O 8: Ultrafast Electron Dynamics I: Surfaces and Interfaces (joint session O/MA)

Time: Monday 10:30–13:15 Location: TRE Phy

O 8.1 Mon 10:30 TRE Phy

Electron dynamics of surface states in momentum space on Cu and Au — •Lukas Hellbrück, Tobias Eul, Martin Aeschlimann, and Benjamin Stadtmüller — Department of Physics and Research Center Optimas, University of Kaiserslautern, Germany

For the development of nanoscale electronic and spintronic devices, it is critical to understand electronic dynamics in solid states systems and at interfaces. Of particular interest is the correlation between the energy and momentum dissipation mechanisms of the hot carriers and the band structure of the material.

In this work, we employ time-resolved two photon momentum microscopy [1] with monochromatic radiation of about 4.6eV to access the electron dynamics in the excited state energy range of typical noble metal surfaces. From these data, we can extract momentum dependent quasi-particle lifetimes of the electrons, providing insight into momentum dependent scattering processes in these materials.

We focus on the (111) and (110) low-index copper and gold surfaces. These surfaces yield a variety of different states in the unoccupied band structure ranging from resonant and off-resonant bulk transitions to Shockley surface states and image potential states. For these different types of states, we observe lifetime differences of several femtoseconds, which points to a complex momentum dependent lifetime of the electrons, even for simple single crystalline materials.

[1] F. Haag et al., Rev. Sci. Instr. 90, 103104 (2019)

O 8.2 Mon 10:45 TRE Phy

Above-threshold multiphoton photoemission from noble metal surfaces — •Marcel Reutzel[1,2], Andi Li[2], and Hrvoje Petek[2] — [1]I. Physikalisches Institut, Georg-August-Universität Göttingen, Germany — [2]Department of Physics and Astronomy, University of Pittsburgh, Pittsburgh, USA

Exciting solid surfaces with intense femtosecond laser pulses prompts electrons of the interrogated material to respond in highly non-linear manner, as is evident in the emission of high-order harmonic radiation and photoelectrons with kinetic energies well above that of the driving photons (above threshold photoemission, ATP). In the solid state, the optical field interacts with the electronic eigenstates defined by the Coulomb potential of a periodic crystal, as well as with its many-body screening response.

In this contribution, we probe the nonlinear photoelectric responses of the pristine silver surfaces in (111) and (110) crystal orientation using interferometrically time-resolved multi-photon photoemission spectroscopy. First, we study the coherent electron dynamics leading to non-resonant coherent mPP of the occupied Shockley surface band of Ag(111). We characterize and benchmark the two-dimensional Fourier transform (2D-FT) spectra based on the photoemission order m and the coherences induced in the sample (m = 2-6) [Phys. Rev. X 9, 01104545 (2019)]. Second, we show that ATP can be used to probe deep points in the Brillouin zone, namely the surface state of Ag(110) at the $\bar{Y}$-point, that is otherwise hidden below the photoemission horizon in lowest order of mPP.

O 8.3 Mon 11:00 TRE Phy

Interfacial carrier and phonon dynamics in nanostructured metal/2D-semiconductor plasmonic heterostructures — •Tommaso Pincelli[1], Thomas Vasileiadis[1], Shuo Dong[1], Samuel Beaulieu[1], Maciej Dendzik[1], Daniela Zahn[1], Sang-Eun Lee[1], Hélène Seiler[1], Yinpeng Qi[1], Patrick Xian[1], Julian Maklar[1], Emerson Coy[2], Niclas Müller[3], Yu Okamura[3], Stephanie Reich[3], Martin Wolf[1], Laurenz Rettig[1], and Ralph Ernstorfer[1] — [1]Fritz-Haber-Institut der MPG, Berlin, Germany — [2]NanoBioMedical Centre, Adam Mickiewicz University, Poznan, Poland — [3]Freie Universität Berlin, Berlin, Germany

Noble metal nanostructures enhance light absorption in semiconductors to produce plasmons, whose decay couples strongly with the generation of hot carriers and non-equilibrium phonons. We combine the use of time and angle-resolved photoemission spectroscopy and femtosecond electron diffraction to investigate both charge carrier and phonon dynamics in a plasmonic metal/semiconductor heterostructure.

