O 80: Ultrafast Electron Dynamics at Surfaces and Interfaces I

Time: Thursday 10:30–13:00 Location: H16

O 80.1 Thu 10:30 H16

Ultra-fast core-level dynamics in semiconducting WSe_2 — •Maciej Dendzik[1], R. Patrick Xian[1], Dmytro Kutnyakhov[2], Shuo Dong[1], Federico Pressacco[3], Davide Curcio[4], Steinn Agustsson[5], Michael Heber[2], Jasper Hauer[1], Wilfried Wurth[2,3], Günter Brenner[2], Yves Acremann[6], Philip Hofmann[4], Martin Wolf[1], Laurenz Rettig[1], and Ralph Ernstorfer[1] — [1]Fritz Haber Institute of the Max Planck Society, Faradayweg 4-6, 14915 Berlin, Germany — [2]DESY Photon Science, Notkestr. 85, 22607 Hamburg, Germany — [3]CFEL, Hamburg University, Luruper Chausee 149, 22761 Hamburg, Germany — [4]Aarhus University, 8000 Aarhus C, Denmark — [5]JGU Mainz, Staudingerweg 7, 55128 Mainz, Germany — [6]ETH Zurich, Otto-Stern-Weg 1, 8093 Zurich, Switzerland

The development of femtosecond XUV sources such as the Free-Electron Laser FLASH [1] combined with recent improvements of electron detection (momentum microscopy [2]) enables studying ultra-fast processes governing the excitations of core-level electrons. Here we present a study of femtosecond dynamics of W4f core levels in semiconducting WSe_2. We find that pumping A-exciton at 800 nm induces changes of both position and shape of observed spectra. In particular, we find a distinct change of the symmetric Lorentzian line profile into a metal-like Doniach-Šunjić asymmetric line shape. We follow the time evolution of induced changes and compare it with the dynamics of valence electrons. [1] W. Ackermann et al., Nat. Phot. 1, 336-342(2007) [2] S.V. Chernov et al., Ultramicroscopy 159, 453-463(2015)

O 80.2 Thu 10:45 H16

Practical Limitations of Floquet Topological Insulators — •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

Recently, using Floquet theory, Oka and Aoki [1] realized that graphene illuminated by circularly polarized light provides and experimental realization of the Haldane model [2] that predicts the occurrence of a quantum Hall effect in the absence of magnetic fields. We used time- and angle-resolved photoemission spectroscopy to search for the predicted Floquet sidebands, changes of the photoemission matrix element due to the induced non-zero z-component of the pseudospin, and band gap opening [3]. After careful optimization of all experimental parameters we observed Floquet sidebands in cleaved bulk WSe2 crystals but not in epitaxial graphene. We attribute the absence of Floquet effects in graphene to scattering times that are short compared to the period of the driving field. Our results have important implications for the experimental realization of Floquet topological insulators and related proposals [4].

[1] T. Oka and H. Aoki, PRB 79, 081406 (2009) [2] F. D. M. Haldane, Phys. Rev. Lett. 61, 2015 (1988) [3] M. A. Sentef et al., Nat. Commun. 6, 7047 (2015) [4] M. Claassen et al., Nat. Commun. 7, 13074 (2016)

O 80.3 Thu 11:00 H16

Valley-polarized excitation in singly-oriented monolayer WS_2/Au(111) — •Hauke Beyer[1], Gerald Rohde[1], Antonija Grubisic Cabo[2], Ankatrin Stange[1], Luca Bignardi[3], Daniel Lizzit[3], Paolo Lacovig[3], Silvano Lizzit[3], Kai Rossnagel[1], Philip Hofmann[2], and Michael Bauer[1] — [1]IEAP, Kiel University, Germany — [2]Department of Physics and Astronomy, Aarhus University, Denmark — [3]Elettra Sincrotrone Trieste, Italy

Time- and angle-resolved photoelectron spectroscopy (trARPES) is employed to study the valley-selective excitation and near-surface dynamics of carriers in singly-oriented monolayer WS_2/Au(111). Upon photoexcitation with circularly polarized light ($\lambda \approx 590$ nm), we observe a selective population of both valence and conduction band at K and K′, respectively. Our results are consistent with the fraction of domains with mirror orientation being $\leq$10 %, a value that was determined in diffraction experiments [1]. The quantitative analysis of the data indicates a valley polarization in the valence band of $\approx$ 90 %, while in the conduction band the valley polarization only reaches values of $\approx$ 70 %. We explain this difference by the small spin-orbit splitting of the conduction band, which promotes intervalley scattering processes. Different carrier dynamics of valence and conduction band population further hint to intervalley scattering being of relevance for the depolarization of the conduction band.

