O 45: Ultrafast Electron Dynamics at Surfaces and Interfaces IV

Time: Wednesday 10:30–12:45 Location: MA 041

O 45.1 Wed 10:30 MA 041

Ultrafast Charge Transfer in Graphene/Ag/SiC Studied Using Time- and Angle-Resolved Photoemission Spectroscopy — •Eduard Moos[1], Zhi-Yuan Deng[1], Hauke Beyer[1], Kai Rossnagel[1,2,3], and Michael Bauer[1,2] — [1]Institute of Experimental and Applied Physics, Kiel University, 24098 Kiel, Germany — [2]Kiel Nano, Surface and Interface Science KiNSIS, Kiel University, 24118 Kiel, Germany — [3]Deutsches ElektronenSynchrotron DESY, Ruprecht Haensel Lab, 22607 Hamburg, Germany

Monoatomic metals intercalated between graphene/SiC interfaces represent a new type of van der Waals heterostructures with extraordinary properties. For instance, silver exhibits a metal-semiconductor transition in the monolayer limit (MLAg) [1, 2]. The low binding energy of the valence band maximum of MLAg, the intrinsic band gap of a bilayer graphene (BLGr) overlayer, and a twisted orientation of the Brillouin zone of silver and graphene overlayer make this heterostructure an interesting model system for the exploration of ultrafast interlayer charge transfer. In this contribution, we use time- and angle-resolved photoemission spectroscopy (TRARPES) with a time resolution of 35 fs to study such types of processes in n-doped BLGr/MLAg/6H-SiC. Following the decay of the photoexcited carriers, we observe clear signatures of a net charge transfer between Ag and graphene on a 50 fs timescale. The detailed analysis of the data allows disentangling and identifying the relevant pathways.

[1] P. Rosenzweig, U. Starke, Phys. Rev. B **101**, 201407(R) (2020).
[2] W. Lee *et al.*, Nano Lett. **22**, 19, 7841-7847 (2022).

O 45.2 Wed 10:45 MA 041

Influence of twist angle on ultrafast charge transfer in WS2-graphene heterostructures — •Niklas Hofmann[1], Leonard Weigl[1], Johannes Gradl[1], Neeraj Mishra[2,3], Stiven Forti[2], Camilla Coletti[2,3], Amir Klein[4], Daniel Hernangomez-Peres[4], Sivan Refaely-Abramson[4], and Isabella Gierz[1] — [1]University of Regensburg, Germany — [2]Istituto Italiano di Tecnologia, Pisa, Italy — [3]Istituto Italiano di Tecnologia, Genova, Italy — [4]Weizmann Institute of Science, Rehovot, Israel

Ultrafast charge separation is crucial for efficiently converting sunlight into electrical energy. This phenomenon commonly occurs in different van der Waals heterostructures [1] where the transfer rates for electrons and holes are determined by the band alignment and interlayer hybridization. Both parameters are expected to be highly sensitive with respect to variations of the twist angle between the layers. This makes the twist angle the decisive tuning parameter for optimizing ultrafast charge transfer processes for various applications. Using time- and angle-resolved photoemission spectroscopy, we investigate the non-equilibrium carrier dynamics in WS2-graphene heterostructures with twist angles of 0° and 30°. We find that, for a twist angle of 0°, hole transfer from WS2 to graphene is significantly faster than electron transfer [2]. For a twist angle of 30°, however, we find no indication for ultrafast charge separation. We interpret our results with the help of ab-initio band structure calculations.

[1] C. Jin et al., Nature Nanotechnology 13, 994 (2018)
[2] S. Aeschlimann et al., Sci. Adv. 6, eaay0761 (2020)

O 45.3 Wed 11:00 MA 041

Probing electron-hole Coulomb correlations in the exciton landscape of a twisted semiconductor heterostructure — •Jan Philipp Bange[1], David Schmitt[1], Wiebke Bennecke[1], Giuseppe Meneghini[2], AbdulAziz AlMutairi[3], Daniel Steil[1], Sabine Steil[1], R. Thomas Weitz[1], G. S. Matthijs Jansen[1], Stephan Hofmann[3], Samuel Brem[2], Ermin Malic[2], Marcel Reutzel[1], and Stefan Mathias[1] — [1]Georg-August-Universität Göttingen, I. Physikalisches Institut, Germany — [2]Philipps-Universität Marburg, Germany — [3]University of Cambridge, U.K.