We grow epitaxial Au nanoislands on single-crystalline bulk WSe_2. The presence of Au nanostructures causes a shortening of the Σ-exciton lifetime and an accelerated lattice heating of WSe_2, which indicate interfacial charge transfer. Furthermore, we observe a non-equilibrium phonon distribution in WSe_2 with sub-bandgap optical pumping, demonstrating increased sensitivity of the semiconductor to near infrared frequencies. The corresponding energy transfer scales nonlinearly with the incident laser fluence, owing to plasmonically-enhanced phonon excitation.

O 8.4 Mon 11:15 TRE Phy

Ultrafast hole-transfer in $MoSe_2$/WSe_2 revealed by polarisation dependent second-harmonic imaging microscopy — •Jonas Zimmermann, Ulrich Höfer, and Gerson Mette — Fachbereich Physik, Philipps-Universität Marburg, Germany

Charge transfer across heterointerfaces plays a fundamental role for the functionality of electronic devices. 2D heterostructures based on transition metal dichalcogenides (TMDC) with their wide variety of materials and stacking combinations represent a unique opportunity for systematic studies of the interfacial charge-carrier dynamics.

Here, we present results on a $MoSe_2$/WSe_2 heterostructure investigated by time-resolved second-harmonic (SH) imaging microscopy. The transient SH-response after resonant optical excitation reveals a delayed filling and an enhanced lifetime for the heterostructure which is absent for the individual monolayers, indicating the formation of interlayer excitons. By careful selection of the polarisation of the probe laser, we are able to enhance the sensitivity to the individual layers in the heterostructure and with that to the charge transfer. Systematic measurements in dependence of the pump photon energy and the probe polarisation exhibit temporal signatures which are attributed to hole transfer from WSe_2 to $MoSe_2$ and vice versa with transfer times of a few hundred femtoseconds in both cases.

O 8.5 Mon 11:30 TRE Phy

Electron transfer dynamics in MoS_2 imaged by time-resolved momentum microscopy — •Lasse Münster[1], Sarah Zajusch[1], Katsumi Tanimura[2], Jens Güdde[1], Robert Wallauer[1], and Ulrich Höfer[1] — [1]Fachbereich Physik, Philipps-Universität Marburg, Germany — [2]Institute of Scientific and Industrial Research, Osaka University, Japan

We investigate the electron dynamics in the topmost layer of MoS_2 after optical excitation above the A-exciton resonance by means of time- and angle-resolved two-photon photoemission with a high harmonic probe. High harmonic generation in krypton is used to produce an almost isolated 7^{th} harmonic of the 400 nm driving laser pulses at a repetition rate of 200 kHz. This is combined with tunable pump pulses in the visible range for resonant excitation. The photoemitted electrons are detected by a momentum microscope with time of flight detection that covers the full photoemission horizon in a single measurement. With a high harmonic probe this results in the imaging of the entire first Brillouin zone in TMDCs.

We observe an instantaneous occupation of the conduction band after optical excitation at all $\overline{K}$-points followed by an ultrafast transfer to the conduction band minima at $\overline{\Sigma}$. For longer delays the excited electron distribution localizes at these high symmetry points which we attribute to electron cooling. The real-space imaging capability of the momentum microscope allows the restriction of such experiments to micrometer-size regions which opens up the possibility to observe momentum-resolved charge transfer in TMDC heterostructures.

O 8.6 Mon 11:45 TRE Phy

Ultrafast electron dynamics in black phosphorus studied by time-resolved photoemission momentum microscopy — •Maciej Dendzik[1,2], Shuo Dong[1], Tommaso Pincelli[1], Samuel Beaulieu[1], Patrick Xian[1], Helene Seiler[1], 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]KTH Royal Institute of Technology, Electrum 229, 164 40 Kista, Sweden

Black phosphorus (BP) has recently emerged as a promising material due to its highly anisotropic thermal, electronic and optical properties. The high hole mobility together with a band-gap tunable by thickness in the visible to mid-IR range make BP attractive for future applications in opto-electronics. Therefore, detailed information on both the equilibrium and excited-state electronic band structure of BP is

of strong interest. Here we present a fs time-resolved study of transient electronic structure of BP in the entire surface Brillouin zone (SBZ) directly imaged with a momentum microscope using a novel setup for XUV time- and angle-resolved photoemission spectroscopy. The measured dispersion is found to be in good agreement with density functional theory calculations. We find that an optical excitation at 800 nm creates a hot-carrier distribution around the SBZ center, which scatters to two other valleys in the conduction band within ca. 15 fs. We further observe a strong linear dichroism in the optical absorption of BP. Interestingly, the inter-valley scattering dynamics are also found to depend on the pump polarization vector direction.

