

O 108: Electron-Driven Processes at Surfaces and Interfaces

Time: Thursday 15:00–17:45

Location: TRE Ma

O 108.1 Thu 15:00 TRE Ma

Probing Optical Modes of Gold Tapers Using Electron Beams — ●NAHID TALEBI¹, SURONG GUO¹, MARTIN ESMANN², MATHIEU KOCIAC³, CHRISTOPH LIENAU², and PETER VAN AKEN¹ — ¹Max Planck Institute for Solid State Research, 70569 Stuttgart, Germany — ²Birkenweg 20 — ³Laboratoire de Physique des Solides, Université Paris Sud, Orsay 91400, France

Three-dimensional gold tapers have fabulous applications in adiabatic nanofocusing (ACS Nano 6 (7), 6040-6048 (2012)), tailoring photoemission processes (ACS Photonics 5 (9), 3584-3593 (2018)), and optical microscopy (Rev. Sci. Instr. 75, 837 (2004)). Recently, it has been also demonstrated that through a combination of adiabatic nanofocusing and scanning-near-field optical microscopy, weak and strong interactions between nanostructures and gold tips can be nicely probed (Nature Nanotechnology 14, 698*704(2019)). Here, we investigate the interaction of optical gold tapers with electron beams. Using electron-energy loss spectroscopy (Nano letters 16 (10), 6137-6144 (2016)) and cathodoluminescence (ACS photonics 6 (10), 2509-2516 (2019)), optical modes of gold tapers are thoroughly investigated, covering both radiating and nonradiating modes. Moreover, we will show that the synchronization of the electron beams with the launched plasmons with high angular momentum orders lead to pronounced resonances in the electron-energy-loss spectra.

O 108.2 Thu 15:15 TRE Ma

Metastable Configurations of a Molecule on a Scanning Probe Tip — ●HADI AREFI^{1,2}, MARVIN KNOL^{1,2}, DANIEL CORKEN³, FRANK S. TAUTZ^{1,2}, REINHARD MAURER³, and CHRISITAN WAGNER^{1,2} — ¹Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, 52425 Jülich, Germany — ²Jülich Aachen Research Alliance (JARA) Fundamentals of Future Information Technology, 52425 Jülich, Germany — ³Department of Chemistry, University of Warwick, Gibbet Hill, Coventry, CV4 7AL, United Kingdom

For scanning probe based molecular nanorobotics and tip-attached single molecule devices, the tip-molecule bonding scheme plays an important role, determining for example, the effective geometric degrees of freedom of the molecule. Exploring various bonding geometries of a PTCDA (3,4,9,10-perylene-tetracarboxylic acid dianhydride) molecule to a pyramid-shaped silver tip, we have studied the potential energy landscape of the molecule using density functional theory. Furthermore, we investigated the effect of the tip shape on the potential energy landscape. Our simulations as well as recent experiments indicate coherently that a bonding scheme different from the originally assumed single tip-molecule bond is present for the PTCDA / Ag-tip system and that the planar PTCDA with carbonyl groups forms two covalent bonds with the tip at sufficiently large tip-surface separations, in similarity to the upright standing PTCDA molecules on a pedestal of two addatoms on the surface [T. Esat, N. Friedrich, F. S. Tautz, and R. Temirov. Nature 558, 573 (2018)].

O 108.3 Thu 15:30 TRE Ma

Two-color coherent control in photoemission from a metal needle tip — ●ANG LI¹, YIMING PAN², PHILIP DIENSTBIER¹, and PETER HOMMELHOFF¹ — ¹Department Physik, Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU), 91058 Erlangen — ²Department of Physics of Complex Systems, Weizmann Institute of Science, Rehovot 7610001, Israel

Nanotips are routinely used as electron sources in high-resolution electron microscopes [1] and as a basis for studying strong-field phenomena at the surface of solids [2]. Two-color laser-induced photoemission from these nanotips shows high visibility in photoemission with femtosecond coherent control [3,4]. Here, we report electron emission studied as a function of the laser wavelength. High visibilities of up to 95.8% in the photoemission has been observed. Further, the photoemission is studied as a function of the ratio of two laser intensities and for a large range of laser intensities. We present an exact quantum mechanical solution based on quantum-pathway interference. Supported by the measured results, this analysis delivers a general explanation to the two-color coherent control in photoemission on a metal nanotip.

