# O 3: Ultrafast Electron and spin dynamics at interfaces I

Time: Monday 10:30–13:00 Location: MA 005

O 3.1 Mon 10:30 MA 005

**Mechanism of spin-dependent electron transfer on ferromagnetic interfaces: theory and application** — •Simiam Ghan, Harald Oberhofer, and Karsten Reuter — Technical University of Munich, Garching, Germany.

Self-assembled monolayers of organic molecules (SAMs) on surfaces show great promise in the emerging field of molecular electronics due to tunable charge transport properties, long-range 2-dimensional order and ease of manufacture. Growth of SAMs on ferromagnetic surfaces offers the additional possibility of spin-dependent transport, making these systems relevant to molecular spintronics in e.g. spin-valves and magnetic tunneling junctions. In order to establish design principles for such applications, a thorough understanding of charge transport mechanisms over SAM-metal interfaces is of great importance.

As an initial benchmark in this direction, we report calculations of spin-dependent electron transport in model systems of Argon monolayers on ferromagnetic Fe(110), Co(0001) and Ni(111) substrates. Spin-polarized charge transfer rates are calculated from the Fermi Golden Rule and explicit time propagation using first-principles parametrized model Hamiltonians. Results are compared to ultrafast core-hole-clock spectroscopy measurements, which found faster transport for minority electrons [1]. After benchmarking the protocol on Ar-metal systems we apply it to thiol-based model SAMs with an aim towards predicting tunable spin-transport behavior.

[1] F. Blobner *et al.*, Phys. Rev. Lett. **112**, 086801 (2014).

O 3.2 Mon 10:45 MA 005

**Time-evolution of optical excitations in Fe/MgO(001) heterostructures from RT-TDDFT** — •Markus Ernst Gruner, Okan Köksal, and Rossitza Pentcheva — Faculty of Physics and Center for Nanointegration, CENIDE, University of Duisburg-Essen

We investigate the real-time (RT) evolution of an optical excitation in a $(Fe)_n/(MgO)_m(001)$ multilayer system in the framework of time-dependent density functional theory (TDDFT). The calculations are carried out with the ELK code using the adiabatic local spin density approximation for exchange and correlation. Starting with a minimum model consisting of a single Fe layer and 3 MgO layers, we vary systematically frequency and fluence of the laser pulse and analyse the time dependent propagation of excitations in terms of the variation of the charge density and electronic density of states, which gives insight into the evolution of orbital polarisation. Apart from laser frequency and intensity, we find that also the polarization of light significantly influences the magnitude and propagation of the excitations. First trends for larger systems are discussed.

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

O 3.3 Mon 11:00 MA 005

**Femtosecond spin-dependent charge transfer at Co/Cu(001) interfaces** — •Andrea Eschenlohr[1], Jinghao Chen[1], Tristan Müller[2], Peter Elliott[2], Eberhard K. U. Gross[2], Uwe Bovensiepen[1], and Sangeeta Sharma[2] — [1]Faculty of Physics, University of Duisburg-Essen — [2]Theory Department, MPI for Microstructure Physics, Halle (Saale)

The elementary processes behind ultrafast spin transfer at epitaxial Co/Cu(001) interfaces due to optical excitation with 1.5 eV photon energy are disentangled by combining femtosecond time-resolved interface-sensitive magnetization-induced second harmonic generation and ab initio time-dependent density functional theory. We obtain a convincing agreement between the observables in theory and experiment, which allows us to directly identify spin-dependent charge transfer between Co and Cu active at $< 30$ fs, and spin-flips mediated by the spin-orbit interaction, which lead to a loss of spatially integrated spin polarization and dominate at $> 30$ fs. In particular, we find that the charge transfer from Cu into Co is also spin polarized.

We acknowledge funding from DFG through SPP 1840 QUTIF.

