O 99: Ultrafast Electron Dynamics IV

Time: Friday 10:30–13:00 Location: H11

O 99.1 Fri 10:30 H11

Large-amplitude coherent phonon dynamics in a van der Waals ferroelectric — •Jan Gerrit Horstmann[1], Quintin Meier[2], Thomas Lottermoser[1], and Manfred Fiebig[1] — [1]Dept. of Materials, ETH Zurich, Switzerland — [2]Institut Néel, CNRS UPR2940, Grenoble, France

We explore the all-optical control of coherent phonon dynamics in the van der Waals ferroelectric WTe_2. Double-pulse optical excitation is harnessed to enhance the amplitude of the prominent interlayer shear mode beyond the limit of single-pulse excitation, with the resulting structural dynamics monitored via time-resolved second harmonic generation. A strong correlation between shear phonon amplitude and damping suggests higher-order coupling to lower-lying modes. At largest initial phonon amplitudes, Fourier transforms of the delay-dependent SHG traces reveal a frequency contribution at twice the shear-mode frequency. We discuss second-harmonic phonon generation and ultrafast phononic modulation of the nonlinear susceptibility as potential underlying mechanisms. Coherent vibrational control over relaxation pathways and electronic properties of ferroic van der Waals materials could enable novel types of devices for energy-efficient electronics.

O 99.2 Fri 10:45 H11

Giant acceleration of polaron transport by ultrafast laser-induced coherent phonon — •Huimin Wang[1,2], Xinbao Liu[1,2], Shiqi Hu[1,2], Daqiang Chen[1,2], Qing Chen[1,2], Cui Zhang[1,3], Mengxue Guan[1,2], and Sheng Meng[1,2,3] — [1]Beijing National Laboratory for Condensed Matter Physics and Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China. — [2]School of Physical Sciences, University of Chinese Academy of Sciences, Beijing 100190, China. — [3]Songshan Lake Materials Laboratory, Dongguan, Guangdong 523808, China

Polaron formation is ubiquitous in polarized materials, but severely hampers carrier transport for which effective controlling methods are urgently needed. Here, we show that laser-controlled coherent phonon excitation enables orders of magnitude enhancement of carrier mobility via accelerating polaron transport in a prototypical material, lithium peroxide (Li2O2). The selective excitation of specific phonon modes, whose vibrational pattern directly overlap with the polaronic lattice deformation, can remarkably reduce the energy barrier for polaron hopping. The strong nonadiabatic couplings between the electronic and ionic subsystem play a key role in triggering the migration of polarons, via promoting phonon-phonon scattering within subpicoseconds. These results extend our understanding of polaron transport dynamics to the non-equilibrium regime and allow for optoelectronic devices with ultrahigh on-off ratio and ultrafast responsibility, competitive with those of state-of-the-art devices fabricated based on free electron transport. H.M.Wang et al. Sci.Adv.9,eadg3833(2023).

O 99.3 Fri 11:00 H11

Ultrafast charge density wave, Van Hove singularity and flat-band dynamics in the Kagome metal CsV_3Sb_5 — •Mohamed Amine Wahada[1], Chris Nicholson[1], Andrea Capa Salinas[2], Brenden R. Ortiz[2], Lawson Lloyd[3], Tommaso Pincelli[3], Tulio de Castro[1], Martin Wolf[1], Ralph Ernstorfer[3], Stephen D. Wilson[2], and Laurenz Rettig[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany — [2]Materials Department and California Nanosystems Institute, University of California Santa Barbara, Santa Barbara, California 93106, USA — [3]Institut für Optik und Atomare Physik, Technische Universität Berlin, 10623 Berlin, Germany

The Kagome metal family AV3Sb5 (A=K, Rb, Cs) features an unconventional superconducting phase, coexisting with a parent charge density wave (CDW) phase. The origin of the CDW is still under debate. Moreover, this family exhibits a number of Van Hove Singularities (VHS) and a flat-band (FB) below the Fermi level, offering a unique platform for high electronic correlation. In order to gain insight into these properties, we study the ultrafast dynamics in CsV3Sb by using time and angle-resolved photoemission spectroscopy (trARPES). We discuss the ultrafast melting of CDW order, concomitant with a shift and a broadening for both the FB and the VHS, all being modulated by coherent phonon modes.

