O 9: Ultrafast Electron Dynamics at Surfaces and Interfaces 2

Time: Monday 15:00–18:15 Location: H3

O 9.1 Mon 15:00 H3

Two distinct 4*f* states in mixed-valent $TmSe_{1-x}Te_x$ — •Chul hee Min[1], Michael Heber[2], Simon Müller[3], Lukas Wenthaus[2], Markus Schoz[2], Dmytro Kutnyakhov[2], Lenart Dudy[4], Hendrik Bentmann[3], Federico Pressacco[2], Matthias Kalläne[1], Woojae Choi[5], Yong Seung Kwon[5], Friedrich Reinert[3], and Kai Rossnagel[1] — [1]IEAP, CAU Kiel, Germany — [2]DESY, Hamburg, Germany — [3]EP7 and ct.qmat, University of Würzburg, Germany — [4]Synchrotron SOLEIL, Saint-Aubin, France — [5]Dep. of EMS, DGIST, South Korea

From the strong electron correlation effects in rare earth compounds, rich phase diagrams emerge with tunable ground states, which underlie a series of unique physical phenomena and quantum states, including quantum criticality, topological Kondo insulators, and diverse charge-neutral quasiparticle formations. However, despite their importance, the nature of such composite quasiparticles is difficult to characterize because all coherent spectral features develop in a similar way. Using angle-resolved photoemission spectroscopy (ARPES) together with time-resolved PES, we have addressed distinct coherent 4*f* features in unique mixed-valence $TmSe_{1-x}Te_x$. Our findings open the path for future investigations of small energy-scale excitations and may provide a framework for understanding the dynamics and entangled nature of correlated electrons.

O 9.2 Mon 15:15 H3

Time- and angle-resolved photoemission study of magnetic topological insulators $MnBi_2Te_4$ and $MnBi_4Te_7$ — •Paulina Majchrzak[1], Klara Volckaert[1], Deepnarayan Biswas[1], Denny Puntel[2], Wibke Bronsch[2], Federico Cilento[2], Xing-chen Pan[3], Yong Chen[1,3], and Søren Ulstrup[1] — [1]Dept. of Physics and Astronomy, Aarhus University, DK — [2]Elettra - Sincrotrone Trieste, IT — [3]Advanced Institute for Materials Research, Tohoku University, JP

Van der Waals heterostructures comprising layers of intrinsically antiferromagnetic topological insulator (TI) $MnBi_2Te_4$ and non-magnetic TI Bi_2Te_3 offer a rich toolbox for engineering exotic quantum phenomena. Magnetic and transport properties of these materials are strongly affected by the interplay between bulk and surface states with divergent topologies resulting from hybridisation between the top layers.

Here, we disentangle those complex interactions in the time domain with TR-ARPES. We discuss the interband dynamics in bulk and surface states as a function of stacking and surface terminations for $MnBi_2Te_4$ and $MnBi_4Te_7$. Our results fill a knowledge gap in understanding of interlayer coupling in $MnBi_2Te_4$-based heterostructures.

O 9.3 Mon 15:30 H3

Electronic and phonon dynamics in 1T-$TiSe_2$ with ultrafast core-level transient absorption spectroscopy in the extreme ultraviolet — •Tobias Heinrich[1], Hung-Tzu Chang[1], Sergey Zayko[1], Murat Sivis[1,2], and Claus Ropers[1,2] — [1]Max Plank Institute for Multidisciplinary Sciences, Göttingen, Germany — [2]4th Physical Institute - Solids and Nanostructures, University of Göttingen, Germany

Extreme ultraviolet (XUV) transient absorption spectroscopy with high harmonic sources is ideally suited to investigate the complex interplay between lattice and electronic degrees of freedom on ultrafast timescale [1]. Here, we compare the experimental result of highly sensitive XUV transient absorption spectroscopy on the charge density wave (CDW) compound 1T-TiSe2 with density functional theory (DFT) simulations to disentangle electronic and phonon contributions to the transient absorption spectra. In addition to photo excited carriers we observe two coherently excited phonon modes that can be assigned to the A_{1g} optical mode (6 THz) and the A_{1g}* (3.3 THz) amplitude mode associated with the CDW formation [2]. The modes show distinct spectral fingerprints which are reproduced by DFT calculations such that their dynamics can be individually analyzed. It is found that only the amplitude mode vanishes at higher fluences, corroborating the proposed mechanism of non-thermal CDW melting [3].

