O 16: Poster Monday: Ultrafast Processes 1

Time: Monday 18:00–20:00 Location: P4

O 16.1 Mon 18:00 P4

Probing photo-induced, ultra-fast dynamics by time-resolved spectroscopic ellipsometry — •Felix-Florian Delatowski[1,3], Krishna Khakurel[1], Jörg Rappich[2], Shirly Espinoza[1], Mateusz Rebarz[1], Martin Zahradnik[1], and Jakob Andreasson[1] — [1]ELI Beamlines, Institute of Physics, Czech Academy of Sciences, Czech Republic — [2]Institute Silicon Photovoltaics, Helmholtz-Zentrum Berlin für Materialien und Energie, Germany — [3]Semiconductor Physics Group, Felix Bloch Institute for Solid State Physics, Universität Leipzig, Germany

Photo-induced phenomena, such as charge carrier and structural dynamics, charge transfer and relaxation processes on sub-picosecond time scales are of great interest for a profound understanding of light-matter interaction. Time-resolved spectroscopic ellipsometry is a promising surface sensitive tool to investigate these phenomena. In this work, we explore the potential of this measurement technique exemplary on germanium and gold and present preliminary experimental results.

In addition to the experimental observations, we also present an approach to predict ellipsometry spectra through ab-initio DFT calculations using the program SIESTA. In order to extend this approach, some preliminary measurements of organic molecules (Pyrimidine) have been performed. We discuss the benefits that the synergy of ab-initio based spectrum predictions and experiments bring in the interpretation of the dynamics in the sample.

O 16.2 Mon 18:00 P4

Tailoring the carrier dynamics of WSe_2 by the adsorption of CuPc — •Gregor Zinke[1], Sebastian Hedwig[1], Benito Arnoldi[1], Martin Aeschlimann[1], and Benjamin Stadtmueller[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

Tailoring the electronic properties and carrier dynamics of 2D-Van-der-Waals materials is a highly promising way to design spin functionalities in low dimensions. This is often achieved by the formation of heterostructures with other Van-der-Waals materials.

Here, we functionalize the carrier dynamics of the prototypical Van-der-Waals material WSe_2 by the adsorption of the aromatic molecule CuPc. Using time- and angle-resolved photoemission with XUV-radiation, we investigate the temporal evolution of the excited states at the K- and Σ-points of WSe_2 after an optical excitation. For the bare WSe_2, we find a spin selective excitation at the K-point depending on the pump light polarization [1], which is followed by a spin-flip scattering from the K- to the Σ-point. After the adsorption of CuPc, the excitation scheme is completely altered. In particular, we uncovered a direct interlayer excitation from the CuPc into the WSe_2 layer that dominates the carrier dynamics of the CuPc/WSe_2 heterostructure.

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

O 16.3 Mon 18:00 P4

Numerous Improvements on Ultrafast Pump-Probe RHEED Experiment — •Jonas Fortmann[1], Christian Brand[1], Thorben Groven[1], Mohammad Tajik[1], Michael Horn-von Hoegen[1], and Thomas Duden[2] — [1]Department of Physics and Center for Nanointegration CENIDE, University of Duisburg-Essen, Lotharstraße 1, 47057 Duisburg — [2]Construction office, Mustangweg 17, D-33649, Bielefeld

During the last year, major improvements have been made to our ultrafast time-resolved reflection high energy electron diffraction (tr-RHEED) experiment. In particular, the previous detection unit consisting of a multichannel plate and a cooled CCD camera has been replaced by a single electron sensitive CMOS based camera (TVIPS TemCam XF-416), which allows spot profile analysis with its superior resolution and signal-to-noise ratio and no blooming. Flatfielding of the detector is achieved with a home-built electron source at 20 keV accompanied by an external EM-deflection unit. In addition, the 80 fs-laser pump pulse ($\lambda = 800$ nm) has been upgraded with an optical parametric amplifier (Topas Prime) that allows excitation of the sample's surface at $1.16 - 2.60\,\mu$m in future experiments. Re-routing of the beamline has shortened the beam paths by ~ 1.5 m and saved half of the mirrors, which provides higher beam stability. Also the third harmonic generation (THG) stage for electron generation in the Au film photocathode has been optimized. A new lab software has been established on the basis of TANGO controls. First experimental data with the new setup are shown.