O 3.4 Mon 11:15 MA 005

**Signature of electron-boson coupling in the non-equilibrium electron population of a Fe-based High-$T_c$ superconductor** — •Isabella Avigo[1], Setti Thirupathaiah[1], Manuel Ligges[1], Thomas Wolf[2], Jörg Fink[3], and Uwe Bovensiepen[1] — [1]Universität Duisburg Essen, Fakultät für Physik, 47057 Duisburg, Germany — [2]Karlsruhe Institute of Technology, Institut für Festkörperphysik, 76021 Karlsruhe, Germany — [3]IFW-Dresden, Institute for Solid State Research, 01171 Dresden, Germany

Understanding the origin of microscopic interactions among the various electronic, phononic, magnetic degrees of freedom of complex materials, such as High-$T_c$ superconductors, is an essential problem in modern solid state physics. A promising approach is the study of non-equilibrium states and their relaxation in the time domain, as specific coupling channels leave their characteristic fingerprints on the energy-resolved relaxation of the excited electronic population. We present a time-resolved photoelectron spectroscopy (trARPES) study conducted on a Fe-based High-$T_c$ superconductor, where the energy-dependent relaxation rates of the excited electronic population show a characteristic step-behavior around the energy of 200 meV above the Fermi level, indicating the coupling of the excited electronic system to a bosonic excitation, to which we assign a magnetic origin. Our findings further confirm and generalize our previous results obtained on a Cu-based High-$T_c$ superconductor. We acknowledge funding from the DFG through the priority program SPP1458 and the EU within the seventh Framework Program under Grant No. 280555 (GO FAST).

O 3.5 Mon 11:30 MA 005

**Femtosecond time-resolved and element-specific x-ray absorption spectroscopy of Fe/MgO** — •Nico Rothenbach[1], Andrea Eschenlohr[1], Katharina Ollefs[1], Carolin Schmitz-Antoniak[2], Soma Salamon[1], Rolf Mitzner[3], Niko Pontius[3], Uwe Bovensiepen[1], and Heiko Wende[1] — [1]University of Duisburg-Essen and CENIDE — [2]Forschungszentrum Jülich GmbH — [3]Helmholtz-Zentrum Berlin

A localized optical excitation of a metal/insulator heterostructure induces ultrafast dynamics in its individual compounds, which can involve charge and spin transfer processes as well as coupling to low energy excitations mediated by e.g. electron-electron and electron-phonon scattering. Femtosecond soft x-ray spectroscopy facilitates to separate and identify these electronic and lattice excitations directly in the time domain and, furthermore, is sensitive to the dynamics of the individual constituents itself due to its element-specific character. We have measured time- and element-resolved x-ray absorption spectroscopy of a $[2\,\mathrm{nm\,Fe}/2\,\mathrm{nm\,MgO}]_8$ multilayer at the Fe $L$- and O $K$-edges with a time resolution of 150 fs. After optically exciting locally Fe with a UV laser pulse of 266 nm wavelength we see a clear pump-induced effect at both edges in fs time resolution. The Fe-signal shows an ultrafast 0.5% dropdown of the signal in 240 fs, followed by a recovery on a 1 ps timescale, while O reaches its maximum not until 1 ps. This slower response suggests that the energy transfer from the metal to the insulator is mediated by phonons, or a combined electronic-phononic process rather than by a direct charge transfer excitation.

O 3.6 Mon 11:45 MA 005

**Highly efficient end station for space-, time- and spin-resolved photoemission spectroscopy at free electron lasers.** — •D. Kutnyakhov[1], F. Pressacco[2], G. Mercurio[2], L. Wenthaus[2], H. Meyer[2], S. Gieschen[2], A. Oelsner[3], C. Tusche[4,5], Y.J. Chen[4,5], D. Vasilyev[6], K. Medjanik[6], G. Brenner[1], S. Dziarzhytski[1], H. Redlin[1], H.J. Elmers[6], G. Schönhense[6], Y. Acremann[7], and W. Wurth[1,2] — [1]DESY, Hamburg, Germany — [2]CFEL, Univ. Hamburg, Germany — [3]Surface Concept GmbH, Mainz, Germany — [4]FZ Jülich GmbH, Germany — [5]Univ. Duisburg-Essen, Germany — [6]Univ. Mainz, Germany — [7]ETH Zürich, Switzerland