[1] A. R. Attar *et al.*, ACS Nano **14**, 11, 15829-15840 (2020)
[2] H. Hedayat *et al.*, New J. Phys. **23**, 033025 (2021)
[3] E. Möhr-Vorobeva *et al.*, Phys. Rev. Lett. **107**, 036403 (2011)

O 9.4 Mon 15:45 H3

Coherent phonon-driven transient modulation of a Dresselhaus-type spin splitting in *Td*-WTe_2 — Petra Hein[1], Stephan Jauernik[1], Hermann Erk[1], Lexian Yang[2,3], Yangpen Qi[4,5], Yan Sun[5], Claudia Felser[5], and •Michael Bauer[1] — [1]Institute of Experimental and Applied Physics, University of Kiel, Germany — [2]State Key Laboratory of Low Dimensional Quantum Physics, Department of Physics, Tsinghua University, Beijing, China — [3]Frontier Science Center for Quantum Information, Beijing, China — [4]School of Physical Science and Technology, ShanghaiTech University, China — [5]Max Planck Institute for Chemical Physics of Solids, Dresden, Germany

Time- and angle-resolved photoemission spectroscopy is used to study transient changes of the electronic structure in the Weyl-semimetal *Td*-WTe_2 in response to the excitation of coherent phonons. A Fourier-transform of the three-dimensional experimental data yields phonon-mode resolved insights into such coupling processes. Results of our analysis reveal a transient modulation of a Dresselhaus-type spin splitting of electronic bands that is selectively driven by the excitation of an interlayer shear mode of the layered compound [1]. The results provide real-time insights into electron-phonon coupled processes that are of vital importance for a light-driven topological phase transition in Td-WTe_2.

[1] P. Hein, et al., Nat. Commun. 11, 2613 (2020).

O 9.5 Mon 16:00 H3

Coherent Control of a Metastable Hidden Phase — J. Maklar[1], S. Dong[1], •J. Sarkar[1], Y. A. Gerasimenko[2], T. Pincelli[1], S. Beaulieu[1], P. S. Kirchmann[3], J. A. Sabota[3], S.-L. Yang[3,4], D. Leuenberger[3,4], R. G. Moore[3], Z.-X. Shen[3,4], M. Wolf[1], D. Mihailovic[2], R. Ernstorfer[1,5], and L. Rettig[1] — [1]Fritz-Haber-Institut der MPG, Berlin, DE — [2]Jožef Stefan Institute, Jubljana, Sl — [3]SLAC National Accelerator Laboratory, California, USA — [4]Stanford University, California, USA — [5]Technical University Berlin, DE

In materials with multiple competing orders, ultrashort light pulses can induce metastable states that are not accessible at thermodynamic equilibrium. One of such, the metallic hidden (H) phase of 1T-TaS_2, is of particular interest as it features an order-of-magnitude change in resistivity, promising for novel energy-efficient high-speed memory devices. We use time- and angle-resolved photoemission spectroscopy (trARPES) to investigate the electronic band structure and formation dynamics of the metastable H-state in 1T-TaS_2. The band structure mapping of H-state reveals suppression of correlation effects and metallization, suggesting a critical role of interlayer stacking order of the TaS_2 sheets in the phase transition. The fluence-dependent dynamics provides strong evidence that the charge density wave amplitude mode governs a collective, ultrafast switching pathway to the H-state. This is further corroborated by demonstrating coherent control of the switching efficiency into the H-phase using a multi-pump-pulse excitation scheme.