An exciton is a two-particle correlated state between an electron and a hole. In the type-II band aligned WSe_2/MoS_2 heterostructure, interlayer excitons can be formed after the resonant excitation of the WSe_2 intralayer excitons. In this process, the exciton's electron transfers across the interface while the hole remains rigid in the MoS_2 layer [1]. Moreover, the inverted process is possible: After exciting MoS_2 intralayer excitons, charge transfer of the exciton's hole across the interface leads to the formation of interlayer excitons. In this case, the exciton's electron remains rigid in WSe_2.

Here, we employ time-resolved momentum microscopy to study the ultrafast hole-transfer mechanism. Interestingly, we find a distinct photoemission feature that can only be described when considering the break-up of the Coulomb correlation between the exciton's electron and hole during the photoemission process [2].

[1] Schmitt, Bange et al., Nature 608, 499-503 (2022).
[2] Bange et al., arXiv:2303.17886 (2023).

O 45.4 Wed 11:15 MA 041

Effect of dynamical screening on the exciton-phono coupling in a layered semiconductor — •Selene Mor[1,2], Valentina Gosetti[1,2,3], Alejandro Molina-Sanchez[4], Davide Sangalli[5], Simona Achilli[5], and Stefania Pagliara[1,2] — [1]Università Cattolica del Sacro Cuore, Brescia, Italy — [2]I-Lamp Research Center, Brescia, Italy — [3]KU Leuven, Leuven, Belgium — [4]University of Valencia, Valencia, Spain — [5]University of Milan, Milan, Italy

The light-mediated interaction of fermionic and bosonic excitations governs the optoelectronic properties of condensed matter systems. In photoexcited semiconductors, the coupling of electron-hole pairs (excitons) to coherent optical phonons enables a modulation of the excitonic resonance on the picosecond timescale [1]. At the same time, due to the Coulombic nature of excitons, their dynamics are sensitive to transient changes in the screening by the photoexcited carriers. Interestingly, the effect of interplay between exciton-phonon coupling and dynamical screening is still elusive. By means of broadband transient reflectance spectroscopy, we set the spectral evidences of either the breaking or the activation of exciton-phonon coupling in a layered semiconductor, and we reveal that dynamical screening can favor the phonon-mediated modulation of the excitonic resonance. These findings move a step forward on the path towards the optical control of fundamental interactions on the ultrafast timescale. [1]Mor, S. et al., Phys. Rev. Research 3, 043175 (2021)

O 45.5 Wed 11:30 MA 041

Ultrafast nano-imaging of dark excitons — •David Schmitt[1], Jan Philipp Bange[1], Wiebke Bennecke[1], Giuseppe Meneghini[2], AbdulAziz AlMutairi[3], Marco Merboldt[1], Jonas Pöhls[1], Sabine Steil[1], Daniel Steil[1], R. Thomas Weitz[1], Stephan Hofmann[3], Samuel Brem[2], G. S. Matthijs Jansen[1], Ermin Malic[2], Stefan Mathias[1], and Marcel Reutzel[1] — [1]Georg-August-Universität Göttingen, I. Physikalisches Institut, Germany — [2]Fachbereich Physik, Philipps-Universität, Marburg, Germany — [3]Department of Engineering, University of Cambridge, Cambridge CB3 0FA, U.K.

The optical response of transition-metal dichalchodgenide (TMDs) heterostructures is determined by the formation of tightly-bound electron-hole pairs, or so-called excitons. For the application of TMDs in optoelectronic devices it is necessary to understand not only the temporal evolution of the formation, thermalization and relaxation of excitons within energy- and momentum-space [1], but also the impact of spatial inhomogeneities to the exciton landscape and ultrafast dynamics on the relevant nanometer to micrometer spatial scale. In this talk, we will demonstrate how our novel method of ultrafast dark-field momentum microscopy [2] can be used to track the ultrafast formation dynamics of excitons with unparalleled simultaneous spatio-temporal and spatio-spectral information.

[1] Schmitt *et al.*, Nature 608, 499-503 (2022)
[2] Schmitt *et al.*, arXiv:2305.18908 (2023)

O 45.6 Wed 11:45 MA 041

Influence of defects on ultrafast charge separation in van der Waals heterostructures — •Johannes Gradl[1], Niklas Hofmann[1], Leonard Weigl[1], Raül Perea-Causín[2], Ermin Malic[3], Daniel Hernangómez Pérez[4], Sivan Refaely-Abramson[4], and Isabella Gierz[1] — [1]University of Regensburg — [2]Chalmers University of Technology — [3]University of Marburg — [4]Weizmann Institute of Science

Ultrafast charge separation is crucial for efficiently converting sunlight into electrical energy. This phenomenon commonly occurs in different van der Waals heterostructures [1] where the transfer rates for elec-

trons and holes are determined by the band alignment and interlayer hybridization. Recently, it has been realized that defects might enhance the lifetime of the charge separated state significantly [2,3]. A detailed microscopic understanding of defect-assisted charge transfer, however, is lacking. We deliberately generate sulfur vacancies in a prototypical WS2-graphene heterostructure and probe their influence on the charge transfer dynamics with time- and angle-resolved photoemission spectroscopy. We interpret our results based on existing theory [4,5].