O 99.4 Fri 11:15 H11

Ultrafast Nonequilibrium Dynamics of Room Temperature Charge Density Wave Fluctuations in 1T-$TiSe_2$ — •Hibiki Orio[1], Samuel Beaulieu[2], Sarath Sasi[2], Akib Jabed[2], Muthu P.T. Masilamani[1], Maximilian Ünzelmann[1], Jan Minar[3], Friedrich Reinert[1], Kai Rossnagel[4], Sotirios Fragkos[2], and Jakub Schusser[1,3] — [1]Universität Würzburg, Experimentelle Physik VII and Würzburg-Dresden Cluster of Excellence, Würzburg, Germany — [2]Université de Bordeaux - CNRS - CEA, CELIA, Talence, France — [3]New Technologies-Research Center, University of West Bohemia, Pilsen, Czech Republic — [4]Institute of Experimental and Applied Physics, Kiel University, Kiel, Germany

$TiSe_2$ exhibits a 2×2×2 charge density wave (CDW) phase below 200 K, driven by the interplay between electron-hole and electron-lattice interactions. Nanoscale CDW domains, referred to as CDW fluctuations, persist even above the transition temperature. This exotic phase provides an intriguing platform for exploring the robustness of electron-hole interactions and electron-phonon coupling at elevated temperatures. Using time- and polarization-resolved XUV momentum microscopy, we investigate CDW fluctuations in $TiSe_2$ and their ultrafast light-induced dynamics. Our band structure mapping reveals that the backfolded Se 4p band associated with the CDW phase remains detectable even at room temperature. Furthermore, we demonstrate the ultrafast melting of these CDW fluctuations on an electronic timescale, followed by rapid recovery modulated by coherent amplitude phonon modes.

O 99.5 Fri 11:30 H11

Coherent phonons and quasiparticle renormalization in semimetals from first principles — •Christoph Emeis, Stephan Jauernik, Sunil Dahiya, Yiming Pan, Carl E. Jensen, Petra Hein, Michael Bauer, and Fabio Caruso — Kiel University, Germany

Coherent phonons offer a powerful means to manipulate structural and electronic properties of materials on ultrafast timescales, making them a key tool for exploring light-induced phase transitions and non-equilibrium dynamical phenomena.

We present an *ab initio* theory for the displacive excitation of coherent phonons in semimetals. We formulate a computational workflow for modelling light-induced structural changes and the resulting transient band-structure renormalization following excitation with a femtosecond laser. This framework is based on first-principles simulations of ultrafast electron and coherent phonon dynamics in the presence of electron-phonon interactions. We validate our approach through a combined theoretical and experimental study of coherent phonons in the semimetal antimony. The robust agreement between photoemission experiments and simulations corroborates our approach and paves the way for new opportunities to control structural and electronic degrees of freedom in semimetals via coherent phonon excitation.

C. Emeis *et al.* arXiv:2407.17118

O 99.6 Fri 11:45 H11

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[1,3], Uwe Bovensiepen[1,3], and Michael Horn-von Hoegen[1,3] — [1]Department of Physics, University of Duisburg-Essen, Germany — [2]Department of Physical Chemistry, Fritz Haber Institute, Germany — [3]Center for Nanointegration Duisburg-Essen, Germany

In condensed matter, microscopic excitations interact on ultrafast time scales. The combination of suitable time domain experiments allows the analysis of processes such as the energy transfer from electrons to the nuclei subsequent to femtosecond laser pulse excitation. In this study, we used 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). While the electrons lose their excess energy within 400 fs, the lattice vibrational displacements gradually increase over 3-8 ps. Within the time gap, the energy is transiently stored in high-frequency phonon modes as simulated in a three-temperature model. Their temperature is experimentally accessible by the transient electron temperature after

equilibration with such hot phonons. The excitation of low-frequency phonons and the subsequent thermalization and equilibration of the lattice are facilitated by anharmonic phonon-phonon coupling within the Pb film.