O 16.4 Mon 18:00 P4

Ultrafast optical spectroscopy of few-layer TMDCs under ultrahigh-vacuum conditions — •Maximilian Franz, Kilian Kuhlbrodt, Jan Gerrit Horstmann, and Claus Ropers — Max-Plank-Institut für Multidisziplinäre Naturwissenschaften, Göttingen, Germany

Optical pump-probe spectroscopy (OPP) has enabled detailed investigations of the nonequilibrium optical properties of atomically thin materials, with prominent examples in graphene or transition metal dichalcogenide (TMDCs) heterostructures [1]. However, these materials are often susceptible to oxidation and contamination under atmospheric conditions, resulting in strongly altered physical and chemical properties [2]. Here, we present the development of an OPP setup for the investigation of dynamics in few-layer TMDCs and heterostructures under ultrahigh vacuum (UHV) conditions. We present first results on the ultrafast phase transition in 1T-TaS_2 samples using time-resolved reflectance and transmittance measurements with fs temporal and μm spatial resolutions. Furthermore, we discuss possible applications, e.g., layer-selective optical excitation of TMDC heterostructures, and the combination of OPP with structure-sensitive techniques like ultrafast low-energy electron diffraction [3].

[1] C. Jin et al., Nature Nanotechnology 13, 994-1003 (2018); [2] Y. Yu et al., Nature Nanotechnology 10, 270-276 (2015); [3] S. Vogelgesang et al., Nature Physics 14, 184-190 (2018)

O 16.5 Mon 18:00 P4

Description for the electronic non-equilibrium after ultrashort laser excitation of metals — •Markus Uehlein, Sebastian T. Weber, and Baerbel Rethfeld — Department of Physics and Research Center OPTIMAS, TU Kaiserslautern, Germany

When a metal is excited by a femtosecond laser pulse in the visible range, electrons absorb the energy, resulting in a non-equilibrium energy distribution. This absorption behavior and the following thermalization to a hot Fermi distribution can be simulated using complex and numerically expensive Boltzmann collision integrals [1]. After the thermalization, the two-temperature model (TTM) can describe the relaxation of the heated electrons and the phonons in a simple way. It is, however, unable to trace non-thermal electrons.

We present an intermediate model, called extended TTM (eTTM). It was developed in Refs. [2, 3] and adds a system of non-equilibrium electrons to the TTM. Besides some improvements to the published versions, we compare the eTTM to the Boltzmann model as well as to the TTM [4]. In particular, we compare the spectral particle dynamics to a time-resolved two-photon photoemission measurement [5].

[1] B. Y. Mueller and B. Rethfeld; PRB **87**, 035139 (2013)
[2] E. Carpene; PRB **74**, 024301 (2006)
[3] G. D. Tsibidis; Appl. Phys. A **124**, 311 (2018)
[4] M. Uehlein, S. T. Weber and B. Rethfeld; Nanomaterials **12**, 1655 (2022)
[5] Y. Beyazit *et al.*; Phys. Rev. Lett. **125**, 076803 (2020)

O 16.6 Mon 18:00 P4

A setup for interferometrically time-resolved multi-photon photoemission benchmarked on Ag(111) — •Hannah Strauch, Marco Merboldt, Jan Philipp Bange, Daniel Steil, G. S. Matthijs Jansen, Sabine Steil, Marcel Reutzel, and Stefan Mathias — I. Physikalisches Institut, Georg-August-Universität Göttingen, Göttingen, Germany

Time- and angle-resolved photoemission is the method of choice to directly study the electron dynamics of condensed matter systems with energy and momentum resolution. After the system is excited by a pump pulse, a delayed second pulse induces photoemission and thus probes the materials response. To complement the obtained insight with information on the coherence of the response, single-color phase-locked pulse pairs can be employed. Here, we present a setup for interferometric time-resolved multi-photon photoemission based on a passively stabilized Mach-Zehnder interferometer which produces phase-

locked pulse pairs and provides an optical delay precision below 60 as. We show interferometric time- and angle-resolved multi-photon photoemission data from the Shockley surface state and the first image potential state of the Ag(111) surface as a benchmark for our setup. Supported by an optical Bloch equation model we explore possibilities to interpret such a data set.

A promising application of the interferometer is in combination a high harmonic generation EUV beamline. This enables double-pump-probe experiments with access to the full Brillouin zone, and allows a comprehensive investigation of optical excitations.

