HL 8: Ultrafast Phenomena I

Time: Monday 15:00–18:00 Location: EW 015

HL 8.1 Mon 15:00 EW 015

Ultrafast Microbeam Diffraction at 15-keV — •Johannes Otto[1,2,3], Leon Brauns[1,2], Rudolf Haindl[1,2], Jan Gerrit Horstmann[1,2,4], Armin Feist[1,2], Murat Sivis[1,2], and Claus Ropers[1,2,3] — [1]Department of Ultrafast Dynamics, Max Planck Institute for Multidisciplinary Sciences, Göttingen, Germany — [2]4th Physical Institute, University of Göttingen, Göttingen, Germany — [3]Max Planck School of Photonics — [4]Present address: Department of Materials, ETH Zürich, Zürich, Switzerland

Ultrafast electron diffraction facilitates the investigation of non-equilibrium structural dynamics [1,2]. In this contribution, we present the development and application of a dedicated setup for microbeam ultrafast electron diffraction (μ-UED), enabling the study of particularly small samples and heterostructures, and promoting experiments on the influence of structural inhomogeneities in non-equilibrium phenomena. We report on the characterization of the spatial and temporal electron beam properties and discuss first time-resolved experiments on a laser-induced structural phase transformation in 1-T tantalum disulfide.

[1] G. Sciaini and R. J. D. Miller, *Rep. Prog. Phys.* **74**, 096101 (2011).

[2] D. Filippetto et al., *Rev. Mod. Phys.* **94**, 045004 (2022).

HL 8.2 Mon 15:15 EW 015

Time-Domain Coherent Phononics in a Rashba Material — •Peter Fischer, Julian Bär, Moritz Cimander, Volker Wiechert, Alfred Leitenstorfer, and Davide Bossini — Department of Physics and Center for Applied Photonics, University of Konstanz, D-78457 Konstanz, Germany

On the basis of spin-orbit coupling, the emerging research field of spintronics is exploring the possibility of using the spin of electrons instead of their charge as carrier of information. This fundamental interaction also enables the conversion of spin currents into charge currents and vice versa, crucial for linking spintronics and conventional electronics. The Rashba effect, an additional contribution to the spin-orbit coupling in materials lacking inversion symmetry, attracts particular attention in this context. We propose that the excitation of coherent phonons of a specific symmetry has the potential to modulate the bulk Rashba spin-orbit coupling and thus the spin-to-charge conversion efficiency on the ultrashort time scale. In our research, we focus on BiTeI, a layered semiconductor exhibiting a giant bulk Rashba effect. On this material, we perform systematic time-resolved pump-probe measurements of the transient reflectivity. The analysis of the acquired time traces confirms the successful excitation of the targeted coherent phonons at a frequency of 2.7 THz. Tuning the pump-photon energy into the visible, near-infrared and even mid-infrared spectral range, we disclose the most favorable conditions for optical generation of these lattice dynamics and thus, in perspective, for the manipulation of the bulk Rashba spin-orbit coupling.

HL 8.3 Mon 15:30 EW 015

Light-Induced Nonthermal Phase Transition to the Topological Crystalline Insulator State in SnSe — •Stefano Mocatti, Giovanni Marini, and Matteo Calandra — Department of Physics, University of Trento, Via Sommarive 14, 38123 Povo, Italy

Femtosecond pulses have been used to reveal hidden broken symmetry phases and induce transitions to metastable states. However, these states are mostly transient and disappear after laser removal. Photoinduced phase transitions toward crystalline metastable states with a change of topological order are rare and difficult to predict and realize experimentally. In this presentation, by means of constrained density functional perturbation theory and non-perturbative light-induced quantum anharmonicity, we show that ultrafast lasers can permanently transform the topologically trivial orthorhombic structure of SnSe into the topological crystalline insulating rocksalt phase via a first-order nonthermal phase transition (1). We describe the reaction path and evaluate the critical fluence and possible decay channels after photoexcitation. Our simulations of the photoexcited structural and vibrational properties are in excellent agreement with recent pump-probe data in the intermediate fluence regime below the transition, with an error on the curvature of the quantum free energy of the photoexcited state that is smaller than 2%.