[1] A. V. Crewe et al., Rev. Sci. Instrum. 39, 576 (1968).

[2] M. Krüger et al., J. Phys. B, 51, 172001 (2018).

[3] M. Förster et al., Phys. Rev. Lett. 117, 217601 (2016).

[4] T. Paschen et al., J. Mod. Opt. 64, 1054-1060 (2017).

O 108.4 Thu 15:45 TRE Ma

Mechanism of ultrafast, spin-dependent electron transfer on ferromagnetic interfaces: an ab initio study — ●SIMIAM GHAN, KARSTEN REUTER, and HARALD OBERHOFER — Chair of Theoretical Chemistry, Technical University of Munich, Garching, Germany.

Using ab-initio calculations, we explore the mechanism behind an observed spin-dependent electron transfer from photoexcited Argon monolayers into ferromagnetic substrates Fe(110), Ni(111) and Co(0001). A generalized Fermi golden rule is used to calculate transfer rates for majority and minority electrons from donor to acceptor-localized diabatic states. The diabatic states are created through partial diagonalization of the Kohn-Sham Hamiltonian, which also yields inter-state couplings. A faster transfer of minority (down) spins is predicted for Fe and Co substrates, as well as nonpreferential transfer for Ni - all in excellent agreement with ultrafast core-hole clock spectroscopic measurements.

The scheme allows us to compare the roles of donor-acceptor couplings and acceptor density of states in determining a final preferential spin transfer, and highlights the importance of the spacial character of participating states for spin transport on interfaces. Initial applications to self-assembled monolayers of organic molecules are discussed, with the prospect of predicting tunable electron (spin) transport behavior for e.g. organic spintronic devices.

O 108.5 Thu 16:00 TRE Ma

A trip down memory lane: non-Markovian electronic friction effects in ultrafast dynamics at metal surfaces — ●CONNOR BOX and REINHARD J. MAURER — Department of Chemistry, University of Warwick, United Kingdom

Electronic friction theory describes how nuclear motion, in the vicinity of a metallic surface, becomes coupled to electron-hole pair excitations. An efficient and practical DFT implementation based on time-dependent perturbation theory [1] has previously been developed, allowing simulation of vibrational decay [2] as well as molecular beam-like surface scattering. [3] In this work; we investigate the importance of memory effects in nonadiabatic energy loss during vibrational decay and molecular scattering. We propose an efficient and robust approach to calculate memory dependent electronic friction and study its effects on a number of well-studied systems including carbon-monoxide adsorbed at Cu(100) and Pt(111). Finally, we explore different approaches to incorporate memory-dependent electronic friction into on-the-fly ab-initio molecular dynamics simulations to achieve a description of nonadiabatic effects beyond the quasi-static limit. [1] PRB 94, 115432 (2016) [2] PRL 116, 217601 (2016) [3] PRL 118, 256001 (2017)

O 108.6 Thu 16:15 TRE Ma

Photocarrier dynamics in CsPbBr₃ perovskite single crystals — ●SERHII POLISHCHUK¹, MICHELE PUPPIN¹, ALBERTO CREPALDI², DMITRY DIRIN³, OLGA NAZARENKO³, MAKSYM KOVALENKO³, MARCO GRIONI², and MAJED CHERGUI¹ — ¹LSU and LACUS, EPFL, Lausanne, Switzerland — ²IoP and LACUS, EPFL, Lausanne, Switzerland — ³LAC, ETHZ, Zurich, Switzerland

