O 19: Poster: Ultrafast Electron Dynamics at Surface and Interfaces I

Time: Monday 18:00–20:00 Location: P2/EG

O 19.1 Mon 18:00 P2/EG

ULTRAFAST TRANSPORT AND ENERGY RELAXATION OF HOT ELECTRONS ON TMDC/Au/Fe/MgO(001) INVESTIGATED BY TIME RESOLVED PHOTOELECTRON SPECTROSCOPY — •Florian Kühne[1], Jesumony Jayabalan[1], Ping Zhou[1], Detlef Diesing[2], Stephan Sleziona[1], Marika Schleberger[1], and Uwe Bovensiepen[1] — [1]Faculty of Physics and Center for Nanointegration, University of Duisburg-Essen — [2]Faculty of Chemistry, University of Duisburg-Essen

Optically excited electrons are of particular interest in solid state physics because analysis of their dynamics allows an understanding of the microscopic interactions. Excited charge carriers in metals and semiconductors relax on a femto- to picosecond timescale due to electron-electron and electron-phonon scattering. Using a back-side pump geometry in time-resolved photoelectron spectroscopy (PES), see Kühne *et al.*, PRR **4**, 033239(2022), and Beyazit *et al.*, PRL **125**, 076803(2020), we demonstrated the analysis of hot electron transport through metallic heterostructures Au/Fe/MgO(001). By pumping the Fe side by femtosecond laser pulses, hot electrons are excited in the Fe layer and injected into the Au layer. They subsequently propagate to the surface, where they are probed by PES. This work aims to employ these heterostructures as electrodes for photo-excited electrons which are injected into transition metal dichalcogenides. Thereby we aim at an analysis of scattering and propagation of electrons through 2D material layers discerning local and non-local effects. Funding by the DFG through Project No. 278162697 - SFB1242 is gratefully acknowledged.

O 19.2 Mon 18:00 P2/EG

T-ReX: a facility for time-resolved optical and angle-resolved photoemission spectroscopies — •Wibke Bronsch[1], Denny Puntel[2], Manuel Tuniz[2], Fulvio Parmigiani[1,2], and Federico Cilento[1] — [1]Elettra-Sincrotrone Trieste, Trieste, Italy — [2]Università degli Studi di Trieste, Trieste, Italy

The T-ReX laboratory at FERMI, Elettra-Sincrotrone Trieste, is a user facility for ultrafast table-top time-resolved spectroscopies [1-3]. Our facility comprises a number of time-resolved optical spectroscopy (TR-OS) setups and an endstation for time- and angle-resolved photoemission spectroscopy (tr-ARPES). The latter is connected to diverse table-top laser sources, allowing for probe energies ranging from 6 to 30 eV. In order to show how the complementarity of these setups can be beneficial for gaining more comprehensive insights on a sample system, we will present our recent experiments on the charge-density-wave materials VTe2 and VSe2, shedding light on phonon dynamics and electronic structures of the samples under investigation.

[1] S. Peli et al., J. Electron Spectrosc. Relat. Phenomena 243, 146978 (2020).
[2] R. Cucini et al., Structural Dynamics 7, 014303 (2020).
[3] M. Perlangeli et al., Optics Express 28, 8819 (2020).

O 19.3 Mon 18:00 P2/EG

Excited state photoemission momentum maps from time-dependent density functional theory — •Melvin Hodžić, Christian S. Kern, Andreas Windischbacher, and Peter Puschnig — Institute of Physics, NAWI Graz, University of Graz, Austria

Angle-resolved photoemission spectroscopy is a powerful experimental technique to reveal the electronic structure of thin films and interfaces. In particular, constant binding energy angular distributions, so called momentum maps, have been shown to be related to the Fourier transform of the electron-emitting initial state molecular orbital, thereby bridging experiment and theory in photoemission orbital tomography (POT). The extension of POT to optically excited states has, of late, been demonstrated via a femtosecond pump-probe setup, paving the way for tracing the momentum distribution of electrons on ultrafast time scales. However, linking the measured momentum maps to the spatial structure of the exciton wave functions has yet to be achieved. We propose a simple procedure which involves a coherent sum of the ground-state Kohn-Sham orbitals that construct the exciton wave function. Here, we validate this approach by solving Casida's equation for a series of organic molecules in the gas phase leading to optical absorption spectra and exciton compositions. With the gained information, momentum maps obtained via linear-response TDDFT are cross-checked by directly simulating angle-resolved photoemission spectra within the framework of real-time TDDFT. To this end, we record the flux of the emitted electrons through a detector surface.