O 16.7 Mon 18:00 P4

Time-resolved ARPES probing *Rabi* Oscillations and *Landau-Zener-Stückelberg* Interferences in Graphene - a Proposal — •Eduard Moos, Hauke Beyer, and Michael Bauer — Institute of Experimental and Applied Physics, Kiel University, Germany

At sufficiently high intensities, the interaction of few-cycle laser fields with solids gives rise to strong-field effects envisioning novel and exciting strategies for controlling optical and electronic properties via the electric field waveform on sub-femtosecond timescales. A striking example is the CEP-control of light-field-driven currents in graphene due to *Landau-Zener-Stückelberg* (LZS) interferences [1] resulting from the complex interplay of field-driven adiabatic intraband and diabatic interband transitions. Simulations show that the LZS-interferences give rise to characteristic asymmetries in the momentum-distribution of the residual conduction band population in the Dirac cone on top of a symmetric quasi-periodic pattern indicative for *Rabi* oscillations. TRARPES using few cycle femtosecond pump laser pulses seems to be in ideal tool for the investigation of these processes and there dependence on parameters such as pulse peak electric field strength E0, laser polarization, and CEP phase. Based on preliminary results on graphite using 7 fs few-cycle NIR pump-pulses we will discuss in this presentation the prospects, but also the challenges that arise in such type of TRARPES experiment.

[1] T. Higuchi, *et al.*, Nature **550**, 224 (2017)
[2] G. Rohde, *et al.*, Phys. Rev. Lett. **121**, 256401 (2018)

O 16.8 Mon 18:00 P4

Laser-based low energy photoelectron diffraction of SnPc/graphite — •Hermann Erk, Stephan Jauernik, Petra Hein, and Michael Bauer — IEAP, CAU Kiel, Germany

Tin-phthalocyanine (SnPc) adsorbed on graphite has been studied using laser-based angle resolved photoemission spectroscopy (ARPES) at 5.9 eV photon energy and low energy electron diffraction (LEED). An ordered SnPc monolayer was prepared by thermal evaporation onto single crystalline graphite (SCG) flakes and subsequent annealing at 370 K. LEED data reveal an incommensurate ordered phase of the SnPc overlayer at room temperature. In the ARPES spectra the SnPc long-range order can be seen due to an adsorbate-induced backfolding of the band structure of graphite from the $\overline{\mathrm{K}}$ point to the center of the Brillouin zone. A comparison of ARPES spectra with simulations of the backfolded band structure under consideration of the LEED data will be discussed. First time-resolved ARPES measurements with the aim to address ultrafast changes in the SnPc long range order will be presented.

O 16.9 Mon 18:00 P4

Type II aligned TMD heterostructures: Probing moiré interlayer excitons with energy-, momentum-, and femtosecond time-resolution — •Marcel Reutzel[1], David Schmitt[1], Jan Philipp Bange[1], Wiebke Bennecke[1], AbdulAziz AlMutairi[2], Giuseppe 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], and Stefan Mathias[1] — [1]I. Physikalisches Institut, Georg-August-Universität Göttingen, Göttingen, Germany — [2]Department of Engineering, University of Cambridge, Cambridge CB3 0FA, U.K. — [3]Fachbereich Physik, Philipps-Universität, 35032 Marburg, Germany

Transition metal dichalcogenides (TMDs) can be stacked into atomically thin p-n junctions, where an optically excited intralayer exciton can decay into interlayer excitons. Here, the electron and the hole contribution to the quasiparticle reside in the neighbouring TMD layers.

On this poster, we show our recent progress towards the identification and characterization of the ultrafast charge transfer process across a type II interface. Using femtosecond momentum microscopy, we identify the distinct momentum fingerprints of various excitonic states in this system, notably the bright and dark intralayer excitons as well as the interlayer exciton. Most intriguingly, the momentum-resolved measurement provides quantitative access to the interlayer exciton wavefunction that is modulated within the moiré potential.
Schmitt *et al.*, *arXiv:2112.05011 (2021).