[1] Luca Bignardi et al., arXiv:1806.04928v2

O 80.4 Thu 11:15 H16

Interplay between CDW quenching and the excited phonon population in TiSe_2, observed via ultrafast transmission electron diffraction — •William WINDSOR[1], Daniela Zahn[1], Helen Seiler[1], Ralph Ernstorfer[1], Laurenz Rettig[1], and Kai Rossnagel[2] — [1]Fritz-Haber-Institut der MPG (DE) — [2]University of Kiel (DE)

TiSe2 is a 2D material which exhibits a charge density wave (CDW) below 200K, with a CDW-driven energy gap of 0.4eV. Here we study ultrafast structural dynamics of the CDW phase following a mid-IR excitation, slightly above the gap. We observe the quenching of the CDW, followed by Subsequent dynamics of the lattice, and a varying excited population of high- and low-wavevector phonons within the inelastic background. Dynamics of the CDW, the lattice and the inelastic background will be comparatively discussed.

O 80.5 Thu 11:30 H16

Real-space order parameter mapping using ultrafast transmission electron microscopy — •Thomas Danz, Till Domröse, and Claus Ropers — 4th Physical Institute – Solids and Nanostructures, University of Göttingen, Germany

Over the past decades, ultrafast optical techniques have considerably shaped our understanding of homogeneous materials, while transmission electron microscopy has greatly contributed to elucidating atomic structures and compositions on the sub-nanometer scale. Combining these concepts, ultrafast transmission electron microscopy (UTEM) allows for resolving femtosecond dynamics in heterogeneous materials using imaging, diffraction, and spectroscopy [1].

The pulsed electron source of the Göttingen UTEM project employs linear photoemission from a nanoscopic Schottky emitter, delivering highly coherent electron pulses with down to 200 fs pulse duration, 0.6 eV energy width, and sub-1 nm focused beam diameter [2].

Here, we demonstrate the ultrafast real-space mapping of the order parameter for a charge-density wave phase transition in the correlated material 1T-TaS_2. Specifically, we track the evolution of domain patterns on femtosecond to picosecond time and nanometer length scales, extracting characteristic observables not accessible by ultrafast electron or x-ray diffraction.

[1] A. H. Zewail, Science **328**, 187 (2010).
[2] A. Feist, Th. Danz *et al.*, Ultramicroscopy **176**, 63 (2017).

O 80.6 Thu 11:45 H16

Atomically-resolved ultrafast dynamics in CDW-Mott-insulator materials — •Shaoxiang Sheng[1], Mohamad Abdo[1,2,3], Moritz Tritschler[1], Luigi Malavolti[1,2,3], Max Hänze[1,2,3], Gregory McMurtrie[1,2,3], Lukas Arnhold[1], and Sebastian Loth[1,2,3] — [1]Universität Stuttgart, Institut für Funktionelle Materie und Quantentechnologien, Stuttgart, Germany — [2]Max-Planck-Institut für Struktur und Dynamik der Materie, Hamburg, Germany — [3]Max-Planck-Institut für Festkörperforschung, Stuttgart, Germany

Electron-electron interaction plays an important role in strongly correlated systems. This may drive the electrons near the Fermi surface to localize resulting in a metal-to-insulator transition. Layered 1T-TaS2, as a model system, presents rich electronic properties, which go through a series of charge-density wave (CDW) phases into a Mott insulating ground state as the temperature decreases.[1] By combining THz pump-probe spectroscopy and low-temperature scanning tunneling microscopy (STM), we can study ultrafast electron dynamics of the Mott phase at the atomic scale. The pump-probe spectra vary strongly on the scale of one unit cell of the CDW. The results shed new light onto the microscopic dynamics of electron motion in the Mott phase.

[1] Sipos, B., et al. Nature Materials 7, 960-965 (2008).

O 80.7 Thu 12:00 H16

Probing non-equilibrium lattice excitations in 1T-TaS2 by ultrafast LEED — •Gero Storeck[1], Theo Diekmann[1], Gerrit Horstmann[1], Simon Vogelgesang[1], Kai Rossnagel[2], and Claus Ropers[1] — [1]4th Physical Institute - Solids and Nanostructures, University of Göttingen, Germany — [2]Institute for Experimental and Applied Physics, University of Kiel, Germany

Transition-metal dichalcogenides (TMDCs) exhibit a multitude of correlation-induced states, such as charge-density wave (CDW) phases coupled to a periodic lattice distortion (PLD). Optical excitation of these states leads to a transient quench of the CDW gap and the excitation of various fluctuation modes. While the Raman-active amplitude modes lead to strong signatures in optical spectroscopy and are frequently observed in photoelectron spectroscopy, phason excitations remain more elusive [1,2].