O 9.6 Mon 16:15 H3

Influence of carbon buffer layer on non-equilibrium carrier dynamics of epitaxial graphene and WS2/graphene heterostructures — •Lukas Bruckmeier[1], Niklas Hofmann[1], Leonard Weigl[1], Johannes Gradl[1], Neeraj Mishra[2,3], Stiven Forti[2], Camilla Coletti[2,3], and Isabella Gierz[1] — [1]Faculty for Physics, University of Regensburg, Regensburg, Germany — [2]Center for Nanotechnology Innovation@NEST, Istituto Italiano di Tecnologia, Pisa, Italy — [3]Graphene Labs, Istituto Italiano di Tecnologia, Genova, Italy

The non-equilibrium photocarrier dynamics of epitaxial graphene on SiC(0001) have been studied in detail in the past. The fact that the graphene layer rests on a second two-dimensional carbon buffer layer (BL) - the well-known $(6\sqrt{3}\times 6\sqrt{3})R30°$ reconstruction - was believed to be of minor importance for the interpretation of the time-resolved data. We use time- and angle-resolved photoemission spectroscopy to show that photoexcitation of the graphene/BL heterostructure is followed by a short-lived transient decrease in binding energy of the Dirac cone indicating a transient charging of the graphene layer with excess electrons. We attribute the transient n-doping of the graphene layer

to the resonant excitation of a direct electronic transition between the non-dispersive states of the BL located 0.5eV below the Fermi level and the Dirac cone. We further show that this direct electronic transition affects the charge transfer dynamics in epitaxial WS2/graphene/BL heterostructures.

O 9.7 Mon 16:30 H3

Ultrafast phonon thermalization in a monolayer crystal — •Hyein Jung — Fritz-Haber-Institut der MPG

Understanding energy flow in semiconductors following a perturbation is key for future applications. Observing dynamic processes on their fundamental time scales enables studying energy transfer processes between intrinsic subsystems, and subsequently understanding the coupling between them. Here we study ultrafast lattice dynamics in monolayer WSe2, carried out using femtosecond (high-energy) electron diffraction (FED).

We studied the lattice response to laser excitation by probing laser-induced variations in Bragg peak intensities, which reflect changes in incoherent lattice vibrations (quantified by atomic mean squared displacements, MSD). We implement a novel approach to analyze such diffraction data, by which we disentangle element-specific vibrational responses in the sub-picosecond time domain.

Through this analysis, we observe a series of steps in the evolution of the lattice response through varying trends of the two elements (W, Se). These observations suggest a cascade of electron-phonon and phonon-phonon scattering processes occurring on short picosecond time scales. We interpret these by means of energy transfer between phonon groups using on ab-initio calculations of the partial phonon density of states phonon. These results demonstrate that our element-specific approach enables a deeper understanding of the cascade of e-ph and ph-ph energy transfer processes following excitation.

O 9.8 Mon 16:45 H3

k-dependent band gap renormalization in monolayer WS2 revealed by tr-ARPES — •Niklas Hofmann[1], Alexander Steinhoff[2], Leonard Weigl[1], Johannes Gradl[1], Tim Wehling[2,3], Sivan Refaely-Abramson[4], Neeraj Mishra[5,6], Stiven Forti[5], Camilla Coletti[5,6], and Isabella Gierz[1] — [1]University of Regensburg, 93040 Regensburg, Germany — [2]University of Bremen, 28359 Bremen, Germany — [3]University of Hamburg, 22607 Hamburg, Germany — [4]Weizmann Institute of Science, 7610001 Rehovot, Israel — [5]Center for Nanotechnology Innovation @NEST, Istituto Italiano di Tecnologia, 56127 Pisa, Italy — [6]Graphene Labs, Istituto Italiano di Tecnologia, 16163 Genova, Italy

Monolayer transition-metal dichalcogenides show strong enhancement of Coulomb interactions due to their reduced dimensionality with immediate effects on both the optical as well as the single-particle band gap. Photogenerated electron-hole pairs have been shown to result in a giant band gap renormalization that has been attributed to efficient screening of the Coulomb interaction. The corresponding band structure changes are predicted to show a pronounced momentum dependence that we resolve using time- and angle-resolved photoemission spectroscopy on monolayer WS2 supported by a graphene substrate. Excellent agreement with ab initio calculations allows us to disentangle the intricate interplay of different many-body contributions to the observed transient band structure renormalization with important implications for the optoelectronic properties of 2D semiconductors.

