O 86: Ultrafast Surface Dynamics I

Time: Thursday 15:00–18:00 Location: S052

O 86.1 Thu 15:00 S052

Imaging ultrafast photo-induced dynamics in semiconducting polymer films with time-resolved photoemission electron microscopy — •Andreas Neff and Katrin Siefermann — Leibniz-Institute of Surface Modification, Leipzig, Germany

Organic semiconductors have great potential for applications in opto-electronic devices. By now, it is well established that the performance of devices critically depends on the detailed morphology of the organic semiconducting films [1]. However, a profound understanding of the correlation between morphology and (photo)physical properties is still missing, and this remains a key challenge to overcome technological hurdles in the field.

We address this challenge with a combination of a photoemission electron microscope (PEEM) and a femtosecond laser system. With this setup, we image the morphology of films of organic semiconductors, in particular the size and orientation of crystallites, with a lateral resolution of ∼100 nm. Ultrafast pump-probe experiments allow us to image photo-induced dynamics with a temporal resolution of 150 fs and a lateral resolution of ∼100 nm. Here, we present results from the ultrafast exciton decay dynamics in P3HT (Poly(3-hexylthiophen-2,5-diyl)) films. We find that these decay dynamics are - as expected but not previously detectable - not the same for all locations on the sample. These results demonstrate the potential of time-resolved PEEM to address key questions with regard to the relationship between nanoscale morphology of organic semiconductors and photo-physical properties.

[1] Y. Diao et al., Nat. Commun. 6, 7955 (2015).

O 86.2 Thu 15:15 S052

Electron-lattice interactions in gold clusters and islands studied with ultrafast electron diffraction — •Thomas Vasileiadis[1], Dawn Wells[2], Lutz Waldecker[1], Roman Bertoni[1], Richard Palmer[2], and Ralph Ernstorfer[1] — [1]Department of Physical Chemistry, Fritz-Haber-Institut, Fardayweg 4-6 14195 Berlin Germany — [2]Nanoscale Physics Research Laboratory, School of Physics and Astronomy, University of Birmingham, Edgbaston Birmingham B15 2TT

Metallic nanoclusters possess a number of functionalities including catalytic activity and fluorescent emission that are absent from their bulk counterparts. In order to shed light into these confinement-induced properties, an in-depth understanding of the interactions between the subsystems of charge carriers and the lattice is essential. For this purpose we employ ultrafast electron diffraction to study size-selected gold nanoclusters and nanoislands supported by two-dimensional substrates. After laser excitation of the electronic subsystem, the evolution of lattice temperature and expansion can be probed from the intensity and position of diffraction peaks respectively. The so-called two-temperature-model allows for a quantification of how strongly the lattice couples to electronic excitations. The results are compared with thin films of gold that approach the bulk limit. We also discuss the effects that possibly rise due to electronic and vibrational coupling with the substrate.

O 86.3 Thu 15:30 S052

Non-thermal phonon distribution in out-of-equilibrium aluminium — •Lutz Waldecker[1], Roman Bertoni[1], Jan Vorberger[2], and Ralph Ernstorfer[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft — [2]Max-Planck-Institut für Physik komplexer Systeme

The interaction between electrons and lattice vibrations is central to both ground state as well as out-of-equilibrium properties of solids. We apply an approach based on femtosecond electron diffraction to retrieve these interactions in laser-excited aluminium at various excitation conditions by measuring the temporal evolution of the atomic mean squared displacement. The data is described with a refined version of the two-temperature model, which is inspired by ab initio theory calculations and allows for transient non-thermal phonon distributions. Our work suggests that assuming thermal phonon distributions, i.e. employing a two-temperature model, can lead to systematic errors in the interpretation of time-resolved experiments.

