O 13: Ultrafast Electron Dynamics at Surfaces and Interfaces II

Time: Monday 15:00–17:30 Location: MA 041

Topical Talk O 13.1 Mon 15:00 MA 041

Photocatalysis at the surface the TiO_2 on its real time — Michael Wagstaffe[1], Lukas Wenthaus[2], Adrian Dominguez-Castro[3], Dmytro Kutnyakhov[2], Siarhei Dziarzhytski[2], Simon Chung[1], Guilherme Dalla Lana Semione[1], Steffen Palutke[2], Federico Pressacco[2], Michael Heber[2], Giuseppe Mercurio[2], Harald Redlin[2], Helena Gleissner[1], Verena Kristin Gupta[3], Nicolai Klemke[4], Yudong Yang[4], Thomas Frauenheim[5], Adriel Dominguez[6], Franz Kärtner[4], Angel Rubio[7], Wilfried Wurth[2], Andreas Stierle[1], and •Heshmat Noei[1] — [1]Centre for X-ray and Nanoscience (CXNS), Deutsches Elektronen-Synchrotron (DESY), Hamburg, Germany — [2]Deutsches Elektronen-Synchrotron (DESY), Notkestr. 85 D-22607, Hamburg, Germany — [3]Bremen Center for Computational Material Science (BCCMS), Uni- versity of Bremen, Bremen, Germany — [4]Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany — [5]Computational Science and Applied Research Institute (CSAR), 518110, Shenzhen, China — [6]Nano-Bio Spectros- copy Group, Departamento de Fisica de Materiales, San Sebastián, Spain; — [7]Max Planck In- stitute for the Structure and Dynamics of Matter, Hamburg, Germany

The femtosecond resolution soft X-ray photoemission spectroscopy results are combined with theoretical calculations to provide crucial insight concerning reaction mechanisms and dynamics of the interaction of water, oxygen and carbon monoxide with the surface of TiO_2 and the interfacial charge transfer during the initial steps of the reaction.

O 13.2 Mon 15:30 MA 041

Photo-induced charge-transfer renormalization in NiO — •T. Lojewski[1], D. Golez[2,3], K. Ollefs[1], L. Le Guyader[4], L. Kämmerer[1], N. Rothenbach[1], R. Y. Engel[5], P. S. Miedema[5], M. Beye[5], G. S. Chiuzbaian[6], R. Carley[4], R. Gort[4], B. E. Van Kuiken[4], G. Mercurio[4], J. Schlappa[4], A. Yaroslavtsev[4,7], A. Scherz[4], F. Döring[8], C. David[8], H. Wende[1], U. Bovensiepen[1,9], M. Eckstein[10], P. Werner[11], and A. Eschenlohr[1] — [1]Univ. Duisburg-Essen and CENIDE — [2]Jozef Stefan Inst. — [3]Univ. of Ljubljana — [4]European XFEL — [5]DESY — [6]Sorbonne Univ. — [7]Uppsala Univ. — [8]PSI — [9]Univ. of Tokyo — [10]Univ. of Hamburg — [11]Univ. of Fribourg

Strongly correlated materials, such as the charge transfer insulator NiO, exhibit fascinating properties due to the interaction-induced localisation of electrons, which competes with their itinerant nature. Here, optical excitation of charge carriers, i.e. photodoping, results in complex dynamics involving both *d-p* and *d-d* excitations. Through a resonant optical pump, femtosecond time-resolved X-ray absorption probe experiment in combination with dynamical mean-field theory, we analyse these dynamics in photo-doped NiO. We find a redshift of the Ni L_3 and O K edges (persisting for > 10 ps), relating to a simultaneous occurrence of Hartree shifts and a renormalisation of local interactions. In addition, we observe a feature below the Ni L_3 edge (at < 1 ps), which we relate to a transient nonthermal population of local many-body multiplets [1]. Financial support by DFG through SFB 1242 is acknowledged. - [1] T. Lojewski et al., arXiv:2305.10145

O 13.3 Mon 15:45 MA 041

Spatio-Temporal Electron Propagation Dynamics in Au/Fe/MgO(001) in Nonequilibrium Analyzed by Femtosecond Two-Photon Photoemission — •Florian Kühne[1], Markus Heckschen[1], Yasin Beyazit[1], Elaheh Shomali[1], Jesumony Jayabalan[1], Ping Zhou[1], Detlef Diesing[2], Markus Gruner[1], Rossitza Pentcheva[1], Axel Lorke[1], Björn Sothmann[1], and Uwe Bovensiepen[1] — [1]Fakultät für Physik, Universität Duisburg-Essen and CENIDE, Duisburg D-47048, Germany — [2]Fakultät für Chemie, Universität Duisburg-Essen and CENIDE

