meanwhile a Rabi oscillation driven by the coherent phonons, as a direct manifestation of the nonadiabatic electron-phonon coupling beyond equilibrium; On the other side, we demonstrate the light-driven coherent phonon can also be utilized to break the crystal symmetry in the altermagnet MnTe. Such symmetry breaking phase holds an extra spin-splitting particularly in the zone center, and thus a strongly enhanced spontaneous anomalous Hall effect.

HL 11.5 Mon 17:00 H14

Semiconductor Bloch Equations and Ehrenfest Dynamics in a Wannier Function Framework: An Integrated Approach to Ultrafast Electron and Ion Dynamics — •Stefano Mocatti, Giovanni Marini, Giulio Volpato, Pierluigi Cudazzo, and Matteo Calandra — Department of Physics, University of Trento, Via Sommarive 14, 38123 Povo, Italy

Real-time simulations of photoexcited semiconductors offer valuable insights but are challenged by the complex interplay of interactions and high computational cost. Here, we present an efficient ab initio scheme within the EPIq code, combining semiconductor Bloch equations with Ehrenfest dynamics in the Wannier representation. Electron-phonon and electron-electron interactions follow a Fan-Migdal+GW approximation, while phonon-phonon interactions include non-perturbative quantum anharmonic effects. Through the real-time tracking of the dynamics, we find that electron-electron interaction dominates photocarrier thermalization, yielding a double chemical potential electronic Fermi distribution. Furthermore, atomic forces converge to those calculated within constrained density functional perturbation theory. This work connects out-of-equilibrium and quasi-equilibrium states, advancing the understanding of ultrafast light-driven phenomena.

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15 min. break

HL 11.6 Mon 17:30 H14

Ultrafast Dynamic Coulomb Screening of X-ray Core Excitons in Photoexcited Semiconductors — •Thomas C. Rossi[1], Lu Qiao[2], Conner P. Dykstra[3], Ronaldo Rodrigues Pela[4], Richard Gnewkow[1,5], Rachel Wallick[3], John H. Burke[3], Erin Nicholas[3], Anne-Marie March[6], Gilles Doumy[6], D. Bruce Buchholz[7], Christiane Deparis[8], Jesus Zuñiga-Pérez[8,9], Michael Weise[10], Klaus Ellmer[10], Mattis Fondell[1], Claudia Draxl[2], and Renske M. van der Veen[1,5,11] — [1]Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Berlin, Germany — [2]Department of Physics and CSMB, Humboldt Universität zu Berlin, Berlin, Germany — [3]Department of Chemistry, University of Illinois at Urbana-Champaign, Urbana, Illinois, USA — [4]Supercomputing Department, Zuse Institute Berlin (ZIB), Berlin, Germany — [5]Institute of Optics and Atomic Physics, Technische Universität Berlin, Berlin, Germany — [6]Chemical Sciences and Engineering Division, Argonne National Laboratories, Lemont, Illinois, USA — [7]Department of Materials Science and Engineering, Northwestern University, Evanston, Illinois, United States — [8]Université Côte d'Azur, CNRS, CRHEA, rue Bernard Gregory, Sophia Antipolis, Valbonne, France — [9]Majulab, International Research Centre Laboratory IRL 3654, Singapore — [10]Optotransmitter-Umweltschutz-Technologie (OUT) e.V., Berlin, Germany — [11]Department of Chemistry, University of Illinois at Urbana-Champaign, Urbana, Illinois, USA

The screening of core excitons is an inherent many-body process that can reveal insight into charge-transfer excitations and electronic correlations. Here we demonstrate the dynamic Coulomb screening induced by photoexcited carriers on core excitons employing X-ray transient absorption (XTA) spectroscopy with picosecond time resolution. Our interpretation is supported by state-of-the-art *ab initio* calculations including many-body perturbation theory. Using ZnO as an archetypal wide band-gap semiconductor, we show that the Coulomb screening modification at the Zn L_3- and K-edge leads to a decrease in the core-exciton binding energy. We also theoretically predict the effect of core-exciton screening on the femtosecond time scale for the case of ZnO, a major step towards hard X-ray excitonics. The results have implications for the interpretation of ultrafast X-ray spectra in general and their use in tracking charge carrier dynamics in complex materials on atomic length scales.

HL 11.7 Mon 17:45 H14

Ultrafast Coherent Dynamics of a Hybrid WS2/Plasmon Structure Probed by Two-Dimensional Electronic Spectroscopy — •Daniel Timmer[1], Moritz Gittinger[1], Daniel C. Lünemann[1], Thomas Quenzel[1], Sven Stephan[1,2], Martin Silies[1,2], Antonietta De Sio[1], and Christoph Lienau[1] — [1]IfP, Carl von Ossietzky Universität Oldenburg, Oldenburg, Germany — [2]ILO, Hochschule Emden/Leer, Emden, Germany

Transition metal dichalcogenide (TMD) monolayers (1L) have been established as important building blocks for quantum materials. Hybridization between light and matter states, in particular in plasmonic nanostructures, offers great opportunities to tailor their optical and electronic properties [1]. Here, we investigate such a hybrid plasmonic structure in the intermediate coupling regime comprised of 1L-WS2 [2] placed on a periodic silver nano-slit array using ultrafast two-dimensional electronic spectroscopy (2DES). We observe a 20-fold increase of the optical nonlinearity and ultrafast coherent plexciton dynamics during the dephasing time (~50 fs). We rationalize our observations via a Tavis-Cummings model that gives rise to collective dark states and a Rabi contraction of the 2-quantum states. Using ultrafast 2DES, we obtain access to probe and disentangle the coherent and incoherent dynamics in TMD-based plasmonic systems. [1]: Timmer et al. "Plasmon mediated coherent population oscillations in molecular aggregates." Nat. Commun. 14.1 (2023): 8035. [2]: Timmer et al. "Ultrafast Coherent Exciton Couplings and Many-Body Interactions in Monolayer WS2" Nano Lett. 24.26 (2024): 8117-8125.