O 86.4 Thu 15:45 S052

Nonequilibrium electron dynamics in laser excited copper, silver and gold — •Sebastian Weber and Baerbel Rethfeld — Fachbereich Physik und Forschungszentrum OPTIMAS, TU Kaiserslautern, Germany

After excitation with an ultrashort 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 ultrafast laser excitation. Here we focus on the study of noble metals which possess characteristic densities of states with an elevated area at a certain distance below the Fermi edge. The results show the thermalization of the electrons within a few femtoseconds. However, electron-phonon coupling hinders the electrons' complete thermalization as long as the temperature of both subsystems differ from each other.

O 86.5 Thu 16:00 S052

Ultrafast Optical Control of the Electronic Properties of ZrTe5 — •Alberto Crepaldi[1], Giulia Manzoni[2], Andrea Sterzi[2], Timo Kuhn[3], Luca Gragnaniello[3], Gabriel Autès[4], Michele Diego[2], Federico Cilento[1], Michele Zacchigna[5], Philippe Bugnon[4], Arnaud Magrez[4], Helmuth Berger[4], Mikhail Fonin[3], Oleg Yazyev[4], Marco Grioni[4], and Fulvio Parmigiani[1,2] — [1]Elettra-Sincrotrone Trieste, Italy — [2]Universitá degli Studi di Trieste, Italy — [3]University of Konstanz, Germany — [4]EPFL, Switzerland — [5]C.N.R.-I.O.M., Italy

ZrTe5 has recently attracted considerable interest owing to some unique, albeit only partially understood, properties. The electrical resistivity exhibits a peak at a temperature where the nature of the charge carriers changes from holes to electrons. The observed negative magneto-resistance has been attributed to the presence of Dirac particles, either three-dimensional or two-dimensional and spin-polarized. Our time and angle-resolved photoelectron spectroscopy (tr-ARPES) study has addressed the origin of the anomalous transport behavior of ZrTe5, while showing the possibility to control the electronic properties of this material via sub-ps IR laser pulses. These observations open the way to the exploitation of ZrTe5 as a platform for magnetoelectric optical and thermoelectric transport applications. Finally, by combining ab initio calculations, ARPES and scanning tunneling microscopy (STM) we are contributing to shed light on the topological nature of ZrTe5, which is shown to be close to transition between strong and weak topological insulator phases.

O 86.6 Thu 16:15 S052

Two-Photon Photoemission on ultrathin CoO films — •Mathias Albrecht[1], Konrad Gillmeister[1], Cheng-Tien Chiang[2,1], and Wolf Widdra[1,2] — [1]Institute of Physics, Martin-Luther-Universität Halle-Wittenberg, Halle (Saale), Germany — [2]Max-Planck-Institute für Mikrostrukturphysik, Halle (Saale), Germany

Unoccupied electronic states of ultrathin cobalt oxide films grown on Ag(001) are studied by time- and angle-resolved two-photon photoemission (2PPE) spectroscopy. With pump/probe photon energies of 4.2 and 1.8 eV, a series of states at 3.55, 3.87 and 4.00 eV above E_F could be identified. The strongest feature at 3.55 eV is assigned as $n = 1$ image potential (IP) state of ultrathin CoO films due to its characteristic parabolic dispersion. At the $\overline{\Gamma}$ point, the IP state has a lifetime of approximately 35 fs. Additionally to the IP states near the vacuum level, an unoccupied CoO conductive band state has been found at 2.3 eV above E_F with a lifetime below 30 fs. Our results are discussed with the theoretical electronic structure of CoO and compared with results for NiO films.

O 86.7 Thu 16:30 S052

Hot electron-spin gas drives ultrafast spin density wave transition in Cr — •Chris Nicholson[1], Claude Monney[2], Robert Carley[3], John Bowlan[4], Björn Frietsch[5], Martin Weinelt[5], and Martin Wolf[1] — [1]Fritz-Haber-Institut, Berlin, Germany — [2]University of Zurich, Switzerland — [3]European XFEL GmbH, Germany — [4]Los Alamos National Laboratory, USA — [5]Freie Universitaet Berlin, Berlin, Germany

Transitions to magnetically ordered states are characterised by an or-

der parameter, defined in the (equilibrium) adiabatic limit. However, it is an open question how the order parameter develops on ultrafast timescales following abrupt excitation by an ultrashort laser pulse and if a general description of such ultrafast phase transitions in terms of a theoretically determined order parameter is still possible [1].