(1) S. Mocatti, G. Marini, M. Calandra, J. Phys. Chem. Lett. 2023, 14, 41, 9329-9334

HL 8.4 Mon 15:45 EW 015

Photoinduced charge density waves in transition metal dichalcogenides — •Kris Holtgrewe, Giovanni Marini, and Matteo Calandra Buonaura — University of Trento, Italy

Phase transitions induced by ultrafast photoexcitation are an active area of research for materials whose properties can be tuned by light. Transition metal dichalcogenides (TMDs) are a promising class of such materials. Previous studies have demonstrated that photoexcited single layers of $MoTe_2$ and WTe_2 show charge density wave (CDW) orders which can be controlled by the photoexcitation strength [1].

In this work, we investigate whether similar phase transitions occur in other 2D TMD homologues, namely MoS_2, WS_2, $MoSe_2$, and WSe_2. Using constrained density functional theory (cDFT) [2], we infer supercell reconstructions from the unstable phonon dispersions of the photoexcited TMDs and analyse their impact on the electronic properties. Our findings reveal that all TMDs undergo a similar CDW-like structural distortion at a material-dependent critical photoexcitation strength. This distortion is characterised by the formation of metal-metal bonds, driven by a charge redistribution between the d orbitals of the metal atoms. Our results provide a comprehensive understanding of the underlying microscopic processes and pave the way for a general strategy to identify further candidates for photoinduced phase transitions.

[1] G Marini et al. Phys. Rev. Lett. 127, 257401 (2021)

[2] G Marini et al. Phys. Rev. B 104, 144103 (2021)

HL 8.5 Mon 16:00 EW 015

Polaron Formation Dynamics in BiOI Nanoplatelets Studied by Time-Resolved PEEM — •Matthias Franz Kestler[1,2], Kyung Chul Woo[2], Justin W. X. Lim[2], Lucas M. Prins[1], Jochen Feldmann[1], and Zhi-Heng Loh[2] — [1]Chair for Photonics and Optoelectronics, Nano-Institute Munich, Department of Physics, Ludwig-Maximilians-Universität (LMU), Königinstr. 10, 80539 Munich, Germany — [2]School of Chemistry, Chemical Engineering and Biotechnology, and School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore 637371, Singapore

BiOI is a nontoxic, stable and polar semiconductor, which shows high conversion efficiencies in photocatalytic water splitting. This is due to an ultrafast and effective charge separation also launching coherent phonons after short laser pulse excitation. The coupling of electronic excitations and phonons should also manifest itself in polaronic effects. To gain unambiguous experimental evidence for polaronic effects we have carried out time-resolved photoemission electron microscopy (TR-PEEM) experiments. This technique offers the unique possibility to learn about the dispersion E(k) of the conduction band (CB) and its occupation with electrons as a function of time after pulsed optical excitation. In order to interpret our data it is not sufficient to describe merely the temporal evolution of the electron distributions in a static CB. We observe that the dispersion of the CB changes itself in time. This is in particular monitored around the Gamma point of the CB. This combined temporal change in band-structure and electron distribution can be explained by the formation of a polaronic excitation.

15 min. break

HL 8.6 Mon 16:30 EW 015

Ultrahigh-Sensitive Optical Detection of Coherent Acoustic Phonons in a GaAs/GaAlAs Superlattice — •Marek Karzel[1], Anton Samusev[1], Alexey V. Scherbakov[1], Tetiana Linnik[1], Mario Littmann[2], Andrey V. Akimov[3], Dirk Reuter[2], and Manfred Bayer[1] — [1]Technische Universität Dortmund, 44227 Dortmund, Germany — [2]Universität Paderborn, 33098 Paderborn, Germany — [3]University of Nottingham, NG7 2RD Nottingham, United Kingdom

