O 101: Focus Session: Structural Dynamics in Nanoscale Materials, Probed by Ultrafast Electron Pulses II

Time: Thursday 15:00–17:30 Location: HE 101

Invited Talk O 101.1 Thu 15:00 HE 101

Ultrafast Structural Dynamics in Organic Molecular Solids — •Heinrich Schwoerer — Max Planck Institut für Struktur und Dynamik der Materie, Hamburg, Germany

Research on functional organic materials is driven by their chemically tunable optoelectronic properties which have led to many commercial applications, notably including molecular semiconductors such as organic light emitting diodes (OLEDs) and organic photodetectors. Comprised of molecules with strong internal covalent bonds which determine the optical characteristics, the macroscopic electronic properties of such crystals are predominantly set by the structural order, which is in turn determined by weak intermolecular forces. This hierarchy of forces opens two approaches for structural dynamics investigations: making use of the crystalline phase of organic molecules to study photo-induced chemical reactions (within the limitations of the confinement), and, looking beyond the unit cell, observing the ultrafast microscopic dynamics responsible for photo-induced macroscopic phase transitions in the entire organic crystal. We will introduce intriguing properties of organic molecular crystals and present recent examples of either of these approaches.

Invited Talk O 101.2 Thu 15:30 HE 101

Ultrafast Electronic Band Gap Control and Self-Protection from a Photoinduced Phase Transition in an Excitonic Insulator — •Julia Stähler — Dept. of Physical Chemistry, Fritz Haber Institute Berlin, Faradayweg 4-6, 14195 Berlin

Ta_2NiSe_5 is proposed to support an excitonic insulator phase below $T_C = 328$ K combined with a structural change. The former occurs in small gap semiconductors with strong electron-hole interaction where excitons form spontaneously and condense into a new insulating ground state. We study the ultrafast electron and lattice dynamics of Ta_2NiSe_5 by means of time- and angle-resolved photoemission spectroscopy (trARPES) and time-resolved coherent optical phonon spectroscopy. We find that the low temperature structural phase persists even for high excitation densities and the photoinduced structural phase transition is hindered by absorption saturation of excitation pulses at a fluence of $F_C = 0.2$ mJ cm^2. We also show that the electronic band gap can be optically controlled by tuning the excitation density. Below F_C, the band gap shrinks transiently due to photoenhanced screening of the Coulomb interaction. However, above F_C, the band gap transiently widens at the Gamma point and recovers to its equilibrium value after 1.5 ps. Hartree-Fock calculations reveal that the band gap widening is due to photoenhancement of the exciton condensate density, persisting until interband carrier relaxation occurs. These results demonstrate the possibility to manipulate exciton condensates with light and gain ultrafast band gap control.

15 min. break

O 101.3 Thu 16:15 HE 101

Optically excited structural transition in atomic wires on surfaces at the quantum limit: a femtosecond ultrafast surface electron diffraction study — Tim Frigge[1], Bernd Hafke[1], Tobias Witte[1], Boris Krenzer[1], Manuel Ligges[1], Uwe Bovensiepen[1], Stefan Wippermann[2], Wolf Gero Schmidt[2], and •Michael Horn-von Hoegen[1] — [1]Fakultät für Physik und CENIDE, Uni Duisburg-Essen, Germany — [2]Theoretische Materialphysik, Uni Paderborn, Germany

The Indium induced (4x1) reconstruction on Si(111) is the prototype for 1D atomic wires at surfaces. At 130 K a metal-insulator transition to the (8x2) ground state takes place. A Peierls-like distortion causes periodicity doubling, opening of a band gap, and formation of a CDW. The non-equilibrium structural dynamics is studied by ultrafast RHEED with a fs-laser system in pump probe setup at temporal resolution better than 300 fs. Upon photo excitation the (8x2) ground state is driven in 350 fs to the (4x1) excited state as observed through the transient spot intensity changes. The transition is described in an accelerated displacive excitation scenario which relies on transient changes in the potential energy surface. The strong coupling between substrate and adsorbate is responsible for the sub-picosecond structural response by dephasing and damping the characteristic phonons in 1/4th of their oscillation period. Transient heating of the In atoms from 30 K to 80 K occurs delayed on a time scale of 2.2 ps. Thus the phase transition is driven by electronic entropy and not thermally.