O 8.7 Mon 12:00 TRE Phy

Carrier dynamics in a laser-excited Fe/(MgO)(001) heterostructure from real-time TDDFT — •Elaheh Shomali, Markus Ernst Gruner, and Rossitza Pentcheva — Department of Physics and Center for Nanointegration, CENIDE, University of Duisburg-Essen, Germany

The interaction of a femtosecond optical pulse with a metal/oxide interface has been addressed based on time-dependent density functional theory (TDDFT) in the real-time domain using the Elk code. We systematically studied electronic excitations of a $Fe_1/(MgO)_3(001)$ heterostructure as a function of laser frequency, peak power density and polarization direction. We find a marked anisotropy in the response to in-plane and out-of-plane polarized light, which changes its character for frequencies lower and higher than the MgO band gap. For laser frequencies between the MgO band gap and the charge transfer gap, interface states resulting from the hybridization of the $d_{3z^2-r^2}$ orbitals of Fe and the p_z orbitals of O at the interface may foster the propagation of excitations into the central layer of MgO. Spin-orbit coupling (SOC) is found to result in a small time-dependent reduction of magnetization only. Finally, we extend our investigation to thicker heterostructures, such as $Fe_3/(MgO)_5(001)$.

Financial support from the DFG within SFB 1242 (project C02) is gratefully acknowledged.

O 8.8 Mon 12:15 TRE Phy

Local and non-local electron dynamics of Au/Fe/MgO(001) heterostructures analyzed by time-resolved two-photon photoemission spectroscopy — Yasin Beyazit[1], Jan Beckord[1], Ping Zhou[1], Jan Philipp Meyburg[2], Detlef Diesing[2], Manuel Ligges[1], and •Uwe Bovensiepen[1] — [1]University of Duisburg-Essen, Faculty of Physics and Center for Nanointegration (CENIDE), 47048 Duisburg) — [2]University of Duisburg Essen, Faculty of Chemistry, Universitätsstr. 5, 45711 Essen

The ultrafast electron dynamics at interfaces is determined by the competition of local scattering processes and transport effects. Employing femtosecond laser pulses in front and back side pumping of Au/Fe/MgO(001) [1] combined with detection in two-photon photoelectron emission spectroscopy we analyze local relaxation dynamics of excited electrons in buried Fe, injection into Au across the Fe-Au interface, and electron transport across the Au layer at 0.6 to 2.0 eV above the Fermi energy. By analysis as a function of Au film thickness we obtain the electron lifetimes of bulk Au and Fe and distinguish the relaxation in the heterostructure's constituents. We conclude further that electron injection across the epitaxial interface proceeds by electron wavepacket propagation. We also show that the excited electrons propagate through Au in a superdiffusive regime determined by few e-e scattering events. Application of such back side pumping photoelectron spectroscopy to further material systems will be discussed. Funding by the DFG through CRC 1242 is gratefully acknowledged.
[1] Alekhin et al., J. Phys. Condens. Matter 31, 124002 (2019)

O 8.9 Mon 12:30 TRE Phy

Time-resolved nonlinear optical spectroscopy of ultrafast charge transfer at the buried GaP/Si(001) interface — •Gerson Mette[1], Jonas Zimmermann[1], Alexander Lerch[1], Kristina Brixius[1], Jens Güdde[1], Andreas Beyer[1], Michael Dürr[2], Kerstin Volz[1], Wolfgang Stolz[1], and Ulrich Höfer[1] — [1]Fachbereich Physik, Philipps Universität Marburg, Germany — [2]Institut für Angewandte Physik, Justus-Liebig-Universität Giessen, Germany

The ongoing miniaturization increases the contribution of interface processes to electronic device properties. For a microscopic understanding, the dynamics of charge transfer across interfaces are particularly important. However, due to the experimental difficulty to detect and isolate the weak interface signature from the dominant bulk signals, direct experimental information about the ultrafast dynamics at buried interfaces is scarce.