We exploit the energy and momentum selectivity of time- and angle-resolved photoemission spectroscopy to address the ultrafast dynamics of the antiferromagnetic SDW photoexcited in epitaxial thin films of chromium. We reveal we are able to quantitatively extract the evolution of the SDW order parameter through the phase transition, which follows the electronic temperature. We observe the complete destruction of antiferromagnetic order on a sub-100fs time scale, which implies that a quasi-equilibrium between electron and spin systems develops on an ultrashort time scale and drives the spin density wave transition in chromium.

[1] P. Beaud et al. Nat. Mater. 13, 1 (2014)

O 86.8 Thu 16:45 S052

Dynamics of photoexcited quasiparticles across the full Brillouin zone of optimally doped $Bi_2Sr_2Cu_2O_{8+\delta}$ probed by time- and angle-resolved XUV photoemission spectroscopy — •Kerstin Hanff[1], Lexian Yang[2], Lars-Philip Oloff[1], Florian Diekmann[1], Gerald Rohde[1], Ankatrin Stange[1], Michael Bauer[1], and Kai Rossnagel[1] — [1]Institute of Experimental and Applied Physics, University of Kiel, 24098 Kiel Germany — [2]Physics Department, Tsinghua University, Beijing 100089, People's Repuplic of China

Conventional angle-resolved photoemission spectroscopy (ARPES) has added tremendously to our understanding of the electronic and superconducting properties of the cuprates, through direct measurement of the momentum-dependent electronic structure, spectral function, and order parameter. Femtosecond time-resolved ARPES has recently provided a complementary direct view on the momentum-dependent dynamics of quasiparticles (QPs) and Cooper pairs. Here, we present the results of time-resolved ARPES on optimally doped $Bi_2Sr_2Cu_2O_{8+\delta}$ using extreme ultraviolet (22 eV) probe pulses delivered by a high-harmonic-generation source. This allows us to study the QP relaxation dynamics in selected momentum-space cuts covering the complete Brillouin zone. Our results show that the relaxation of photoexcited QPs is almost momentum-independent taking place on femto- and picosecond time scale.

O 86.9 Thu 17:00 S052

Femtosecond time-resolved photoemission of the photo-induced valence transition in $YbInCu_4$ — •Florian Diekmann, Lars-Philip Oloff, Kerstin Hanff, Michael Bauer, and Kai Rossnagel — Institut für Experimentelle und Angewandte Physik, Universität Kiel, 24098 Kiel, Germany

Below a temperature of 42 K, $YbInCu_4$ displays a first-order isostructural transition from a semimetallic, paramagnetic local-moment phase with an Yb valence of 2.94 to a metallic, non-magnetic, valence-fluctuating heavy-fermion phase with a Yb valence of 2.84 [1]. Here, we use time- and angle-resolved extreme ultraviolet photoemission spectroscopy to directly probe the photo-induced valence transition in the electronic structure relaxation dynamics near the Fermi level on the femtosecond time scale. Temperature- and pump fluence-dependent results are presented and compared to the results of time-resolved reflectivity measurements.

[1] H. Sato *et al.*, Phys. Rev. Lett. **93**, 246404 (2004).