HL 11.8 Mon 18:00 H14

Excited-state symmetry breaking and antisymmetric mode brightening in quadrupolar dye — •Somayeh Souri[1], Katrin WInte[1], Daniel Lünemann[1], Fulu Zheng[2], Mohamed Madjet[2], Teresa Kraus[3], Elena Mena-Osteritz[3], Peter Bäuerle[3], Sergei Tretiak[4], Antonietta De Sio[1], and Christoph Lienau[1] — [1]Oldenburg University, Germany — [2]Bremen University, Germany — [3]Ulm University, Germany — [4]Los Alamos National Laboratory, USA

Quadrupolar acceptor-donor-acceptor (A-D-A) dyes are chemically tunable materials displaying a rapid photo-induced charge transfer of interest for applications in solution-processed photovoltaics. The origin of the ultrafast charge transfer is unknown. Using sub-10-fs ultrafast spectroscopy, we investigate the excited-state dynamics of a prototypical A-D-A with comparable electronic and vibronic coupling strengths. Our results reveal that vibronic couplings to high-frequency C-C-stretching vibrations on each arm of the quadrupolar dye induce a double-minimum potential energy surface (PES) in the excited-state S1 driving symmetry breaking along the antisymmetric vibrational coordinate (Q-). Upon excitation, this induces periodic splitting of the optically launched coherent wavepacket along Q- with 20-fs period, rapidly relaxing into local minima of the symmetry-broken PES within less than 100 fs. Our results demonstrate highly nonadiabatic vibronic quantum dynamics, theoretically predicted for this class of dyes [1], and unravel their role for the ultrafast charge transfer in this class of molecules. [1] J. Chem. Phys. 141, 164317 (2014).

HL 11.9 Mon 18:15 H14

Phonon-driven exciton population oscillations in Methylammonium Lead Bromide Perovskites. — •Mohsin Sayar[1], Katrin Winte[1], Daniel Timmer[1], Somayeh Souri[1], Davide Ceratti[2], David Cahen[2], Christoph Lienau[1], and Antonietta De Sio[1] — [1]Carl von Ossietzky Universität, Oldenburg, Germany. — [2]Weizmann Institute of Science, Rehovot, Israel

Halide perovskites exhibit unique optoelectronic properties significantly influenced by electron-phonon interactions. Recent work shows that the internal fields induced by coherent lattice motions can transiently control ultrafast excitonic optical response in bulk $CsPbBr_3$[1]. Here, we demonstrate this behaviour also in $CH_3NH_3PbBr_3$ across different crystal phases, using temperature-dependent ultrafast transient reflectivity with 10 fs time resolution. Following resonant exciton excitation reveals coherent low-frequency phonon oscillations at 40cm^{-1} and 67cm^{-1}, corresponding to Pb-Br-Pb bending and stretching modes[2] that most strongly couple to the exciton. Additionally, we observe faster oscillations with dominant period of 105 fs in all crystal phases, arising from coherent exciton population transfer between 1s and 2p excitonic states, off-resonantly driven by the low-frequency phonon fields. We rationalize these results by a phenomenological model accounting for the coupling of excitons to the low frequency phonon modes and coupling of 1s-2p vibronic manifolds via the phonon fields[1]. These results may have important implications for transiently modifying the optoelectronic properties of perovskites.

HL 11.10 Mon 18:30 H14

Coherent suppression of high-harmonic generation in Dirac materials — •Wolfgang Hogger[1], Alexander Riedel[1], Debadrito Roz[2], Angelika Knothe[1], Cosimo Gorini[3], Juan-Diego Urbina[1], and Klaus Richter[1] — [1]Institute for theoretical physics, University of Regensburg, Germany — [2]Indian Institute of Science, Bengaluru 560012, India — [3]Université Paris-Saclay, CEA, CNRS, SPEC, 91191, Gif-sur-Yvette, France

The study of high-harmonic generation in solids by intense laser pulses provides a fascinating platform for studying ultra-fast electron dynamics and material properties, where the coherent character of the electron dynamics is a central aspect. Starting with the semiconductor Bloch equations, we show the ubiquitous presence of a mechanism suppressing the high harmonic spectrum arising from the coherent superposition of intra- vs inter-band contributions to the total signal [1]. We provide evidence for the generality of this phenomenon by extensive numerical simulations exploring the parameter space of this coherent suppression of high harmonic generation in systems of massive Dirac Fermions (as a prototypical model for topologically non-trivial matter [2]), systems with a pseudo- relativistic dispersion. We supplement our numerical observations with analytical results for a simplified single-mode analysis.

[1] Y. Murakami and M. Sch*uler, Phys. Rev. B 106, 35204 (2022)
[2] C.-X. Liu, X.-L. Qi, H. Zhang, X. Dai, Z. Fang, and S.-C. Zhang, Physical Review B 82, (2010)