O 119: Non-Equilibrium Daynamics in Light-Driven Materials: Theory Meets Experiment

Time: Friday 11:30–13:00 Location: MA 141

O 119.1 Fri 11:30 MA 141

Long-lasting nonequilibrium in the electron system of laser-excited copper — •Sebastian T. Weber and Baerbel Rethfeld — Department of Physics and Research Center Optimas, University of Kaiserslautern, Erwin Schroedinger-Strasse 46, 67663 Kaiserslautern, Germany

After excitation with an ultra-short laser pulse, the electrons in a metal are in a strong nonequilibrium state. During thermalization, they interact also with the lattice, transferring energy to the phonons. On the basis of complete Boltzmann-type collision integrals, a method allowing to treat materials with an arbitrary density of states has been developed [Phys. Rev. B 87, 035139 (2013)]. This method provides insights into the response of different material classes to ultra-fast laser excitation. Here, we focus on the study of copper which possesses a characteristic densities of states with an elevated area at a certain distance below the Fermi edge.

The results show the thermalization of the electrons towards Fermi distributions in a few femtoseconds. However, electron-phonon coupling hinders the electrons' complete thermalization as long as the quasi-temperatures of subsystems differ from each other. We present results showing these thermal and non-thermal non-equilibria and their timescales of relaxation.

O 119.2 Fri 11:45 MA 141

Relaxation of a laser-induced phonon nonequilibrium distribution — •Isabel Klett and Bärbel Rethfeld — Department of Physics and Optimas Research Center, University of Kaiserslautern, Germany

Femtosecond laser irradiation of solids leads to a thermodynamic nonequilibrium within and between the electron and phonon subsystems of the material. Due to collision processes, both subsystems relax into new thermodynamic equilibria within different respective thermalization times. In many studies, the nonequilibrium state within the phonon system is neglected and its influence on other relaxation processes is unclear. Here, we present a model for the description of the phonon nonequilibrium and its relaxation [1]. Phonon-phonon interactions are described with Boltzmann collision integrals. From this, an energy-dependent relaxation time can be extracted and inserted into a relaxation-time approach. Within the frame of this model, we study the thermalization of a nonequilibrium phonon distribution induced by ultrafast laser irradiation. It takes place on a timescale of some hundreds of femtoseconds. Additionally, we discuss the energy transfer between Fermi-distributed electrons and nonequilibrium phonons and compare this to the energy transfer between two equilibrated subsystems.

[1] I. Klett and B. Rethfeld, arXiv:1710.02355

O 119.3 Fri 12:00 MA 141

Hot phason dynamics in 1T-TaS2 probed by ultrafast LEED — •Theo Diekmann[1], Gerrit Horstmann[1], Simon Vogelgesang[1], Gero Storeck[1], Kai Rossnagel[2], and Claus Ropers[1] — [1]4th Physical Institute - Solids and Nanostructures, University of Göttingen, Friedrich-Hund-Platz 1, Germany — [2]Institute for Experimental and Applied Physics, University of Kiel, Germany

We recently developed an ultrafast low-energy electron diffraction (ULEED) setup for the time-resolved study of structural dynamics. To implement this approach in a backscattering geometry, a nanoscopic needle emitter is utilized in a miniaturized electrostatic lens geometry as a high-brightness photoelectron source [1]. In a first application, we investigated phase-ordering kinetics in the structural transition between CDW-phases in 1T-TaS2 [2].

Here, we study collective phase-excitations of the incommensurate CDW state, so-called phasons. By a comparison of the pump-induced intensity suppressions of Bragg- and CDW-satellite-reflections, we determine the time-dependent temperature changes in the respective subsystems. Our results reveal a strong initial energy transfer to the phason system, followed by a long-lived non-equilibrium between phasons and phonons, indicating an inhibited electron-phason relaxation channel. We attribute this observation to a gap-induced decoupling of electron and phason systems.

[1] G. Storeck et al., Structural Dynamics 4.4, 044024 (2017). [2] S. Vogelgesang et al., Nature Physics (2017), advance online publication.