The optical detection of coherent acoustic phonons is a primary tool in ultrafast acoustics. It engages the application of phonons in quantum technology, which is especially promising for nanoscale communications. The ultrahigh-sensitive detection of optically generated phonons with frequencies above 20 GHz can be accomplished due to optical

probing in the spectral vicinity of polariton resonance, where a strong permittivity dispersion is observed [1, 2]. In this work, we monitor the propagation of the optically generated coherent phonon wave packet in a semiconductor superlattice of $GaAs/Ga_{0.67}Al_{0.33}As$ at a temperature of 10 K. With the experimental scheme, we achieve a remarkably high detection sensitivity confirmed by probing coherent acoustic wave packets with less than (on average) one phonon within the bandwidth of interest. The lattice strain induced by such a weak elastic perturbation is of an order of 10^{-9} with the corresponding lattice displacement of only $\approx 10^{-16}$ m.

[1] A. N. Poddubny, et al., Phys. Rev. B 89, 235313 (2014).
[2] M. Kobecki, et al., Phys. Rev. Lett. 128, 157401 (2022).

HL 8.7 Mon 16:45 EW 015

Dynamic Observation of Projected Potentials in Switching Semiconductor Diodes by Time-Resolved Electron Holography — •Tolga Wagner, Hüseyin Çelik, Simon Gaebel, Dirk Berger, and Michael Lehmann — Technische Universität Berlin, Germany

Off-axis electron holography (EH) in a transmission electron microscope (TEM) provides access to nanometer resolved information about the projected electromagnetic potentials of investigated samples. As existing ways for realizing time-resolved measurements in a TEM (e.g. by stroboscopic illumination) have proven to be disadvantageous for EH, such investigations have so far been limited to static measurements.

Recently, a simple, yet promising approach to realize robust time-resolved measurements of periodic processes with nanosecond time resolution in an electron holographic setup by means of interference gating (iGate) was presented. It's based on the synchronized destruction of an interference pattern, relative to an investigated process, where interferometric information is only generated in short undisturbed intervals of a period.

In this presentation, the first application of iGate to switching semiconductor nanostructures, driven at a repetition rate of 3 MHz is demonstrated. By having access to individual frames of the projected electric potential in the area of space charge regions during switching, iGate provides a completely new visual insight into the dynamics of the involved charge carriers with nanometer and nanosecond resolution.

HL 8.8 Mon 17:00 EW 015

A machine learning approach to non-thermal melting — •Andrea Corradini, Giovanni Marini, and Matteo Calandra — Department of Physics, University of Trento, via Sommarive 14, 38123 Povo, Italy

Optical laser irradiation of an insulator promotes a fraction of its valence electrons to the conduction band. Often, irradiation transfers electrons from bonding to antibonding states, leading to a weakening of covalent bonds between ion cores. This process is responsible for the insurgence of various non-equilibrium phenomena in photoexcited materials, like non-thermal melting, that occurs when bond weakening is so strong that the crystal structure dissolves and ions rearrange into a new disordered structure. Non-thermal melting on the femtosecond timescale has been observed in photoexcited silicon. Expensive ab initio calculations have also been able to model this process, characterizing its microscopic origin and the resulting disordered structure [1]. Here, we find strong evidence of non-thermal melting in photoexcited silicon by fitting gaussian approximation machine learning potentials to photoexcited equilibrium phases of silicon. Electrons in these phases are treated as a hot Fermi gas for metallic configurations and with the two Fermi level approach for insulating ones [2]. Our findings successfully reproduce most results from real-time time-dependent density functional theory at much lower computational cost and allow us to fully characterize the liquid phases of photoexcited silicon.

