HL 23: Ultra-fast phenomena

Time: Tuesday 9:30–12:30 Location: POT 112

HL 23.1 Tue 9:30 POT 112

Atomic disorder in electroninc materials revealed by decoherence — •Samuel Palato[1], Hélène Seiler[1], Patanjali Kambhampati[2], Parmeet Nijjar[3], and Oleg Prezhdo[3] — [1]Frits-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14169 Berlin — [2]Department of Chemistry, McGill University, 801 rue Sherbrooke O., Montréal, Qc, Canada H3A 0B8 — [3]Department of Chemistry, University of Southern California, Los Angeles, California 90089

Disorder in electronic materials is a challenge for both experimental studies and theoretical description. Coherent dynamics provide a sensitive spectroscopic probe of electronic disorder. We exploit recent advances in multidimensional spectroscopy to study coherent dynamics in the model system of CdSe nanocrystal quantum dot (QD). Coherence mapping in both amplitude and phase reveals the nature of the coherent dynamics as vibrational or electronic. According to the standard model for the electronic structure of semiconductor QDs, decoherence is dominated by inhomogeneity in the sizes of the nanocrystals. Predictons from this model are inconsistent with the observation. Instead, we show decoherence arises naturally when accounting for the individual atoms by performing *ab initio* molecular dynamics of a single QD. Accounting for atomic positions results in a complicated electronic manifold. This atomistic disorder is intrinsic to the QD, and is expected to be a general phenomenon in nanostructures.

HL 23.2 Tue 9:45 POT 112

Ultrafast Laser Excitation of ZnO: A First-Principles Study — •Xiao Chen and Silvana Botti — Institut für Festkörpertheorie und -optik, Friedrich-Schiller-Universität Jena, Max-Wien-Platz 1, 07743 Jena, Germany

Laser device miniaturization is a focused area of nano optics, and ZnO nanowire lasers are attracting large interest in this context. ZnO is a well studied gain medium, and the faceted cylindrical geometry of ZnO nanowires forms a waveguide cavity. Recent experiments [1] focused on the pumping phase, and more specifically the multi-photon absorption and tunneling regimes. In this presentation, we discuss first-principles calculations of electronic excitations in ZnO, driven by strong lasers. These simulations were realized using time-dependent density functional theory, implemented in the open source code Octopus [2].

[1] R. Hollinger et al. Nano Lett. **19**, 6, 3563 (2019).
[2] X. Andrade et al., Phys. Chem. Chem. Phys, **17** 31371 (2015)

HL 23.3 Tue 10:00 POT 112

Optical Vortex Core Switching in Polariton Condensates — •Matthias Pukrop[1], Stefan Schumacher[1,2], and Xuekai Ma[1] — [1]Department of Physics and CeOPP, Paderborn University, Paderborn, Germany — [2]College of Optical Sciences, University of Arizona, Tucson, AZ 85721, USA

Vortices are topological objects carrying quantized orbital angular momentum, also known as topological charge, and have been widely studied in many physical systems. In those with spin degree of freedom the elementary excitations are so called half-vortices (HVs), referring to a vortex state carrying a topological charge in only one circular polarization component. Here we demonstrate the existence of localized half-vortices in spinor polariton condensates, non-resonantly excited by a linearly polarized ring-shaped pump [1,2]. In the core region of the half-vortex the condensate is circularly polarized, while it is linearly polarized elsewhere. With TE-TM splitting, the pseudospin structure of the condensate gives rise to solutions with broken cylindrical symmetry. The attractive cross-interaction between different spin components can be used to realize optical vortex core switching between left- and right-circularly polarized HV states [1]. This switching process reverses the circular polarization in the HV core. It can be easily detected by measuring the polarization resolved intensity in the vortex core region. The same method can also be applied to higher order states, enabling multi-level switching configurations.

[1] M. Pukrop et al., arXiv:1907.10974 (2019).
[2] X. Ma et al., Physical Review Letters 121, 227404 (2018).