Here, we present an ultrafast low-energy electron diffraction (ULEED) study of the incommensurate and nearly commensurate CDW phases of 1T-TaS_2 [3]. We trace the non-equilibrium dynamics of the CDW amplitude, and study the population of phonon and phason modes by their influence on the diffraction intensity of the lattice Bragg peaks, the PLD satellite peaks as well as the diffuse background scattering.

[1] Lee, W. S. et al., Nature Communications 3, (2012).
[2] Liu, H. Y. et al., Physical Review B 88, (2013).
[3] Wilson et al., Advances in Physics 24, 117-201 (1975).

O 80.8 Thu 12:15 H16

Ultrafast exciton dynamics in monolayer WS2 probed by femtosecond time-resolved (non-)linear optical spectroscopies. — •Stefano Calati[1], Selene Mor[1], Sarah King[1,2], and Julia Stähler[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin (DE) — [2]University of Chicago, Department of Chemistry, Chicago, USA

Controlling the optical properties of transition metal dichalcogenides (TMDCs) on ultrashort timescale is foundamental for device applications. Our aim is to unveil the foundamental processes responsible for the exciton dynamics of these materials. We present our work on pristine monolayer WS2 using femtosecond time-resolved reflectivity contrast (tr-RC) and electronic sum frequency generation (tr-eSFG) optical spectroscopies [1]. We present tr-RC measurements of monolayer WS2 with unprecedented time resolution. We discover an unexpected transient blueshift of the A excitonic resonance, which depends on the excitation density and photon energy. We identify and discuss the ultrafast processes responsible for the observed dynamics. Furthermore, we report intense eSFG generation from a monolayer WS2. We identify a resonant contribution of the A exciton, and the transient changes are compared to the dynamics observed in the linear optical experiment. These studies provide the basic framework for future studies on energy and charge transfer at TMDC-based hybrid interfaces.

[1] L. Foglia et al., Appl. Phys. Lett. 109, 202106 (2016)

O 80.9 Thu 12:30 H16

Inter- and intralayer electron dynamics in MoS_2 probed by 2PPE — •Robert Wallauer, Johannes Reimann, Jens Güdde, and Ulrich Höfer — Fachbereich Physik und Zentrum für Materialwissenschaften, Philipps-Universität, 35037 Marburg

We use time- and angle-resolved two photon photoemission (2PPE) with a high-harmonic probe for the investigation of electron dynamics of MoS_2 in momentum space. For this purpose, we combined a high-repetition rate high-harmonic source with a 3D (k_x, k_y, E) electrostatic electron spectrometer [1]. The pump laser is tunable within the visible from around 500 - 700 nm. At our high-harmonic photon energy of 23.5 eV we essentially probe only the first layer, which differs from the bulk with respect to band energies.

By tuning our pump laser above and below the band gap of the first layer we are able to selectively induce the excitation from the valence into the conduction band at $\overline{\mathrm{K}}$ within the first layer or alternatively in deeper layers. For all photon energies, the relaxation to the conduction band minimum at $\overline{\Sigma}$ is observed. The relaxation dynamics, however, varies with pump photon energy, which allows us to distinguish between charge transfer within the first layer and charge transfer between adjacent layers.

[1] R. Wallauer et al., Appl. Phys. Lett. 109, 162102 (2016).

O 80.10 Thu 12:45 H16

Decoding the Ultrafast Formation of a Fermi-Dirac Distributed Electron Gas — •Gerald Rohde, Ankatrin Stange, Arne Müller, Marcel Behrendt, Lars Oloff, Kerstin Hanff, Hauke Beyer, Thies Albert, Petra Hein, Kai Rossnagel, and Michael Bauer — Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, 24098 Kiel, Germany

A photoexcited electron gas is phenomenologically often described within multitemperature models, which consider the electronic system being internally thermalized for all times. Ultrafast nonthermal phenomena taking place right after photoexcitation are still largely unexplored. Time- and angle-resolved photoemission spectroscopy operated near the Fourier-limit and at a temporal resolution of ≈10 fs is used to sample the ultrashort time frame of 50 fs in the formation of a Fermi-Dirac distributed electron gas in graphite following an impulsive photoexcitation [1]. We identify and dissect experimentally characteristic stages associated with different interaction processes among the involved degrees of freedom, which have been predicted theoretically in the past [2,3]. The scenario involves electron-photon, electron-electron, and electron-phonon interaction affecting the response of the system on different timescales. Our results reveal experimentally the complexity of the transition from a nascent nonthermal towards a thermal electron distribution in a graphitic material with reduced screening.

[1] G. Rohde *et al.*, Phys. Rev. Lett. *accepted* (2018)
[2] M. Mittendorff *et al.*, Nano Lett. 14, 1504 (2014)
[3] T. Winzer *et al.*, J. Phys. Condens. Matter 25, 054201 (2013)