High repetition rate XUV and soft X-ray free electron lasers (FELs) such as FLASH at DESY, Hamburg offer unique possibilities for time-resolved photoelectron spectroscopy (TR-PES). To fully exploit these possibilities it is necessary to use very efficient photoelectrons detection schemes. Combining a time of flight momentum microscope with the FEL as a source is ideal for time- and angle-resolved PES and TR-X-ray photoelectron diffraction to study ultrafast electron- and lattice dynamics. The momentum microscope allows simultaneous detection of the entire band structure with unprecedented efficiency in the full surface Brillouin zone with 3.4 $Å^{-1}$ dia. and 4 eV binding energy range or the angular pattern of core level photoelectrons, respectively, for each time step in a pump-probe experiment. Adding the imaging

spin detector extends the capability to detect the spin polarized band structure of the material. The set up was commissioned at FLASH and first results will be presented. Funding BMBF 05K16PGB.

O 3.7 Mon 12:00 MA 005

**Intra-atomic Delays in Attosecond Time-resolved Solid State Photoemission** — Fabian Siek[1], Sergej Neb[1], Peter Bartz[1], Matthias Hensen[1], Christian Strüber[1], Sebastian Fiechter[2], Miquel Torrent-Sucarrat[3,4,5], Vyacheslav M. Silkin[3,4,5], Eugene E. Krasovskii[3,4,5], Nikolay M. Kabachnik[6,7], Stephan Fritzsche[8], Ricardo Díez Muiño[4,9], Pedro M. Echenique[3,4,9], Andrey K. Kazansky[3,4,5], Norbert Müller[1], •Walter Pfeiffer[1], and Ulrich Heinzmann[1] — [1]University of Bielefeld, Germany — [2]Institut für Solare Brennstoffe, Germany — [3]University of the Basque Country, Spain — [4]Donostia International Physics Center, Spain — [5]IKERBASQUE, Basque Foundation for Science, Spain — [6]Skobeltsyn Institute of Nuclear Physics, Lomonosov Moscow State University, Russia — [7]European XFEL GmbH, Germany — [8]Helmholtz-Institut Jena, Germany — [9]Centro de Física de Materiales, Spain

Attosecond time-resolved photoemission on the semiconductor $WSe_2$ allows investigating temporal delays from different initial states with unprecedented resolution. The observed delays cannot be attributed to photoelectron propagation in the solid but intra-atomic corrections such as the formation of a centrifugal barrier must be taken into account to match the experimental results. This sheds new light on the fundamental mechanism involved in the photoemission process from solids on the very initial stage of photoelectron emission, i.e. for the very short time window the emitted electron still resides inside the atom from which it is emitted.

O 3.8 Mon 12:15 MA 005

**Femtosecond momentum- and energy resolved electron thermalization dynamics in Cu(100) single crystal studied by time-resolved ARPES** — •S. Y. Agustsson[1], D. Vasilyev[1], D. Kutnyakhov[2], K. Medjanik[1], S. Babenkov[1], S. Dziarzhytski[2], H. Redlin[2], D. Curcio[3], F. Pressacco[4], C. Tusche[5,6], Y. Chen[5,6], K. Bühlmann[7], Y. Acremann[7], W. Wurth[2,4], J. Demsar[1], G. Schönhense[1], and H. Elmers[1] — [1]Univ. Mainz — [2]DESY Photon Science — [3]Univ. Aarhus, Denmark — [4]Univ. Hamburg, — [5]FZ Jülich — [6]Univ. Duisburg-Essen — [7]ETH Zürich

The understanding of the photoexcited electron-electron and electron-phonon thermalization dynamics in simple metals and their relation to the inter-subsystem (electron-phonon) coupling strength presents a foundation for similar studies in advanced quantum matter. However, despite the three decades of ongoing research, the understanding is still elusive. Here, we present the first time-, energy-, and momentum resolved photoemission studies of carrier dynamics in Cu (100) single crystal. We used a time of flight momentum microscope with the FEL (FLASH at DESY) as a source (55 eV, <100 fs) to track the changes in the electronic distribution function following photoexcitation with <100 fs NIR pulses from a synced high-repetition rate Ti:Sa amplifier. We demonstrate that the intra-band photoexcitation with an s-polarized light results in changes in the electronic distribution function, which is strongly momentum-dependent. Surprisingly, the momentum anisotropy is observed throughout the electron thermalization process.