[1] Nat. Nanotechnol. 13, 994 (2018)
[2] Sci. Adv. 7, eabd9061 (2021)
[3] Phys. Rev. Lett. 127, 276401 (2021)
[4] Phys. Rev. B 107, 075419 (2023)
[5] Nano Lett. 23, 5995 (2023)

O 45.7 Wed 12:00 MA 041

Ultrafast SHG imaging microscopy: imhomogeneities in space and time — •Marleen Axt[1], Markus B. Raschke[2], Gerson Mette[1], and Ulrich Höfer[1] — [1]Fachbereich Physik, Philipps-Universität Marburg, Germany — [2]Department of Physics and JILA, University of Colorado, Boulder, USA

Ultrafast processes in 2D materials such as exciton or charge-transfer dynamics exhibit a broad range of time scales. They have been found to vary from sample to sample, are substrate dependent, or influenced by edges, grain boundaries and defects. Distinguishing the influence of these extrinsic effects from the intrinsic carrier dynamics has been challenging. In particular for 2D heterostructures, the emergent quantum phenomena are controlled by interlayer coupling and crystallographic orientation that are particularly sensitive to structural heterogeneities. Here, we present multi-scale imaging in a combination of time-resolved second-harmonic generation microscopy with nano-optical imaging to probe WS_2/WSe_2 heterostructures that reveal a large influence of different inhomogeneities on carrier dynamics.

O 45.8 Wed 12:15 MA 041

Coherent Time- and Angle-Resolved Photoelectron Spectroscopy in a Low Energy Electron Microscope — •Alexander Neuhaus[1], Pascal Dreher[1], Florian Schütz[2], Helder Marchetto[2], Torsten Franz[2], Michael Horn-von Hoegen[1], and Frank J. Meyer zu Heringdorf[1,3] — [1]Faculty of Physics and Center for Nanointegration, Duisburg-Essen (CENIDE), University of Duisburg-Essen, 47048 Duisburg, Germany — [2]ELMITEC Elektronenmikroskopie GmbH, 38678 Clausthal-Zellerfeld, Germany — [3]Interdisciplinary Center for the Analytics on the Nanoscale (ICAN), 47057 Duisburg, Germany

In modern photoemission microscopes the electron optics allows different imaging modes such as real space imaging or momentum microscopy. Here, we describe how a spectroscopic and low energy electron microscope (SPE-LEEM) can be equipped with an additional slit at the entrance of the hemispherical analyzer to enable a ARPES like mode with micrometer spatial selectivity while maintaining all other imaging modes. We use a photogrammetric calibration to correct for image distortions caused by the magnetic projective system and to calibrate the momentum and energy axis. We demonstrate the capabilities of the new ARPES mode by analyzing the time-resolved nonlinear electron emission from a plasmonic nano-focus on a Au(111) platelet using a femtosecond laser.

O 45.9 Wed 12:30 MA 041

Modelling the temporal evolution of photoemission momentum maps of CuPc/Cu(001)-2O — •Alexa Adamkiewicz[1], Miriam Raths[2], Marcel Theilen[1], Lasse Münster[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

Using time-resolved photoemission orbital tomography on a well-ordered monolayer of CuPc on Cu(001)-2O, we find a temporal evolution of the momentum pattern which gradually changes from a HOMO into a LUMO pattern [1]. This can be traced back to the presence of two excitation pathways with different dynamics: transient population of the CuPc LUMO via resonant HOMO→LUMO transition and coherent two-photon photoemission from the HOMO. For a theoretical description of the ultrafast electron dynamics, we numerically solve the optical Bloch equations for a three-level system, using a density matrix approach. This allows us to disentangle photoemission contributions from both excitation pathways. The model well reproduces the respective momentum distribution patterns as well as the measured shift in kinetic energy of photoelectrons for different detuning of the pump photon energy with respect to resonant excitation.

[1] A. Adamkiewicz *et al.*, J. Phys. Chem. C **127**, 20411 (2023).