Optically excited electrons in metals travel ballistically on a femtosecond timescale until they scatter due to electron-electron or electron-phonon interactions. To gain a deeper understanding of the ballistic and few times scattered electrons under non-equilibrium conditions we combine time-resolved two-photon photoelectron emission spectroscopy with real-time time-dependent density functional theory(RT-TDDFT) as well as a random-walk-like transport simulation, see Heckschen et al., PRX Energy **2**, 043009(2023). Here, we discuss the experimental details using a back-side pumped geometry on a MgO/Fe/Au epitaxial heterostructure probing the Au surface. The time-delayed response of the photoelectrons at lower energies is used to investigate transport and scattering pathways. Analyzing this time delay of 10 to 100fs shows an apparent acceleration with increasing film thickness. We find the electron trajectories angular dependence is of key importance to explain this effect. Funding by the DFG through Project No. 278162697 - SFB1242 is gratefully acknowledged.

O 13.4 Mon 16:00 MA 041

Spatio-Temporal Electron Propagation Dynamics in Au/Fe/MgO(001) in Nonequilibrium: A Random Walk Simulation — •Markus Heckschen[1], Yasin Beyazit[1], Elaheh Shomali[1], Florian Kühne[1], Jesumony Jayabalan[1], Ping Zhou[1], Detlef Diesing[2], Markus E. Gruner[1], Rossitza Pentcheva[1], Axel Lorke[1], Björn Sothmann[1], and Uwe Bovensiepen[1] — [1]Fakultät für Physik, Universität Duisburg-Essen and CENIDE, Duisburg D-47048, Germany — [2]Fakultät für Chemie, Universität Duisburg-Essen and CENIDE, Essen D-45711, Germany

Electron transport in metals can be either ballistic, superdiffusive or diffusive. We analyze time resolved two-photon photoelectron emission (2PPE) spectroscopy performed in a back-pump front-probe geometry on MgO/Fe/Au which provides information about the nonequilibrium electron distribution as a function of time, energy and Au layer thickness [1].

Here, we present a trajectory-based Monte-Carlo simulation of classical, quasiballistic electrons that undergo inelastic electron-electron scattering. Our simulation reproduces well the experimentally observed 2PPE spectra and allows us to identify high-energy electrons passing ballistically through the sample, as well as low-energy electrons which are transported in a superdiffusive manner.

[1] M. Heckschen et al., PRX Energy **2**, 043009 (2023).

O 13.5 Mon 16:15 MA 041

Thermal boundary conductance of ultrathin epitaxial Pb films on Si(111) — •Christian Brand[1], Tobias Witte[1], Mohammad Tajik[1], Jonas D. Fortmann[1], Michael Horn-von Hoegen[1], Laurenz Rettig[1,2], and Uwe Bovensiepen[1] — [1]University of Duisburg-Essen, Duisburg, Germany — [2]Fritz Haber Institute, Berlin, Germany

The non-equilibrium dynamics of electrons in a metal subsequent to excitation with a fs-laser pulse couple to other degrees of freedom during the relaxation phase of the system. Here we have studied the electron and lattice dynamics in ultrathin Pb films grown on Si(111) by means of time-resolved photoemission spectroscopy and reflection high-energy electron diffraction. After thermalization of the electron and lattice system, the transient cooling of the film is determined by heat transport across the interface into the substrate, i.e., by the thermal boundary conductance (TBC) on time scales of a few 100 ps. We experimentally find for crystalline Pb films with low defect density at a base temperature of 19.3 K a TBC of less than 2 MW/m^2K. We discuss the results in terms of diffuse and acoustic mismatch models.

O 13.6 Mon 16:30 MA 041

Electron-phonon coupling in ultrathin Pb films on Si(111): Where the heck is the energy? — •Mohammad Tajik[1], Tobias Witte[1], Christian Brand[1], Laurenz Rettig[2], Björn Sothmann[1], Uwe Bovensiepen[1], and Michael Horn-von Hoegen[1] — [1]Department of Physics, Universität Duisburg-Essen, Lotharstraße 1, 47057 Duisburg, Germany — [2]Department of Physical Chemistry Fritz Haber Institute Faradayweg 4-6 14195 Berlin Germany

In this work, we studied the heat transfer from electron to phonon subsystem within a five monolayer thin epitaxial Pb film on Si(111) upon fs-laser excitation. The response of the electron subsystem was determined using time-resolved photoelectron spectroscopy (tr-PES) while the lattice excitation was measured by means of the Debye-Waller effect in time-resolved reflection high-energy electron diffraction (tr-RHEED). The electrons lose their heat in less than 0.7 ps while the lattice temperature rises slowly in 3.5 to 8 ps. This raises the question where is the energy hidden for 3-7 ps? Within a three-temperature model we used three heat baths, namely electrons, high-frequency and low-frequency phonon modes to simulate the observations. We pro-

pose that the hidden energy is transiently stored in high-frequency phonon modes at the zone boundary for which the tr-RHEED is insensitive and which are excited in less than 0.7 ps. The excitation of low-frequency acoustic phonons, i.e., thermalization of the lattice is facilitated through anharmonic phonon-phonon interaction.