O 101.4 Thu 16:30 HE 101

Phase-ordering kinetics of charge density waves mapped by ultrafast LEED — •Simon Vogelgesang[1], Gero Storeck[1], Jan Gerrit Horstmann[1], Theo Diekmann[1], Murat Sivis[1], Sebastian Schramm[1], Kai Rossnagel[2], Sascha Schäfer[1], and Claus Ropers[1] — [1]IV. Physical Institute - Solids and Nanostructures, University of Göttingen, Germany — [2]Institute for Experimental and Applied Physics, University of Kiel, Germany

Recently, ultrafast low-energy electron diffraction was developed for the investigation of structural dynamics in thin films in a transmission geometry [1]. In order to extend this approach to back-scattering diffraction for the analysis of surface dynamics, we employ miniaturized laser-driven electron sources featuring nanometric metal tip photocathodes [2]. Here, we investigate dynamics of charge density wave (CDW) phases at the surface of the quasi-two-dimensional material 1T-TaS_2 [3]. Specifically, we trace the laser-induced transition from the room-temperature nearly-commensurate (NC) to the high temperature incommensurate (IC) CDW phase. Performing a thorough spot-profile analysis, we identify a coarsening behavior in the newly created IC phase exhibiting significant CDW disorder. This growth of the IC CDW coherence length is attributed to the annihilation of dislocation-type topological defects, as corroborated by numerical simulations in a Ginzburg-Landau approach.

[1] M. Gulde et al., Science 345, 200 (2014). [2] G. Storeck et al., Structural Dynamics 4, 044024 (2017). [3] S. Vogelgesang et al., Nature Physics (2017), advance online publication, doi:10.1038/nphys4309.

O 101.5 Thu 16:45 HE 101

Ultrafast structural dynamics of transition metal dichalcogenide heterostructures — •Daniela Zahn, Thomas Vasileiadis, Lutz Waldecker, and Ralph Ernstorfer — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany

Vertical heterostructures of two-dimensional crystals offer intriguing new perspectives for the fabrication of novel nanodevices [1]. Since all devices are operated in non-equilibrium conditions, interactions between the different degrees of freedom play an important role in their functionality. These interactions can be studied using femtosecond pump-probe techniques. In particular, femtosecond electron diffraction allows to directly observe the structural response to photoexcitation [2]. Electron-phonon and phonon-phonon coupling as well as electronic and vibrational coupling across interfaces can be studied.

We focus on heterostructures with staggered (=type II) band alignment. This particular band alignment leads to photo-induced charge separation across the interface and is therefore interesting for many applications, e.g. photovoltaic devices. We present a study on multilayer WSe_2/WS_2 heterostructures using two different excitation wavelengths. Different dynamics are observed, suggesting that the interfacial carrier transfer depends on the initial hot carrier distribution in k-space.

[1] A. Geim, I.V. Grigorieva, Nature 499, 419-425 (2013).
[2] L. Waldecker, R. Bertoni, R. Ernstorfer, JAP 117, 044903 (2015).

O 101.6 Thu 17:00 HE 101

Laser control of ultrafast nonthermal melting in silicon. — •Tobias Zier, Eeuwe S. Zijlstra, and Martin E. Garcia — Theoretische Physik, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany

Femtosecond-laser pulses can induce structural phenomena, like, solid-to-solid phase transitions and ultrafast melting in crystalline structures. The main reason for the appearence of such effects is the ultrafast modification of the bonding properties in the induced nonthermal state consisting of extremely hot electrons and nearly not effected cold ions. Although melting is a stochastic process in thermodynamical equilibrium, we show that in the laser excited nonthermal case some coherences are preserved or created. Moreover, by performing ab initio

molecular dynamics simulations of the excitation of silicon by a series of laser pulses we demonstrate that it is possible to control nonthermal melting by light. Analyzing the energy flow in quasimomentum space, we found that the ultrafast disordering atomic motion can be stopped and redirected depending on the delay between the pulses. Essential for the controlling mechanism is the appearance of an intermediate state in the excitation process that shows dominantly thermal phonon squeezing.

O 101.7 Thu 17:15 HE 101

Ab-initio calculation of electron impact ionization cross sections for atoms in exotic electron configurations — •John Bekx[1,2], Sang-Kil Son[1], Robin Santra[1,2], and Beata Ziaja[1,3] — [1]DESY, Hamburg, Germany — [2]University of Hamburg, Hamburg, Germany — [3]INP, Krakow, Poland

Coherent diffraction imaging uses intense femtosecond pulses of hard X-rays. When such pulses interact with the imaged sample, photoelectrons from atomic inner shells are ionized. In light elements, the core holes formed relax predominantly through Auger decay, whose characteristic lifetime is usually between 1 and 10 fs. During this time interval, due to the presence of many energetic electrons excited by X-rays, the ions with core holes may also undergo impact ionization. In this study, we investigate the impact ionization cross sections of ions with such exotic electron configurations and compare them to those of ground state atoms and ions. For calculating the doubly differential cross section, both energy- and angle-resolved, we extend the ab-initio toolkit XATOM [1]. We use the Born approximation for the impact electron, and apply the Hartree-Fock-Slater model for the description of the complex many-electron system under consideration. Our predictions have important implications for understanding radiation damage in X-ray free-electron laser experiments. We also expect applications in the area of electron diffraction, where similar bound-electron configurations could arise.

[1] Z. Jurek, et al., J. Appl. Crystallogr. **49**, 1048-1056 (2016).