O 9.9 Mon 17:00 H3

Pump helicity-dependent anisotropic population dynamics in the topological insulator Sb2Te3 — •Jan Böhnke[1], Haydar Altug Yildirim[1,3], Stephan Schmutzler[1], Jaime Sánchez-Barriga[2], Oliver Rader[2], Cornelius Gahl[1], and Martin Weinelt[1] — [1]Fachbereich Physik, Freie Universität Berlin, Berlin, Germany — [2]Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Berlin, Germany — [3]Leibniz-Institut für Astrophysik, Potsdam, Germany

The specific spin texture of Dirac cone like topologically protected surface states (TSS) and the long electron mean free path allow for spin-polarized currents at the surface of topological insulators. Optical control of such currents has been discussed controversially.

We investigated the role of direct and indirect population channels for the unoccupied TSS on Sb2Te3 in 2D momentum space by time and angle-resolved two-photon photoemission spectroscopy. Excitation with 1.55 eV photons leads to an initially anisotropic population of the Dirac cone, dependent on the helicity of the excitation pulse and the azimuthal orientation of the sample. The strongest anisotropy is found in the energy range of the warped Dirac cone. It exhibits predominantly a 3-fold symmetry originating from the symmetry group of the bulk material. This contribution accordingly does not correspond to a macroscopic current in the TSS. On a time scale of 100 fs the population anisotropy is masked by electrons scattering from the bulk conduction band into the TSS.

O 9.10 Mon 17:15 H3

Spatio-temporal imaging of bright and dark excitonic quasiparticles in twisted TMD heterostructures — •Jan Philipp Bange[1], David Schmitt[1], Wiebke Bennecke[1], AbdulAziz AlMutairi[2], Guiseppe Meneghini[3], Daniel Steil[1], R. Thomas Weitz[1], Sabine Steil[1], G. S. Matthijs Jansen[1], Samuel Brem[3], Ermin Malic[3], Stephan Hofmann[2], Marcel Reutzel[1], and Stefan Mathias[1] — [1]I. Physikalisches Institut, Georg-August-Universität Göttingen, Germany — [2]Department of Engineering, University of Cambridge, U.K. — [3]Fachbereich Physik, Philipps-Universität Marburg, Germany

In two-dimensional van-der-Waals semiconductors, the weak Coulomb screening of charge carriers leads to exciting new material properties, such as bright and dark excitons with large binding energies. Consequently, when creating a heterostructure from two transition-metal dichalcogenide (TMD) monolayers with a type II band alignment, interlayer excitons can be formed [1]. Because dark excitons are not directly accessible with optical techniques, the spatial and lateral dynamics on the fundamental nanometer length scale remain largely unexplored. How do dark and interlayer quasiparticles form, relax and diffuse in the presence of a heterojunction, stress fields and inhomogeneities? Here, we address this question using time-resolved momentum and dark-field photoemission microscopy which enables us to study the ultrafast formation dynamics of different excitonic species in twisted WSe_2/MoS_2 heterostructures.

[1] Schmitt *et al.*, arXiv2112.05011 (2021).

O 9.11 Mon 17:30 H3

SHG imaging microscopy of ultrafast charge-transfer dynamics in twisted TMDC heterostructures — •Marleen Axt, Jonas E. Zimmermann, Gerson Mette, and Ulrich Höfer — Fachbereich Physik, Philipps Universität Marburg, Germany

Two-dimensional heterostructures of transition metal dichalcogenides (TMDC) represent very well-defined model systems of van-der-Waals interfaces. Many material combinations feature a type-II band alignment, which can separate photoexcited electrons and holes into different layers through ultrafast charge transfer leading to the formation of so-called interlayer excitons or interface excitons.

