O 79: Poster Session VI: Ultrafast electron dynamics at surface and interfaces II

Time: Wednesday 13:30–15:30 Location: P

O 79.1 Wed 13:30 P

Momentum Microscope vs. Hemispherical analyzer - a quantitative comparison of electron analyzer performance for time-resolved ARPES experiments — Julian Maklar, Tommaso Pincelli, Samuel Beaulieu, Shuo Dong, Maciej Dendzik, Martin Wolf, Ralph Ernstorfer, and •Laurenz Rettig — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

For angle-resolved photoelectron spectroscopy (ARPES), an important element is the employed electron analyzer. While hemispherical analyzers with angle-dispersing electron lenses have been the working horse for decades, recently time-of-flight based momentum microscopes have shown a huge improvement in parallel detection efficiency, allowing for simultaneous detection of multiple Brillouin zones without the need to rearrange the sample geometry. However, one drawback of such instruments, in particular in time-resolved studies, arises from the large energy and momentum range covered simultaneously, which in combination with detection limitations of delay-line detectors can severely reduce the effective detection rate for selected energy-momentum regions compared to conventional hemispherical analyzers. Additionally, the high electron energies employed in the more complex electron lens system designs impose new constraints in terms of space charge. Using our XUV time-resolved ARPES system hosting both a hemispherical analyzer (SPECS Phoibos 150) and a momentum microscope (SPECS Metis 1000) in one experimental setup, we quantitatively compare the advantages and disadvantages of both types of analyzers for various kinds of trARPES experiments. Rev. Sci. Instr. 91, 123112 (2020)

O 79.2 Wed 13:30 P

Dynamic screening of quasiparticles in WS_2 monolayers — •Stefano Calati[1,2], Qiuyang Li[3], Xiaoyang Zhu[3], and Julia Stähler[1,2] — [1]Department of physical chemistry, Fritz-Haber-Institut der MPG, Berlin — [2]Institut für Chemie, Humboldt-Universität zu Berlin — [3]Department of Chemistry, Columbia University, New York

The low dimensional nature of TMDCs and the resulting reduced screening influence their non-equilibrium optical properties, as dynamic screening by photoexcited quasiparticles governs the transient response. Here, we investigate the respective roles of excitons and quasi-free carriers on the dynamic response of WS_2 monolayers on SiO_2. We find drastic changes in the reflectivity/transmittance contrast upon photoexcitation. The main observation is a pump photon energy-dependent blue/red shifts of the neutral exciton. Based on a phenomenological model, we disentangle the different impact of excitons and free carriers on the renormalization of the quasi-free-particle band gap and exciton binding energy. This work unravels and quantifies the competition and interplay of the multiple electronic and thermal processes contributing to the recovery of the system upon photoexcitation.

O 79.3 Wed 13:30 P

Transient band structure renormalizations and ultrafast exciton dynamics in fullerene thin films — •Sebastian Hedwig, Sebastian Emmerich, Benito Arnoldi, Johannes Stöckl, Benjamin Stadtmüller, and Martin Aeschlimann — Department of Physics and Research Center OPTIMAS, University of Kaiserslautern, Erwin-Schrödinger-Straße 46, 67663 Kaiserslautern, Germany

Fullerenes have been subject to intense research in the past decades, with promising potential for their implementation in optoelectronic devices. Resonant optical excitation of a fullerene thin film leads to a transient renormalization of the polaronic valence band structure which follows the timescale of CT-exciton formation and decay, as recently shown for C_{60} [1,2]. Embedding metal atoms or clusters into the carbon cage can alter the cage symmetry and density of states leading to new relaxation channels, such as the cluster-cage electron transfer. Here, we present time resolved fs UV-pump XUV-probe photoemission studies carried out on thin films of C_{60} and the endohedral metallofullerene $Sc_3N@C_{80}$. Both fullerene complexes reveal almost identical band structure changes upon optical excitation evolving on similar timescales. For $Sc_3N@C_{80}$, we also show that the exciton and polaron dynamics are strongly altered upon K intercalation of the pristine film. This enables us to reveal the role of the cluster-cage charge transfer on the ultrafast carrier dynamics in fullerene thin films [3].

