HL 30: Ultrafast Phenomena II

Time: Wednesday 10:00–12:15 Location: EW 015

HL 30.1 Wed 10:00 EW 015

Ultrafast coherent exciton dynamics and manybody interactions in a WS2 monolayer — Daniel Timmer[1], •Moritz Gittinger[1], Daniel Lünemann[1], Sven Stephan[1,2], Martin Silies[1,2], Antonietta De Sio[1], and Christoph Lienau[1] — [1]University of Oldenburg, Germany — [2]University of Applied Sciences Emden, Germany

Monolayers of transition metal dichalcogenides (TMDCs) are quantum materials with fascinating optoelectronic properties. In particular, valleytronic applications are exploiting the spin-selectivity that is provided by the valley pseudospin and large energy splitting between A and B excitons due to strong spin-orbit coupling. However, manybody interactions lead to rapid valley depolarization. In particular, exchange interactions lead to a mixing of A and B excitons. We use ultrafast pump-probe and two-dimensional electronic spectroscopy (2DES), the ideal tool to investigate coherent couplings and manybody interactions in semiconductors, to directly probe these A-B exciton interactions in a monolayer of WS2. We observe the signatures of coherent couplings between the A and B excitons, cross-peaks in the 2DES maps. The associated coherent Rabi oscillations with ~11.5-fs period are also resolved. In addition, the 2DES data gives detailed insight into the formation of manybody interactions on a tens of fs timescale. The observed coherent couplings are likely reflecting a Dexter-type intervalley exchange interaction. Broadband ultrafast spectroscopy with sufficiently high time-resolution proofs therefore essential in directly probing the coherent quantum dynamics in the time domain.

HL 30.2 Wed 10:15 EW 015

Ultrafast phonon-driven exciton Rabi oscillations in halide perovskites — X.Trung Nguyen[1], Katrin Winte[1], Daniel Timmer[1], Yevgeny Rakita[2], Sufyan Ramzam[1], Caterina Cocchi[1], Michael Lorke[3], Frank Jahnke[3], David Cahen[2], Christoph Lienau[1], and •Antonietta De Sio[1] — [1]Universität Oldenburg — [2]Weizmann Institute of Science — [3]Universität Bremen

Coupling electromagnetic radiation with matter is highly promising for tailoring optoelectronic properties and photoinduced dynamics of functional materials. Here we show that even the internal fields induced by coherent lattice motions can transiently control the ultrafast excitonic response in halide perovskites(HaPs). Temperature-dependent ultrafast two-dimensional electronic spectroscopy reveals that the characteristic low-frequency phonons of the soft lead-halide lattice induce strong mixing of 1s and 2p exciton manifolds in $CsPbBr_3$ and $MAPbBr_3$. This results in side-peaks in the spectra and 100-fs-Rabi oscillations in the dynamics. The temperature-dependence of the Rabi oscillations provides insight into dephasing mechanisms within the excited exciton manifold. Our results show that the conventional Fröhlich model is insufficient to describe the behavior of these materials. Instead, they indicate the importance of nonadiabatic couplings. An important implication is that intrinsic optical phonon fields in HaPs may be exploited to transiently modify their optoelectronic properties, and to explore fundamental field-matter interactions toward shaping coherent exciton dynamics by strong and ultrastrong coupling to phonons.

HL 30.3 Wed 10:30 EW 015

Transient core-hole screening investigated by time-resolved X-ray absorption spectroscopy at the Zn K-edge of ZnO — •Thomas C. Rossi[1], Lu Qiao[2], Keith Gilmore[2], Ronaldo Rodrigues Pelà[2], Claudia Draxl[2], and Renske M. van der Veen[1] — [1]Helmholtz Zentrum Berlin für Materialien und Energie GmbH, 14109 Berlin, Germany — [2]Department Physics and IRIS Adlershof, Humboldt-Universität zu Berlin, D-12489 Berlin, Germany

Understanding the ultrafast electronic and lattice response of photoexcited semiconductor materials at the atomic level is crucial for the realization and optimization of devices. Here, we report on the picosecond dynamics of atoms and photoexcited charge carriers above the optical band gap of ZnO in oriented nanorods and thin films by time-resolved X-ray absorption spectroscopy (TRXAS) at the Zn K-edge. The transient signal reveals the non-local screening of the core-hole potential by photogenerated electron-hole pairs, often overlooked in previous studies on photoexcited semiconductors. State-of-the-art calculations with the Bethe Salpeter equation are able to reproduce the spectral features and the non-linear effect of the core-hole screening on the transient with the excitation density. Theoretical predictions are made on the effect of the pump/probe polarization and efficiency of the non-local screening on the femtosecond timescale. This work highlights the simultaneous sensitivity of TRXAS to incoherent atomic motions and delocalized photoexcited carriers with chemical element sensitivity, which opens new perspectives for the study of photoexcited semiconductors in complex materials or in optoelectronic devices.