O 19.4 Mon 18:00 P2/EG

Ultrafast all-optical manipulation of the charge-density-wave in VTe2 — •Manuel Tuniz[1], Wibke Bronsch[2], Denny Puntel[1], Davide Soranzio[3], Davide Bidoggia[1], Steven Johnson[3], Maria Peressi[1], Fulvio Parmigiani[1,2], and Federico Cilento[2] — [1]Dipartimento di Fisica, Università degli Studi di Trieste, Italy — [2]Elettra - Sincrotrone Trieste S.C.p.A., Italy — [3]Institute for Quantum Electronics, ETH Zurich, 8093 Zurich, Switzerland

By means of broadband time-resolved optical spectroscopy (TR-OS) we investigated the ultrafast reflectivity changes caused by collective and single particle excitations in the charge-density wave (CDW) system VTe2. This material has been recently subject of investigation since the modifications in its electronic structure triggered by the CDW formation are strongly orbital-dependent and may give rise to a topological change in specific bands [1]. In our contribution, we show by means of TR-OS measurements the possibility to optically excite the amplitude mode (AM) of the CDW phase and therefore couple to the CDW condensate [2]. Moreover, through double pump experiments, we show the possibility to control the intensity and the phase of the amplitude mode of the system.

References
[1] Mitsuishi, N. et al. Nat. Commun. 11, 2466 (2020).
[2] Schaefer, H. et al. Phys. Rev. B 89, 045106 (2014).

O 19.5 Mon 18:00 P2/EG

Modification of the ultrafast charge carrier dynamics in a molecule/TMDC heterostructure — •Sebastian Hedwig[1], Matthias Rüb[1], Benito Arnoldi[1], Kai Rossnagel[3,4], Benjamin Stadtmüller[1,2], and Martin Aeschlimann[1] — [1]Department of Physics, University of Kaiserslautern and Research Center OPTIMAS, 67663 Kaiserslautern, Germany — [2]Institute of Physics, Johannes Gutenberg University Mainz, 55128 Mainz, Germany — [3]Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, 24118 Kiel, Germany — [4]Ruprecht Haensel Laboratory, Deutsches Elektronen-Synchrotron DESY, 22607 Hamburg, Germany

Tailoring the properties of layered van der Waals materials is one of the most intriguing challenges in the field of 2D materials. While for transition metal dichalcogenides (TMDCs) this is mainly achieved by the formation of heterostructures with other 2D materials, our approach is to functionalize the properties of TMDCs by the adsorption of molecular films. Here we present our recent fs-time-resolved optical-pump XUV-probe ARPES experiments on a $C_{60}/TiSe_2$ heterostructure. $TiSe_2$ was selected since it can host a charge density wave phase at low sample temperature that can be melted by fs pulses [1]. We find a clear modification of the optically induced phase transition dynamics from the CDW into the semi-metallic phase. This observation is discussed in the light of interactions and the interfacial charge carrier dynamics at the $C_{60}/TiSe_2$ interface.

[1] Rohwer T., et al.; Nature, 471, 490, (2011)

O 19.6 Mon 18:00 P2/EG

Analysis of Nonlocal Correlations in 1*T*-TaS_2 Driven Out of Equilibrium — •Jesumony Jayabalan[1], Florian K. Diekmann[2], Ping Zhou[1], Kai Rossnagel[2], and Uwe Bovensiepen[1] — [1]Fakultät für Physik, Universität Duisburg-Essen, Germany — [2]Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, Germany

Driving correlated materials out of equilibrium on ultrafast time scales can provide information about the correlation phenomena and their complex free energy landscapes. Time- and angle-resolved photoemission spectroscopy provided access to the ultrafast dynamics of Doublons [Ligges et al., PRL 120, 166401 (2018)] and concluded on presence of empty and doubly occupied sites at few tens of femtosecond timescales which match calculated spectra with 5% static hole doping. Thus the doublon decay was concluded to be mediated in 1*T*-TaS_2 by the presence of holes. We propose experiments aimed at a doping dependent relaxation dynamics measurements that would reveal the

role of screening and charge excitations on such photo-induced states. Two different approaches will be used for doping: 1) 1 T-TaS_2 crystals are grown by Ta substitution to induce a less than half filled band and hole doping and 2) surface doping by adsorption of alkali adsorbates on the cleaved 1 T-TaS_2 surface. Such doping depending studies at the timescale of the charge density wave amplitude mode potentially allow to examine the role of strong correlations in the coupled electron-lattice dynamics of 1 T-TaS_2 as well as possibilities to control two-dimensional correlated surfaces via photo excitation of the substrate.