[1] B. Stadtmüller et al., Nat Commun 10, 1470 (2019)
[2] S. Emmerich et al., J. Phys. Chem. C 124, 23579-23587 (2020)
[3] S. Emmerich et al., arXiv:2002.04576 (2020)

O 79.4 Wed 13:30 P

Optical driven 4f-spin and orbital transitions in rare-earth metals — •N. Thielemann-Kühn[1], T. Amrhein[1], W. Bronsch[1], S. Jana[2], N. Pontius[2], R. Y. Engel[3], P. S. Miedema[3], M. Beye[3], B. E. van Kuiken[4], M. Teichmann[4], R. E. Carley[4], L. Mercadier[4], A. Yaroslavtsev[4], G. Mercurio[4], L. Le Guyader[4], N. Agarwal[4], A. Scherz[4], P. M. Oppeneer[5], M. G. Weinelt[1], and C. Schüssler-Langeheine[2] — [1]FU Berlin — [2]HZB — [3]DESY — [4]EuXFEL — [5]Uppsala University

$4f$ rare earth (RE) metals exhibit strongly localized $4f$ states carrying the largest magnetic moment among the elements of the periodic table. Due to their energy-level scheme, $4f$ states are not directly accessible with optical wavelengths in contrast to the delocalized $5d6s$ valence electrons, that mediate the *inter*-atomic $4f$ exchange coupling. Despite the strong *intra*-atomic exchange coupling, local $5d$-$4f$ excitations have neither been observed upon optical excitation nor were considered to be relevant for non-equilibrium dynamics in RE metal based systems. In an X-ray absorption experiment at the EuXFEL we found that the $5d$ and $4f$ system interact directly via inelastic electron scattering and $4f$-$5d$ electron transfer, initiating transitions to higher energetic $4f$ configurations, which becomes pronouncedly visible in a change of the multiplet structure. The observed $4f$ electronic excitation are highly element-specific and strongly depend on pump pulse parameter. They directly cause changes in exchange and electron-phonon coupling and thus open a door for more fundamental understanding of non-equilibrium dynamics.

O 79.5 Wed 13:30 P

Extension of the two-temperature model capturing nonequilibrium electrons after ultrashort laser excitation — •Markus Uehlein, Sebastian T. Weber, and Baerbel Rethfeld — Department of Physics and Research Center OPTIMAS, TU Kaiserslautern, Erwin-Schroedinger-Strasse 46, 67663 Kaiserslautern, Germany

The understanding of materials' reaction on a laser excitation with pulse durations below one picosecond is of significant importance for industrial applications. The energy of ultrashort laser pulses in the visible range is absorbed by the electrons. This creates a non-equilibrium in the electron system that thermalizes typically in the following hundreds of femtoseconds [1]. On longer time scales, electrons and phonons relax to a joint temperature. The latter relaxation process is described by the well-known two-temperature model (TTM) [2]. However, this neglects the electronic non-equilibrium completely.
In this presentation, we investigate an extension of the TTM developed by Carpene [3] and Tsibidis [4]. This extension adds a system of nonthermal electrons to the TTM. We introduce several improvements and discuss selected results of the extended two-temperature model.
[1] B. Y. Mueller and B. Rethfeld, PRB **87**, 035139 (2013)
[2] S. I. Anisimov *et al.*, JETP **39**, 375 (1974)
[3] E. Carpene, PRB **74**, 024301 (2006)
[4] G. D. Tsibidis, Appl. Phys. A **124**, 311 (2018)

O 79.6 Wed 13:30 P

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 examine the effect of spin-orbit coupling.

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O 79.7 Wed 13:30 P

Tracing magnetization dynamics in Fe at the BZ edge by TR-ARPES — •Christian Strüber, Xinwei Zheng, Shabnam Haque, and Martin Weinelt — Freie Universität Berlin, Berlin, Germany

Time- and angle-resolved photoemission spectrocopy (TR-ARPES) allows for the investigation of magnetization dynamics by observing shifts in the electronic band structures and transient changes of the electronic population [1]. The focus of previous experiments often lay on the centre of the Brillouin zone (BZ). However, large parts of the electrons contributing to magnetism are distributed over the whole BZ.

We prepare single-crystalline layers of Fe on W(110) at 300 K and measure TR-ARPES signals at cryogenic temperatures (<100 K). In a pump-probe experiment photoelectrons are excited by ultrashort monochromatic XUV pulses (27.2 eV, 10 kHz, <100 fs) generated via HHG in an argon target. Demagnetization dynamics in the thin-film samples are pumped by intense NIR (770 nm) pulses. We measure the electronic excitation and energy shifts of majority and minority spin bands at the centre of the BZ and for high lateral momentum. We observe different dynamics of bulk and surface resonance states.

Additionally, we observe kinetic energy modulations before the temporal overlap resulting from an interaction with the ponderomotive potential of the pump pulse in front of the Fe sample. We investigate these modulations as a function of kinetic energy, emission angle and pump incidence angle and compare to results in Gd [2].

[1] B.Frietsch et al., *Science Advances* **6**,39(2020)

[2] U. Bovensiepen et al. *Physical Review B* **79**, 045415 (2009)