HL 30.4 Wed 10:45 EW 015

Lightwave-control of on-chip femtosecond electronics — •Johannes Schmuck[1,2], Johannes Gröbmeyer[1,2], Maximilian Auers[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 Technolgy (MCQST), Munich, Germany

Ultrashort laser pulses allow the generation of light-driven electrical currents [1]. By different photoemission processes the generation of THz electric pulses on on-chip circuits is possible. The corresponding pulses with increased bandwidths up to 10 THz can propagate along macroscopic striplines on a millimeter scale [2]. Rather than using femtosecond photoswitches based on photoconductivity or biased tunneling barriers in nanoscale metal junctions to drive the pulses [3], we investigate direct control of the electronic pulses by single-cycle laser pulses.

[1] M. Ludwig et al. Sub-femtosecond electron transport in a nanoscale gap. Nat. Phys. 16, 341-345 (2020). [2] C. Karnetzky et al. Towards femtosecond on-chip electronics based on plasmonic hot electron nano-emitters. Nat Commun 9, 2471 (2018). [3] J. Gröbmeyer et al. Space-charge limited and ultrafast dynamics in graphene based nano-gaps. Appl. Phys. Lett. 123, 013504 (2023).

15 min. break

HL 30.5 Wed 11:15 EW 015

Full calculation of the observable for HHG in solids — •Francisco Navarrete and Dieter Bauer — Institute of Physics, University of Rostock, 18051 Rostock, Germany

When an intense infrared laser interacts with a target, it can produce radiation at frequencies that are multiples of the central frequency of the driving source. This is known as high-harmonic generation (HHG), which was first investigated for atomic and molecular targets [1] and later for condensed matter [2]. The growing attention to explain the impact of the quantum nature of the driving pulse [3], along with the recognized discrepancies between the spectra in HHG in solids obtained from numerical calculations and experiments [4], has encouraged the pursuit of new methods to compute the spectra [5]. In this study, we demonstrate the distinctions in the harmonic spectrum of HHG in solids by comparing the conventional calculation method of the observable [6] with an alternative approach that takes into account typically overlooked terms. Additionally, we examine the underlying physics behind these dissimilarities. [1] M. Lewenstein et al., Phys. Rev. A 49, 2117 (1994) [2] S. Ghimire et al., Nat. Phys. 7, 138 (2011) [3] M. Lewenstein et al., Nat. Phys. 17, 1104 (2021) [4] I. Floss et al., Phys. Rev. A 97, 011401(R) (2018). [5] A. Gorlach et al., Nat Commun 11, 4598 (2020) [6] B. Sundaram and P. W. Milonni, Phys. Rev. A 41, 6571(R) (1990)

HL 30.6 Wed 11:30 EW 015

Modelling dephasing effects in High Harmonic Generation in a real space Tight-Binding-approach — •Martin Thümmler[1], Alexander Croy[1], Ulf Peschel[2], and Stefanie Gräfe[1] — [1]Institute of Physical Chemistry, University of Jena — [2]Institute of Condensed Matter Theory and Optics, University of Jena

High harmonic generation in bulk materials is commonly described by semiconductor Bloch equations employing ultrashort dephasing times (~ 2 fs). Those are chosen to match the clear harmonic peaks observed in experiments, but contradict spectroscopic measurements of narrow linewidth excitonic peaks in semiconductors. Therefore the physical origin of these short dephasing times is currently strongly debated.

In this talk, we focus on real space dephasing models starting from our periodic real space Tight-Binding model. Extending this model, we discuss the effect of different real space dephasing mechanisms on

the calculated high harmonic spectra.

HL 30.7 Wed 11:45 EW 015

Time-resolved on-chip electronics in the THz regime — Johannes Schmuck[1,2], •Maximilian Auers[1,2], Johannes Gröbmeyer[1,2], Nina Pettinger[1,2], Sergey Zherebtsov[1,2], and Alexander Holleitner[1,2] — [1]Walter Schottky Institute, TU Munich — [2]MCQST

Our study explores on-chip circuits driven by ultrashort femtosecond pulses, which allow an increased bandwidth of up to 10 THz [1]. The circuits employ macroscopic striplines spanning on the micrometer scale to facilitate the propagation of corresponding electronic pulses. We present a two-temperature heat model investigating the heat dissipation across the metal electrodes in combination with different substrate materials. Such stripline circuits mark a pivotal advancement toward the integration of femtosecond electronics within on-chip quantum circuits.

[1] C. Karnetzky et al. Towards femtosecond on-chip electronics based on plasmonic hot electron nano-emitters. Nat Commun 9, 2471 (2018).

HL 30.8 Wed 12:00 EW 015

First-Order Rhombohedral to Cubic Phase Transition in Photoexcited GeTe — •Matteo Furci, Giovanni Marini, and Matteo Calandra — Department of Physics, University of Trento, Via Sommarive 14, 38123 Povo, Italy

Photoexcited GeTe undergoes a non-thermal phase transition from a rhombohedral to a rocksalt crystalline phase. The microscopic mechanism and the nature of the transition are unclear. By using constrained density functional perturbation theory and by accounting for quantum anharmonicity within the stochastic self-consistent harmonic approximation, we show that the non-thermal phase transition is strongly first order and does not involve phonon softening, at odd with the thermal one. The transition is driven by the closure of the single particle gap in the photoexcited rhombohedral phase. Finally, our work shows how ultrafast XRD data are consistent with a coexistence of the two phases, as expected in a first order transition. Our results are relevant for the understanding of phase transitions and bonding in phase change materials.