Here, we will show that the experimental challenges can be overcome by optical second-harmonic generation (SHG), a technique which is intrinsically highly interface sensitive. We are investigating the ultrafast charge-carrier dynamics at the buried interface of GaP on Si(001) by time-resolved optical-pump SHG-probe spectroscopy. Photon energy dependent measurements reveal the existence of electronic interface states in the band gap of both materials. Charge carriers excited via these interface states are efficiently injected within a few hundred femtoseconds from the GaP/Si interface into the Si substrate resulting in the build-up of an electric field on a picosecond time scale.

O 8.10 Mon 12:45 TRE Phy

Pump-probe second harmonic spectroscopy of molecule/metal interfaces — Jinghao Chen, Ping Zhou, Uwe Bovensiepen, and •Andrea Eschenlohr — Faculty of Physics, University Duisburg-Essen, Lotharstr. 1, 47057 Duisburg, Germany

Achieving a microscopic understanding of charge transfer dynamics and the relaxation of optically excited electrons and holes at molecule/metal interfaces requires an interface-sensitive analysis on the respective femtosecond timescales. Second harmonic spectroscopy (SHS) [1] is such an interface-sensitive probe in centrosymmetric materials. We employ a non-collinear optical parametric amplifier in the visible wavelength range (1.9-2.5 eV) for pump-probe SHS with <20 fs pulse duration. A prototypical molecule/metal interface is prepared by adsorption of iron octaethylporphyrin (FeOEP) molecules on Cu(001) [2] and analyzed *in situ* in ultrahigh vacuum. We find a molecule-induced resonance at about 2.2 eV fundamental photon energy in the second harmonic spectrum of one monolayer of FeOEP/Cu(001). At this resonance, we observe a markedly slower relaxation time of the pump-induced changes in SHS compared to the bare Cu(001) surface, which indicates an increased lifetime of the electronic molecular state.

We thank H. Wende, J. Güdde and E. Riedle for valuable experimental advice, and the German Research Foundation for funding via SPP 1840 QUTIF and Sfb 1242.

[1] T. F. Heinz, C. K. Chen, D. Ricard, and Y. R. Shen, Phys. Rev. Lett. **48**, 478 (1982); U. Höfer, Appl. Phys. A **63**, 533 (1996).

[2] H. C. Herper et al., Phys. Rev. B **87**, 174424 (2013).

O 8.11 Mon 13:00 TRE Phy

Dynamics of interfacial electron-hole separation in an organic heterojunction monitored by femtosecond time-resolved X-ray photoelectron spectroscopy — •Friedrich Roth[1], Mario Borgwardt[2], Lukas Wenthaus[3], Johannes Mahl[2], Steffen Palutke[4], Günter Brenner[4], Serguei Molodtsov[1,5], Wilfried Wurth[3,4,6], Oliver Gessner[2], and Wolfgang Eberhardt[3] — [1]Institute of Experimental Physics, TU Bergakademie Freiberg, Leipziger Straße 23, D-09599 Freiberg, Germany — [2]Chemical Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA — [3]Center for Free-Electron Laser Science / DESY, D-22607 Hamburg, Germany — [4]Deutsches Elektronen-Synchrotron DESY, Notkestraße 85, 22603 Hamburg, Germany — [5]European XFEL GmbH, Holzkoppel 4, 22869, Schenefeld, Germany — [6]Universität Hamburg, Luruper Chaussee 149, 22761, Hamburg, German

The dynamics of ultrafast photon-to-charge conversion in a copper-phthalocyanine (CuPc)-C_{60} heterojunction is studied by femtosecond time-resolved X-ray photoemission spectroscopy (tr-XPS) at the free-electron laser FLASH. The technique provides site-specific access to electron dynamics and monitors the generation and decay of interfacial charge-transfer (ICT) states after excitation with 775 nm photons. A previously unobserved channel for ICT separation into mobile charge carriers with an efficiency of 22 ± 7 % is identified, providing a direct measure of the internal quantum efficiency of the heterojunction for this channel.