[1] Liu, Wen-hao *et al.*. *Sci. Adv.* 8, eabn4430 (2022).
[2] Marini, G. and Calandra, M. *Phys. Rev. B* 104, 144103 (2021)

HL 8.9 Mon 17:15 EW 015

Molecular dynamics simulations of ultrafast thermodynamics in SrTi03 — •Fredrik Eriksson, Erik Fransson, Richard Matthias Geilhufe, and Paul Erhart — Departement of physics, Chalmers university of technology, Gothenburg, Sweden

The possibility to control and probe materials in the ultrafast regime has opened up several new areas in fundamental research and engineering. In this regard characterization of ultrafast time-dependent thermodynamic properties within the material is crucial [1]. Here, we analyze these aspects by molecular dynamics for the cubic perovskite SrTi03. In particular we artificially excite the ferroelectric optical phonon mode at the zone center, mimicking a laser response. The mode interactions and decay pathways are analyzed and compared to theoretical and experimental data. We find that after about 500fs of pumping the ferroelectric mode, a higher frequency optical mode at Gamma, around 6THz, is strongly excited and pushed out of its equilibrium distribution. This observation is in good agreement with recent experimental work using time-resolved X-ray scattering where this upconversion was also observed [2]. The method allows for a detailed understanding of the entropy production in the system from first principles. In addition, the microscopic mechanism responsible for the upconversion are outlined.

[1] Caprini et al. arXiv:2302.02716v2 (2022)
[2] Kozina et al. Nat. Phys. 15, 387-392 (2019)

HL 8.10 Mon 17:30 EW 015

Ultrafast Nanobeam Electron Diffraction of Charge-Density Wave Phase Transitions at Megahertz Rates — •Till Domröse[1,2] and Claus Ropers[1,2] — [1]Max Planck Institute for Multidisciplinary Sciences, Göttingen, Germany — [2]4th Physical Institute, University of Göttingen, Germany

Ultrafast electron diffraction (UED) is capable of unveiling ultrafast structural dynamics in functional materials [1]. Its intrinsic spatial averaging, however, limits the characterization of nanoscale heterogeneity, which often decisively influences the dynamics, or even is a source of functionality itself. Here, we demonstrate how the high-coherence electron source of the Göttingen Ultrafast Transmission Electron Microscope (UTEM) [2] enables the formation of highly collimated, nanometer-sized, femtosecond electron pulses for UED. We investigate laser-induced charge-density wave dynamics in the strongly-correlated materials 1*T*-TaS_2 and 1*T*-$TaTe_2$ at an unprecedented repetition rate of up to 2 MHz. Therein, the high reciprocal-space resolution allows us to identify a light-induced hexatic state via a three-dimensional characterization of transient disorder [3]. Furthermore, the significant signal enhancement in combination with the small probe volume facilitates access to laser-induced phase transitions with high sensitivity, paving the way for the investigation of non-equilibrium structural dynamics in heterogeneous systems on their intrinsic timescales.

[1] D. Filippetto *et al.*, Rev. Mod. Phys. **94**, 045004 (2022)
[2] A. Feist *et al.*, Ultramicroscopy **176**, 63-73 (2017)
[3] T. Domröse *et al.*, Nat. Mater. **22**, 1345-1351 (2023)

HL 8.11 Mon 17:45 EW 015

Institute of Physics, Rostock University, 18051 Rostock, Germany — •Siamak Pooyan and Dieter Bauer — Institute of Physics, Rostock University, 18051 Rostock, Germany

It has been found previously that the presence or absence of topological edge states in the Su-Schrieffer-Heeger (SSH) model in its topological and trivial phase, respectively, has a huge impact on harmonic generation spectra. More specifically, the yield of harmonics for harmonic orders that correspond to photon energies below the band gap is many orders of magnitude different in the trivial and topological phase. It is shown in this work that this effect is still present if nearest-neighbor electron-electron interaction is taken into account, i.e., if a Hubbard term is added to the SSH Hamiltonian. To that end, finite SSH-Hubbard chains at half filling are considered that are short enough to be accessible to exact diagonalization but already showing edge states in the topological phase. We show that the huge difference in the harmonic yield between trivial and topological phase can be reproduced with few-level models employing only the many-body ground state and a few excited many-body states in the trivial and topological phase, respectively.