O 86.10 Thu 17:15 S052

Transient band gap enhancement of the photoexcited excitonic insulator phase in quasi-1D Ta_2NiSe_5 — •Selene Mor[1], Marc Herzog[1], Claude Monney[2], Martin Wolf[1], and Julia Staehler[1] — [1]Fritz-Haber-Institut der MPG, Dept. of Phys. Chem., Berlin, Germany — [2]University of Zurich, Physics Dept., Switzerland

Strong electron-hole interaction in small gap semiconductors can lead to spontaneous formation of excitons resulting in an excitonic insulator (EI) phase. Ta_2NiSe_5 (TNS) has been proposed as candidate for such a phase transition (PT) in combination with a structural change at $T_c \approx 328$ K. In order to unveil how the monoclinic/EI phase stabilizes in TNS, we monitor the non-equilibrium dynamics after photoexcitation using time-resolved optical and photoemission spectroscopy. Time-resolved photoemission in the EI phase shows a strong excitation-density-dependent valence band depletion, until absorption saturation is reached at a critical fluence F_{sat}. This is also reflected in a saturation threshold of the overall transient optical response. A coherent phonon at 4 THz, which is specific of the monoclinic phase, persists above F_{sat}, indicative of a hindered photoinduced PT. Time-resolved photoemission below F_{sat} reveals a band gap shrinking due to photoenhanced screening of Coulomb interaction. However, above F_{sat} this process competes with a delayed band gap widening that we attribute to increased excitonic correlations. After $\sim$ 1.5 ps, excess energy is transferred to the lattice and the band gap shrinking is driven quasi-thermally. These complex dynamics support the key role of electron-hole correlations as origin of the EI phase in TNS.

O 86.11 Thu 17:30 S052

Tracing thermal and nonthermal phase transition by pump-probe schemes. Part I: material behavior under irradiation — •Nikita Medvedev[1], Victor Tkachenko[1], and Beata Ziaja[1,2] — [1]CFEL at DESY, Notkestr. 85, Hamburg, Germany — [2]Institute of Nuclear Physics, Polish Academy of Sciences, Radzikowskiego 152, 31-342 Krakow, Poland

We study theoretically effects of thermal and nonthermal phase transitions in semiconductors irradiated with femtosecond laser pulses. Our developed hybrid code XTANT combines: (i) Monte Carlo method tracing highly-excited nonequilibrium electrons; (ii) Boltzmann collision integrals to calculate energy exchange between atoms and low-energy electrons; (iii) tight binding molecular dynamics following atomic motion on the evolving potential energy surface. This combined approach allows to model both, thermal (triggered by heating of atoms via electron-phonon coupling) and nonthermal (triggered by changes of the interatomic potential through electron excitation) phase transition.

In the first part of the report, we present the XTANT model and discuss the behavior of irradiated semiconductors occurring at different timescales. We demonstrate how the electronic kinetics at femtosecond timescales impacts the atomic dynamics. The nonthermal phase transitions occur within a few hundred femtoseconds. Typically at lower excitation doses, thermal heating may lead to structural changes at picoseconds timescales. Examples of irradiated silicon and GaAs will be presented and compared to experiments.

O 86.12 Thu 17:45 S052

Tracing thermal and nonthermal phase transitions in solids with pump-probe scheme. Part II: Response of optical properties — •Victor Tkachenko[1], Nikita Medvedev[1], and Beata Ziaja[1,2] — [1]CFEL at DESY, Notkestr. 85, 22607 Hamburg, Germany — [2]Institute of Nuclear Physics, Polish Academy of Sciences, Radzikowskiego 152, 31-342 Krakow, Poland

Electronic excitation and atomic dynamics after femtosecond laser pulse irradiation affect electronic structure, thus modifying optical properties of irradiated materials. During the transition of the material to non-equilibrium state and its later relaxation to a new equilibrium phase, its optical properties are influenced by both electronic and atomic dynamics. Optical coefficients can be measured in experiments and used as signatures of phase transitions. Pump-probe techniques tracing evolution of optical properties in irradiated material are modelled with our in-house developed XTANT code. As the code is a unified hybrid model that combines Monte-Carlo scheme, temperature equation, molecular dynamics technique and tight-binding formalism, it allows to trace structural transformation of diamond into graphite. The presented method proved to be feasible to evaluate complex dielectric function of the material and hence to calculate reflection, transmission and absorption coefficients of diamond at each stage of the graphitization process. Comparison with experimental data confirms the accuracy of our method.