O 19.7 Mon 18:00 P2/EG

Disentangling HOMO and LUMO excitation in momentum space by means of time-resolved photoemission orbital tomography — Alexa Adamkiewicz[1], Miriam Raths[2], •Marcel Theilen[1], Monja Stettner[2], Sabine Wenzel[2], Mark Hutter[2], Sergey Soubatch[2], Christian Kumpf[2], Francois C. Bocquet[2], Robert Wallauer[1], F. Stefan Tautz[2], and Ulrich Höfer[1] — [1]Philipps-University Marburg, Germany — [2]Peter Grünberg Institute (PGI-3), Jülich Research Centre, Germany

Time-resolved photoemission orbital tomography allows to trace electron dynamics in momentum space by measuring the angle-resolved photoemission intensity on an ultrafast timescale. For molecule-metal interfaces it proved to be a powerful technique to investigate the population dynamics of molecular states [1]. Here, we find for a well-ordered monolayer of CuPc on Cu(001)-2O a delay-dependent change in the momentum pattern of the molecular LUMO. In addition, we show how the temporal evolution of the momentum distribution can be systematically disentangled from contributions of the projected HOMO by varying the pump photon energy and polarization. By aligning the polarization of the pump pulse along the molecular axis of CuPc molecules, we are able to selectively excite molecules of a specific orientation. The temporal evolution of the excitation and decay from the LUMO can be well described by solving the optical Bloch equations for a three-level system.

[1] R. Wallauer *et al.*, Science 371, 1056-1059 (2021).

O 19.8 Mon 18:00 P2/EG

Spin- and valley-dependent charge carrier dynamics in a WSe_2 bulk crystal — •Gregor Zinke[1], Sebastian Hedwig[1], Benito Arnoldi[1], Martin Aeschlimann[1], and Benjamin Stadtmüller[1,2] — [1]Department of Physics and Research Center OPTIMAS, TU Kaiserslautern, 67663 Kaiserslautern, Germany — [2]Institute of Physics, Johannes Gutenberg University Mainz, 55128 Mainz, Germany

The intriguing spin functionalities of transition metal dichalcogenides such as WSe_2 are rooted in the spin- and valley-degrees of freedom. These are responsible for the particular spin-texture of TMDCs as well as the helicity- and valley-dependent light absorption. However, despite the clear understanding of the initial spin-and valley-dependent light-matter interaction, a clear picture of the spin-dependent intra- and intervalley scattering of electrons and holes is still elusive. Here, we focus on the ultrafast carrier dynamics in a bulk crystal of WSe_2. Using spin-, time- and angle-resolved photoemission with XUV-radiation, we simultaneously image the temporal evolution of the excited carriers in the conduction band as well as the corresponding hole dynamics within the valence band on a fs timescale. The spin-polarization of the initially excited carriers is controlled by the helicity of the optical excitation. We will show clear changes of the spin-polarization of the excited electrons after intra-band scattering from the K- to the Σ-point, and discuss these results in regards to the influence of electron-electron and electron-phonon scattering for the spin-polarized electron population in the conduction band.

O 19.9 Mon 18:00 P2/EG

Ultrafast dynamics of CT excitons in 2d heterostructures investigated with SHG imaging microscopy — •Marleen Axt, Jonas E. Zimmermann, Gerson Mette, and Ulrich Höfer — Fachbereich Physik, Philipps-Universität Marburg, Germany

The exciton dynamics in vertically stacked TMDC heterostructures can be modified by variation of the twist angle between the two layers. Using time-resolved second hamornic (SH) imaging microscopy we observe the ultrafast formation of charge transfer (CT) excitons in micrometer-sized MoS_2/WSe_2 heterostructures with different stacking angles. We make use of the strong anisotropy of the nonlinear SH response to selectively probe the carrier dynamics in individual layers of the system. For misaligned layers with a stacking angle of 16° the formation of CT excitons takes place within 85 fs after resonant excitation of the WSe_2 A excition. In contrast, an alignment close to 2H stacking (52°) leads to an enhanced electronic coupling between the layers resulting in a much faster CT exciton formation time of only 12 fs. Moreover, different formation and relaxation pathways are identified depending on the pump-photon energy.