HL 23.4 Tue 10:15 POT 112

Comparison of atomic pathways during nonthermal melting in the isostructural elements germanium and silicon — •Tobias Zier and Martin Garcia — Theoretische Physik, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany

Intense femtosecond-laser pulses can induce extreme non-equilibrium conditions in solids. As a consequence the crystalline structure of the solid exhibits nonthermal effects, like, coherent phonons, thermal phonon squeezing, or nonthermal melting. In the latter case the interatomic bonding is broken by the laser pulse, which excites a large amount of electrons from bonding into antibonding states. The ensuing atomic forces, that raise because of the change in the interatomic bonding, accelerate the atoms away from their initial positions. In a recent publication we could resolve the atomic motions and pathways during nonthermal melting in silicon by performing molecular dynamics simulations using our own electronic-temperature density-functional theory code CHIVES.[1] Here, we study the atomic motion during nonthermal melting in the isostructural element germanium, which has almost the same lattice parameter than silicon but less strong interatomic bonding. We will identify the most important melting directions and compare them to the results found in silicon, in order to find similarities and/or differences in the underlying melting mechanism. Our findings will help to find a general theory that describes the atomic pathways during nonthermal melting.

[1] T. Zier, E. S. Zijlstra, M. E. Garcia, Phys. Rev. Lett. **116**, 153901 (2016).

HL 23.5 Tue 10:30 POT 112

Structural Dynamics in Nanostructured Systems Probed by Ultrafast Transmission Electron Microscopy — •Nora Bach[1], Armin Feist[2], Marcel Möller[2], Claus Ropers[2], and Sascha Schäfer[1] — [1]Institute for Physics, University of Oldenburg, Germany — [2]4th Physical Institute - Solids and Nanostructures, University of Göttingen, Germany

A successful approach to investigate ultrafast nanoscale structural dynamics and to disentangle different excitation mechanisms in spatially inhomogeneous systems is based on local diffractive probing with nanofocussed femtosecond electron pulses in an ultrafast transmission electron microscope (UTEM).

Employing the advanced electron pulse properties of the Göttingen UTEM [1,2] in ultrafast convergent electron beam diffraction (U-CBED) mode [3], we study local dynamics in a multi-component model system consisting of a metal-/semiconductor hybrid structure. Ultrashort optical excitation of platinum stripes on a silicon membrane results in the generation of hot electrons and their subsequent coupling with the underlying silicon substrate. Pronounced lattice distortions are quantitatively tracked by U-CBED, and experimental results are compared to finite element simulations in order to provide deeper insights into local couplings and the efficiency of electronic and phononic transport channels across interfaces.

[1] A. Feist, N. Bach, et al., Ultramicroscopy 176, 63 (2017).
[2] N. Bach et al., Structural Dynamics 6, 014301 (2019).
[3] A. Feist et al., Structural Dynamics 5, 014302 (2018).

30 min. break

HL 23.6 Tue 11:15 POT 112

Femtosecond laser-induced electron emission from nanodiamond-coated tungsten needle tips — •Alexander Tafel, Stefan Meier, Jürgen Ristein, and Peter Hommelhoff — Department Physik, Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU), 91058 Erlangen

We present femtosecond electron emission from a tip-shaped metal-semiconductor heterostructure. By coating tungsten needle tips with thin nanodiamond (50-200 nm), we combine the high electron beam quality of tip-shaped emitters with the robustness and the negative electron affinity of diamond.
Due to the high peak intensity of femtosecond laser pulses, electrons can be photoexcited for wavelengths from the infrared (1932 nm) to the ultraviolet (235 nm) because multiphoton excitation becomes efficient over the entire spectral range. Depending on the laser wavelength, we find different dominant emission channels identified by the number of photons needed to emit electrons. Based on the band alignment between tungsten and nanodiamond, the relevant emission channels can be identified as specific transitions in diamond and its graphitic bound-

aries. Emission is stable at all wavelenths and bunch charges tested (up to 400 electrons per pulse). We infer a normalized emittance of less than 0.20 nm rad and a normalized peak brightness higher than $1.2 \cdot 10^{12} \mathrm{Am}^{-2}\mathrm{sr}^{-1}$.

Reference: A. Tafel et al., Phys. Rev. Lett. 123, 146902, **2019**.

HL 23.7 Tue 11:30 POT 112

Femtosecond electron diffuse scattering probes momentum-resolved phonon dynamics in black phosphorus — •Hélène Seiler[1], Daniela Zahn[1], Marios Zacharias[1], Patrick Hildebrandt[1], Thomas Vasileiadis[1], Will Windsor[1], YingPeng Qi[1], Maciej Dendzik[1], Christian Carbogno[1], Claudia Draxl[2], Fabio Caruso[2], and Ralph Ernstorfer[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany. — [2]Department of Physics, Humboldt-Universität zu Berlin, Brook-Taylor-Straße 6, 12489 Berlin, Germany