O 3.9 Mon 12:30 MA 005

**Ultrafast electron and spin dynamics in antiferromagnetic rare-earth intermetallics** — •Y. W. Windsor[1], C. Nicholson[1], A. Fedorov[2], M. Puppin[1], K. Kummer[3], K. Kliemt[4], C. Krellner[4], C. Schüssler-Langeheine[5], N. Pontius[5], U. Staub[6], M. Wolf[1], R. Ernstorfer[1], D. V. Vyalikh[7], and L. Rettig[1] — [1]Fritz-Haber-Institut (DE) — [2]IFW Dresden (DE) — [3]ESRF (FR) — [4]Goethe-Univ. (DE) — [5]HZB (DE) — [6]PSI (CH) — [7]DIPC (ES)

Antiferromagnets (AFM) promise much faster magnetization dynamics than ferromagnets (FM), and so are promising candidates for future ultrafast spintronic applications. Here we study the ultrafast electron and spin dynamics in RE-Rh2Si2 AFMs (RE is a rare earth), which exhibit an intriguing coupling of bulk AFM order to a FM surface evidenced by a spin-split surface state [1].

Using XUV time-resolved ARPES (trARPES) and time-resolved resonant X-ray diffraction (trRXD), we obtain a comprehensive view of the ultrafast demagnetization of the bulk AFM and the FM surface (following optical excitation). We observe similar dynamics for the two orders (bulk AFM order and FM spin split surface state), indicating a strong coupling between the two. Comparison of the demagnetization timescales for different RE ions suggests a strong influence of the 4f L moment and spin-lattice coupling on demagnetization, similar to studies on RE metals [2]. Finally, we observe a coherent rotation of the entire AFM structure, which can be controlled by laser fluence.

[1] Güttler, Sci.Rep. 6, 24254 (2016)
[2] Wietstruck, PRL 106, 127401 (2011)

O 3.10 Mon 12:45 MA 005

**Direct observation of magnetite surface dynamics by Fast Scanning Tunneling Microscopy** — Barbara A. J Lechner, Alexander Bourgund, Ueli Heiz, and •Friedrich Esch — Department of Chemistry & Catalysis Research Center, Technical University of Munich, Lichtenbergstr. 4, D-85748 Garching

Magnetite shows a rich surface chemistry and is a well-known particle support in catalysis [1]. While its surface dynamics have been studied extensively with conventional STM and other techniques [1-3], little is known about these surface processes when studied at the atomic scale at elevated temperatures, with the required high temporal resolution. We present results on the Fe3O4(001) surface dynamics taken with a specially developed FastSTM add-on module [4] that boosts the temporal resolution of our commercial STM up to 12 frames/s, demonstrating the potential of this technique: Surface species and defects that appear similar topographically unravel their different dynamic behavior when increasing temperature stepwise (up to 800 K). In particular, we follow the mobility of H adatoms and of domain boundaries, we observe the subsurface diffusion of Fe atoms and their exchange with the bulk and provide additional insight into the high-temperature transition [3]. Furthermore, the surface morphology is studied under reducing or wet conditions, as well as in an oxygen atmosphere.

References [1] G. S. Parkinson, Surf. Sci. Rep. 2016, 71, 272. [2] R. Bliem et al., Angew. Chem. Int. Ed. 2015, 54, 13999. [3] N. C. Bartelt et al., Phys. Rev. B 2013, 88. [4] F. Esch et al., Rev. Sci. Instrum. 2011, 82, 53702 and NFFA project JRA1.