O 99.7 Fri 12:00 H11

Resolving ultrafast atomic motion in WTe_2 — •Hanqian Lu[1,2], Victoria C. A. Taylor[2], Hyein Jung[1,2], Jannik Malter[1,2], Ralph Ernstorfer[1,2], and Yoav W. Windsor[1,2] — [1]Technische Universität Berlin, 10623 Berlin, Germany — [2]Fritz Haber Institute of the Max Planck Society, 14195 Berlin, Germany

WTe_2 is a layered transition metal dichalcogenide with a distorted layered structure leading to distinct material properties. Here we study its lattice dynamics following ultrafast photoexcitation, using femtosecond electron diffraction (FED). We observe a strong coherent lattice response as well as growth of incoherent atomic vibrations, which we qualitatively describe as previously done. With the goal of describing such motions microscopically, we develop a quantitative model that extracts both the coherent and the incoherent atomic motions of each W and Te atom in the unit cell.

O 99.8 Fri 12:15 H11

Structural dynamics of the silicon (111)-(7×7) surface upon optical excitation studied by ultrafast reflection high-energy electron diffraction — •Jonas Darius Fortmann[1], Birk Finke[1], Christian Brand[1], and Michael Horn-von Hoegen[1,2] — [1]Faculty of Physics, University of Duisburg-Essen, 47057 Duisburg, Germany — [2]Center for Nanointegration (CENIDE), University of Duisburg-Essen, 47057 Duisburg

We present first results on the ultrafast structural dynamics of the Si(111)-(7×7) surface subsequent to an optical excitation. The surface is excited by an 800 nm fs-laser pulse at various fluences. The structural dynamics is followed through ultrafast reflection high-energy electron diffraction (URHEED) at a sample temperature of 80 K by means of the Debye-Waller effect. The surface has been excited for incident fluences $> 1.6\,\mathrm{mJ/cm^2}$ at a drop of RHEED intensity by only 0.8%. For the highest fluence of $5.4\,\mathrm{mJ/cm^2}$ the intensity drop was 2% which is equivalent to a rise of surface temperature by 7 K. The fluence dependence is explained by linear absorption through the metallic surface state of the 7×7 reconstruction. For higher fluences we observe an additional three photon absorption due to bulk excitation in order to overcome the direct bandgap. The recovery to the groundstate occur via several processes at different time constants. The system has not fully recovered the groundstate after 200 μs, which indicates the population of a long-lived electronic state.

O 99.9 Fri 12:30 H11

Ultrafast imaging ellipsometry or interferometry - Which one is more suitable to measure the dielectric function of laser-excited materials? — •Markus Olbrich, Theo Pflug, Andy Engel, and Alexander Horn — Laserinstitut Hochschule Mittweida, Hochschule Mittweida, Technikumplatz 17, 09648 Mittweida, Germany

Changes of the dielectric function of materials due to excitation by ultrafast laser radiation are crucial for understanding fundamental processes such as absorption of electromagnetic radiation or the relaxation of the excited electrons resulting in changes in the density of state of the material. The transient dielectric function can either be measured by ultrafast ellipsometry or interferometry. This study evaluates the advantages and disadvantages of each method exemplarily for an excited thin gold film induced by ultrafast laser radiation ($\lambda = 800\,\mathrm{nm}$, $\tau_H = 40\,\mathrm{fs}$) at fluences below and above the ablation threshold. The focus of the presentation is on characterizing the methodology including the experimental effort, the obtained signal-to-noise ratio, the data evaluation particularly in the limits of the applied optical model, as well as the physical interpretation of the measured data.

O 99.10 Fri 12:45 H11

Anisotropic electronic response in hexagonal boron nitride to laser excitations from real-time time-dependent density functional theory — •Cheng Wang, Markus E. Gruner, and Rossitza Pentcheva — Department of Physics, University of Duisburg-Essen

Hexagonal boron nitride (h-BN) has a graphite-derived structure where nitrogen (N) and boron (B) atoms occupy alternating sites. In the framework of real-time time-dependent density functional theory (RT-TDDFT), as implemented in the Elk code, we systematically study the response of bulk and single layer h-BN to laser pulses with in- and out-of-plane polarization, different photon energies, duration and fluences. Our analysis focuses on transient charge redistribution and changes in occupation numbers, revealing a marked dependence on both polarization and frequency in bulk and monolayer h-BN. For a photon energy of 4.8 eV, slightly above the DFT band gap, we find a notable charge transfer from N to B p_z-orbitals in both bulk and monolayer for in-plane polarization. At 9.5 eV, excitations occur primarily between occupied and unoccupied N p_x-orbitals with a small charge transfer to B. In the monolayer, these excitations are suppressed for out-of-plane polarization. Finally, we compare our findings for monolayer h-BN with analogous calculations for graphene.

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