We employ femtosecond electron diffuse scattering in combination with first principle calculations to reveal the consequences of atomic and electronic structure anisotropy on phonon dynamics in prototypical anisotropic semiconductor black phosphorus. We show the presence of two dominant channels for electron-phonon scattering, namely intravalley scattering within the Gamma valley and intervalley scattering towards the Y valley. Phonons subsequently build up along the Gamma-X path over tens of picoseconds, as a result of phonon-phonon coupling. These measurements provide insights into the origin of anisotropic non-equilibrium properties with unprecedented level of detail. Our work provides direct experimental evidence that a transient nonthermal phonon distribution exists for tens of picoseconds in black phosphorus and highlights the key role of phonon-phonon scattering in phonon thermalization. That thermalization in black phosphorus is limited by phonon-phonon scattering is expected to have consequences on heat and electrical transport properties.

HL 23.8 Tue 11:45 POT 112

Photoelectron spectroscopy combining an ultrashort infrared field with an attosecond pulse pair — •Jan Vogelsang, Sara Mikaelsson, Chen Guo, Cord L. Arnold, Mathieu Gisselbrecht, Anders Mikkelsen, and Anne L'Huillier — Department of Physics, Lund University, 221 00 Lund, Sweden

We perform photoelectron spectroscopy in helium gas and at a zinc oxide (ZnO) surface using two attosecond pulses with a fixed delay and a 7-fs short infrared laser pulse. The experiments are enabled by a 200-kHz repetition rate OPCPA laser system, generating pairs of attosecond pulses in a high pressure gas jet. The measured photoelectron kinetic energy spectra are strongly modulated due to the interference of the electron matter waves emitted by the pair of attosecond pulses, showing the well-known odd-order harmonics. Additionally, the infrared dressing field leads to a periodic modulation of the spectra when we change the temporal delay, very much like in an attosecond streaking experiment, but with the spectral resolution of the harmonic frequency comb.

HL 23.9 Tue 12:00 POT 112

Non-perturbative subcycle nonlinearities of ultrastrong light-matter coupling — •Joshua Mornhinweg[1], Maike Halbhuber[1], Cristiano Ciuti[2], Dominique Bougeard[1], Rupert Huber[1], and Christoph Lange[1] — [1]University of Regensburg, Germany — [2]Université de Paris, France

In the ultrastrong-coupling regime, the rate of energy exchange between the light field of an optical resonator and an electronic excitation - the vacuum Rabi frequency, Ω_R - is comparable to the carrier frequency of light, ω_c, and anti-resonant interaction terms govern the dynamics. In this setting, novel quantum effects including the vacuum Bloch-Siegert shift, modified electronic transport, or light-induced superconductivity have been explored. Yet, little is known about the nonlinearities of this extreme regime of light-matter interaction. Here, we investigate extreme subcycle nonlinearities of Landau cavity polaritons with a coupling strength of $\Omega_\mathrm{R}/\omega_\mathrm{c} = 0.6$. Two-dimensional terahertz (THz) spectroscopy probes the response with amplitude and phase resolution, on a subcycle scale. In a strong-field setting of multiple THz photons per Landau electron, the dynamics include pump-probe as well as four and six-wave mixing processing observed for each of the polaritons. Most importantly, we observe off-diagonal contributions, which our Liouville path analysis links to nonlinear interactions between the individual polariton states and to a collapse of the normal-mode approximation. Our quantitative microscopic theory links the nonlinearities to coherent Coulomb correlations resulting from non-perturbative excitation of the Landau system.

HL 23.10 Tue 12:15 POT 112

Ultrafast long-range energy transport via light-matter coupling in organic semiconductor films — •Raj Pandya and Akshay Rao — Cavendish Laboratory, JJ Thomson Avenue, CB3 0HE, Cambridge, United Kingdom

Efficient energy transport over macroscopic length scales is highly desirable in organic semiconductors. Here, we show this can be achieved at room temperature in a range of chemically diverse, organic semiconductor thin films through strong light-matter coupling to form exciton-polaritons, despite the absence of an external cavity. We directly visualize energy transport via femtosecond transient absorption microscopy with sub-10 fs temporal and sub-10 nm spatial precision and find energy transport lengths of up to ~270 nm at effective velocities of up to ~5 x106 m s-1. We find additional evidence of strong light-matter coupling via peak splittings in the reflectivity spectra and emission from collective polariton states. These results and the design rules that follow will enable a new generation of organic optoelectronic and light harvesting devices based on robust cavity-free exciton-polaritons.