O 13.7 Mon 16:45 MA 041

Non-Equilibrium Pathways for Excitation of Bulk and Surface Phonons through Anharmonic Coupling — C. Brand[1], V. Tinnemann[1], A. Hanisch-Blicharski1[1,3], M. Tajik[1], •J. D. Fortmann[1], A. Kassen[1], F. Thiemann[1], and M. Horn-von Hoegen[1,2] — [1]Department of Physics, University of Duisburg-Essen, 47048 Duisburg, Germany — [2]Center for Nanointegration (CENIDE), 47048 Duisburg, Germany — [3]present address: Institut für IT-Management & Digitalisierung, FOM Hochschule, Germany

Upon impulsive optical excitation of solid-state materials, the non-equilibrium flow of energy from the excited electronic system to the lattice degrees of freedom typically happens in a few picoseconds. Here we identified the surface of thin Bi films grown on Si(001) as an additional subsystem which is excited much slower on a 100 ps timescale that is caused by decoupling due to mismatched phonon dispersions relations of bulk and surface. Anharmonic coupling among the phonon systems provides pathways for excitations which exhibits a 1/T-dependence causing a speed-up of surface excitation at higher temperatures. A quantitative justification is provided by phonon Umklapp processes from lattice thermal conductivity of the Bi bulk. Three-temperature model simulations reveal a pronounced non-equilibrium situation up to nanoseconds: initially, the surface is colder than the bulk, that situation is then inverted during cooling and the surface feeds energy back into the bulk phonon system.

O 13.8 Mon 17:00 MA 041

Temporal evolution of energy-resolved non-thermal electron densities — •Christopher Seibel, Markus Uehlein, Tobias Held, Sebastian T. Weber, and Baerbel Rethfeld — Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau

When a metal is irradiated with an ultrashort laser pulse, the initially Fermi-distributed electrons are excited to a state far from equilibrium. These hot electrons can be exploited for numerous processes and applications, such as photodetection, solar energy conversion, and photocatalysis. However, the non-thermal electrons rapidly thermalize by collisions with each other, thereby limiting their availability for specific applications. Thus, it is crucial to understand the microscopic processes that determine the timescales of hot electron thermalization.

Here, we use a kinetic model based on full Boltzmann collision integrals to trace the non-equilibrium dynamics of the electronic distribution function during excitation and thermalization. We evaluate the time-dependent electron densities in various energy intervals and show the dependence of the dynamics of these spectral electron densities on the excitation conditions. We find energy regions where the interplay between primary and secondary electron generation leads to a behavior revealing a long-lasting non-equilibrium that cannot be explained with a single relaxation time [1].

[1] C. Seibel *et al.*, J. Phys. Chem. C (2023), DOI: 10.1021/acs.jpcc.3c04581

O 13.9 Mon 17:15 MA 041

Band-resolved relaxation of laser-excited gold — •Tobias Held, Stephanie Roden, Pascal D. Ndione, Sebastian T. Weber, and Baerbel Rethfeld — Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau

When a noble metal is irradiated with a short-pulsed laser in the visible spectrum, sp- and d-electrons are excited into energetically higher free states while the phonons are not directly affected. This process increases the energy content of the electron system, alters the partial electron densities and induces non-equilibrium electron distributions. We investigate how the non-equilibrium evolves in the individual bands towards Fermi distributions using the Boltzmann equation with full Boltzmann collision integrals for the excitation, electron-electron scattering and electron-phonon scattering, respectively.

Following the fully kinetic stage, when a temperature description is valid, an occupational non-equilibrium can still persist between sp- and d-electrons [1]. We investigate how this occupational non-equilibrium affects the electron-phonon coupling parameter, finding a strong dependence of the coupling strength on the band occupation [2]. We also observe signatures of features of the partial densities of states in the coupling parameter.

[1] P. D. Ndione, S. T. Weber, D. O. Gericke and B. Rethfeld, Scientific Reports 12, 1 (2022)
[2] T. Held, S. T. Weber and B. Rethfeld, arXiv:2308.01067 (2023)