O 119.4 Fri 12:15 MA 141

Photon-dressed states viewed by time- and angle-resolved photoemission spectroscopy — •Sven Aeschlimann[1], Mariana Chavez-Cervantes[1], Razvan Krause[1], Camilla Coletti[2], Kai Rossnagel[3], and Isabella Gierz[1] — [1]Max Planck Institute for the Structure and Dynamics of Matter, Center for Free Electron Laser Science, Hamburg, Germany — [2]Center for Nanotechnology @ NEST, Istituto Italiano di Tecnologia, Pisa, Italy — [3]Institute of Experimental and Applied Physics, Christian-Albrechts-Universität zu Kiel, Kiel, Germany

According to Floquet theory, the coherent interaction between Bloch states and strong photon fields generates side bands of the unperturbed band structure with the possibility to induce topological phase transitions when time reversal symmetry is broken with circularly polarized light[1]. The effect has been observed in Bi_2Se_3 using tr-ARPES[2]. There are predictions for related phase transitions in graphene[1] and transition metal dichalcogenides[3].

We present evidence for the generation of photon-dressed states in graphene and WSe_2 using tr-ARPES at extreme ultraviolet wavelengths. We discuss the key requirements for the experimental observation of these states, including the suppression of coherent interactions between the pump field and the photo-emitted electrons and a careful tuning of pump wavelength as well as pump and probe pulse durations.

[1] T. Oka and H. Aoki, PRB 79, 081406 (2009) [2] Y.H. Wang et al., Science 342, 453 (2013) [3] M. Claassen et al., Nature Communications 7, 13074 (2016)

O 119.5 Fri 12:30 MA 141

Light-induced femtosecond dynamics in C_{60}-films — •Razvan Krause, Mariana Chavez-Cervantes, Sven Aeschlimann, Andrea Cavalleri, and Isabella Gierz — Max Planck Institute for the Structure and Dynamics of Matter, Center for Free Electron Laser Science, Hamburg, Germany

Motivated by the recent discovery of a light-induced superconducting-like state in K_3C_{60} fullerides far above the critical temperature of 20 K [1] we are trying to unveil the underlying mechanism with time- and angle-resolved photoemission spectroscopy (tr-ARPES). We started by photo-exciting carriers across the HOMO-LUMO gap in C_{60} films grown on Si(111) with a resonant 1.5 eV femtosecond laser pulse, revealing an excited state life time of 50 fs. We are now exploring the possibility of driving large structural distortions of the C_{60} molecule through resonant excitation of the T_{1u} phonon mode at 7μm wavelength, which has been proposed to be responsible for the light-induced superconducting-like state in K_3C_{60} [1]. We present optical as well as tr-ARPES data that allow us to assess both the amplitude of the light-induced carbon displacements as well as the response of the electronic structure. We end by discussing the influence of doping and crystalline order for the light-induced C_{60} dynamics.

[1] Mitrano et al., Nature 530, 461 (2016)

O 119.6 Fri 12:45 MA 141

Ultrafast dynamics of bands and bonds in In/Si(111) nanowires probed by trARPES at 500 kHz — •Chris W. Nicholson[1], Andreas Lücke[2], Wolf Gero Schmidt[2], Michele Puppin[1], Laurenz Rettig[1], Ralph Ernstorfer[1], and Martin Wolf[1] — [1]Fritz-Haber-Institut, Berlin, Germany — [2]Universität Paderborn, Germany

In a Born-Oppenheimer description, nuclear dynamics evolve across a free energy surface determined by the occupation of electronic states as a function of nuclear coordinates. Ultrafast photo-induced phase transitions (PIPTs) provide a test case for how the forces and resulting nuclear motion along the reaction coordinate originate from a non-equilibrium population of excited electronic states.

Utilizing femtosecond time and angle resolved photoemission spectroscopy (trARPES) with a novel 500 kHz XUV laser source, we obtain direct access to the transient electronic structure during an ultrafast PIPT in a model system: the structural transition in In/Si(111) nanowires. A detailed reaction pathway is laid out, including temporally separated electronic and structural phase transitions. Comparison with ab initio molecular dynamics simulations reveals the crucial role played by localized photo-holes in shaping the potential energy

landscape. Our detailed insights enable a combined momentum and real space description of ultrafast PIPTs including the ultrafast formation of chemical bonds [1].

[1] Nicholson et al. Submitted (2017)