O 16.10 Mon 18:00 P4

Influence of the static dielectric permittivity on ultrafast quasiparticles dynamics in WS_2 monolayers — •Subhadra Mohapatra[1,2], Stefano Calati[1,2], Quiyang Li[3], Xiaoyang Zhu[3], and Julia Stähler[1,2] — [1]Humboldt-Universität zu Berlin, Institut für Chemie,Berlin,Germany — [2]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Abt. Physikalische Chemie, Berlin, Germany — [3]Columbia University, New York City, New York

In our recent fluence-and photon energy-dependent studies [1,2] of quasiparticle dynamics in WS_2 monolayers on fused silica (FS) and Si-SiO_2 substrates, we observed that excitonic screening solely reduces the binding energy of the excitons, leading to a transient blue-shift of the exciton resonance while quasi free carrier screening effectively shows a red shift, as the carrier-induced screening leads to a larger band gap renormalization than binding energy reduction. Further investigation of such fluence-dependent quasiparticle dynamics studies using a higher dielectric permittivity of a sapphire substrate, we found that scattering rates, relaxation time constants, and band gap renormalization are not influenced by the dielectric permittivity. On the contrary, the ratio of dynamic screening parameter of the excitons and their Bohr radius is approximately 4 times higher in sapphire than for FS, which must be a direct consequence of the increased dielectric permittivity likely leading to more localized excitons.

References:
[1] Calati et al. PCCP 23(39) (2021).
[2] Calati et al. arXiv:2204.02125 (2022).

O 16.11 Mon 18:00 P4

Ultrafast Transport and Energy Relaxation of Hot Electrons in Au/Fe/MgO(001) Investigated by Linear Time-resolved Photoelectron Spectroscopy — •Florian Kühne[1], Yasin Beyazit[1], Detlef Diesing[2], Ping Zhou[1], Jesumony Jayabalan[1], and Uwe Bovensiepen[1] — [1]University of Duisburg-Essen,Physics — [2]University of Duisburg-Essen, Chemistry

Optically excited electrons and holes are of particular interest in solid-state physics because analysis of their dynamics allows a microscopic understanding of the interactions in non-equilibrium states. Here we want to discern the relaxation by such local inelastic processes and non-local transport effects. To analyze the ultrafast dynamics of charge carriers in the vicinity of the Fermi energy E_F, femtosecond time-resolved linear photoelectron spectroscopy was applied. We report on first experimental results obtained by using 1.55 eV pump and 6 eV probe photons on an Au/Fe/MgO(001) epitaxial heterosystem, complementary to previous work in Beyazit et al., PRL 125, 076803 (2020). By pumping the Fe side, hot electrons are excited in the Fe layer, and subsequently injected into the Au layer and propagate to the surface, where they are probed by photoelectron emission spectroscopy. In the Fe side pumped data, we observe thickness dependent differences in relaxation compared to the Au side pumped data. In Au side pumping we observe efficient transport into the Fe layer. We will present an energy E dependent analysis of the propagation and energy density $U(E, d_{Au})$ of electrons above E_F, which together with a two-temperature model shows a super diffusive transport limit.

O 16.12 Mon 18:00 P4

Investigations of polarons in hematite α-Fe_2O_3(1-102) by means of nc-AFM and KMC — •Jesús Redondo[1,2], Vít Gabriel[1], Giada Franceschi[3], Igor Sokolović[3], Dominik Wrana[1], Florian Kraushofer[3], Erik Rheinfrank[3], Michele Riva[3], Gareth S. Parkinson[3], Michael Schmid[3], Ulrike Diebold[3], Pavel Kocán[1], and Martin Setvin[1,3] — [1]Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic — [2]Faculty of Chemistry, University of the Basque Country, San Sebastián, Spain — [3]Institute of Applied Physics, Vienna University of Technology, Vienna, Austria

Polarons are known to strongly influence the catalytic activity and the electronic, magnetic, and structural properties of transition metal oxides and halide perovskites. The study of polaron formation and dynamics is fundamental to understanding the actual mechanisms and yields of catalytic reactions in these materials. A new method for the investigation of electron and hole polarons is demonstrated. Charge

carriers are injected with the AFM/STM tip into the surface of natural, Ti- and Ni-doped α-Fe2O3(1-102). The injected charges form a cloud of electrons or holes trapped in the lattice. This cloud expands due to electrostatic interactions and thermally activated polaron hopping. Controlled annealing of the sample and characterization by Kelvin probe force microscopy (KPFM) provides information on polaron dynamics; these results are compared to kinetic Monte Carlo (KMC) simulations and the dependence of the polaron hopping activation energy on the doping is shown.