We investigate the ultrafast charge-transfer dynamics in TMDC heterostructures as a function of the stacking angle using time-resolved second-harmonic generation (SHG) imaging microscopy. This experimental technique combines the advantages of SHG with high temporal and spatial resolution. For differently twisted MoS_2/WSe_2 heterostructures the electron transfer from WSe_2 to MoS_2 after resonant excitation (1.70 eV) was found to depend considerably on the twist angle. The transfer time is reduced from 85 fs down to 12 fs when going from a larger rotational mismatch (16°) towards 2H-stacking (52°).

O 9.12 Mon 17:45 H3

Spin and charge carrier dynamics at a CuPc/WSe_2 heterostructure — •Sebastian Hedwig[1], Gregor Zinke[1], Benito Arnoldi[1], Martin Aeschlimann[1], and Benjamin Stadtmüller[1,2] — [1]Department of Physics and Research Center OPTIMAS, TU Kaiserslautern, Erwin-Schroedinger-Str. 46, 67663 Kaiserslautern, Germany — [2]Institute of Physics, Johannes Gutenberg University Mainz, Staudingerweg 7, 55128 Mainz, Germany

2D-Van-der-Waals systems are a highly intriguing class of low dimensional materials with promising spin functionalities for future nanoscale spintronic applications. Here, we show our approach to control the spin and charge carrier dynamics of the Van-der-Waals material WSe_2 by the adsorption of CuPc molecules. We conduct time-, angle- and spin-resolved photoemission experiments to investigate the optically excited carrier dynamics at the K- and Σ-points of WSe_2. After an initial spin-selective excitation at the K-point, depending on the pump light polarization [1], we observe that the subsequent intraband scattering from the K- to the Σ-point of the bare WSe_2 conduction band coincides with a change of the carriers spin polarization. Both, the optical excitation and the subsequent relaxation process can be actively modified by appropriate adsorption of CuPc. In particular, the dominant optical excitation at the K-point of WSe_2 is replaced by a

direct interlayer excitation from the CuPc into the WSe_2 layer. We will show that the strength of the interlayer excitation can be tuned and controlled by the polarization of the exciting light field.

[1] Bertoni et al.; Phys. Rev. Lett. 117, 277201 (2016)

O 9.13 Mon 18:00 H3

Subcycle lightwave-ARPES in the strong-field regime — Suguru Ito[1], Manuel Meierhofer[2], Josef Freudenstein[2], Dmytro Afanasiev[2], •Jens Güdde[1], Rupert Huber[2], and Ulrich Höfer[1] — [1]Fachbereich Physik, Philipps-Universität Marburg, Germany — [2]Fakultät für Physik, Universität Regensburg, Germany

Angle-resolved photoemission spectroscopy (ARPES) combined with THz excitation enables the investigation of lightwave-driven Dirac currents in the surface state of topological insulators with sub-cycle time resolution [1]. At low THz frequencies and moderate effective field strengths of a few kV/cm at the surface, we have shown that the current dynamics is dominated by intraband acceleration of the electrons within the surface band of Bi_2Te_3.

Here, we will show how such experiments can be extended to the strong-field regime at driving frequencies of 25-40 THz. This is challenged by strong energy and momentum streaking of the photoelectrons after photoemission as well as the requirement of ultrashort pulses for photoemission which also introduce considerable energy broadening of the ARPES spectra. We will discuss how these challenges can be successfully overcome in order to enable lightwave-ARPES at field strength of $\sim$1 MV/cm despite surface screening. This paves the way for visualizing strong-field phenomena such as high-harmonic generation (HHG) [2] and the emergence of Floquet-Bloch states directly in the band structure on a sub-optical-cycle time scale.

[1] J. Reimann *et al.*, Nature 562, 396 (2018).
[2] C. P. Schmid *et al.*, Nature 593, 385 (2021).