The ability to harvest hot carrier energy can elevate the power conversion efficiency of modern single junction solar cells by up to 25%. In conventional solar materials the photogenerated charge carriers lose the excess electronic energy before the charge collection happens. The understanding of the energy dissipation mechanisms is the fundamental step towards the emergence of hot carrier photovoltaics. The time- and angle-resolved photoelectron spectroscopy (TR-ARPES) is a direct probe of the electronic structure and carrier dynamics of solar materials. The photogenerated electron cooling to the conduction band minimum has been visualized and tracked by TR-ARPES in fully-inorganic CsPbBr₃ perovskite single crystals. The results yield the cooling timescale of approximately 250 fs, and the sub-ps population decay. While hot phonon bottleneck and Auger heating are considered to govern electron cooling dynamics, the surface and Auger recombination, and the diffusion into the bulk supposedly define the apparent population decay. These findings showcase the capabilities of this experimental approach and constitute an important milestone in understanding the processes controlling the ultrafast photocarrier dynamics

in CsPbBr₃ semiconductor.

O 108.7 Thu 16:30 TRE Ma

Simulating Electromigration in Metallic Atomic Contacts — ●MARKUS RING¹, FABIAN PAULY^{1,2}, and PETER NIELABA¹ — ¹Universität Konstanz, Konstanz, Deutschland — ²Okinawa Institute of Science and Technology, Okinawa, Japan

This talk presents the results of ab-initio simulations of metallic contacts with the aim to understand current-induced structure changes and switching. To do this the effect of the electronic motion is integrated into a Langevin equation for the atoms.

The metallic contacts are investigated with density functional theory (DFT) for the atomic and electronic structure as well as the interaction between the two. All of these properties are calculated perturbatively with nonequilibrium Green's function methods to determine transport properties. Among those properties are current-induced forces acting on the atoms. Those forces are modeled in a Langevin equation for the atoms, whose dynamics and kinetics are analyzed.

The primary results are threshold voltages at which vibration modes of the system turn unstable in the presence of non-equilibrium transport. These threshold voltages are investigated for a significant sample of contact realisation changing distance and lead structure for four metals (Pb, Au, Al, Cu) and compared with experimental switching voltages. Finally the unstable vibration modes are used to search and find additional stable conductance states. Overall this is an approach to bridging the time-scale gap between electronic and atomic motion and simulating switching and electromigration of atomic contacts from first principles.

O 108.8 Thu 16:45 TRE Ma

Superoxide formation near DMSO/Cu(111) interfaces — ●ANGELIKA DEMLING¹, SARAH B. KING², and JULIA STÄHLER¹ — ¹Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14169 Berlin — ²Department of Chemistry and James Franck Institute, University of Chicago, 929 E 57th St Chicago, IL 60637

DMSO is a common non-aqueous solvent with the potential to be used in lithium-oxygen batteries due to its ability to catalyze the superoxide formation [1]. In this study we investigate the electron dynamics in DMSO films on Cu(111) using time- and angle-resolved two-photon photoemission and disentangle the individual steps of charge transfer leading to superoxide formation:

Electrons are injected from the metal to the DMSO, where they form small polarons on sub-picosecond time scales. The subsequent trapping extends the electronic lifetimes to several seconds [2]. Co-adsorption of molecular oxygen quenches the trapped electron signal and leads to the appearance of an occupied state whose binding energy is in good agreement with electrochemical data of superoxide [3]. These results may contribute to a basic understanding of charge transfer processes in novel battery systems.

[1] K. M. Abraham, *J. Electrochem.* 162, A3021, (2015)

[2] S. B. King et al., *J. Chem. Phys.*, 150, 041702 (2019)

[3] D. T. Sawyer et al., *J. Electroanal. Chem.* 12, 90 (1966)

O 108.9 Thu 17:00 TRE Ma

Structural Dynamics in the Si(111)-In Atomic Wire System Studied by femtosecond-RHEED: Excitation, Metastable State and Relaxation — ●CHRISTIAN BRAND¹, BERND HAFKE¹, TOBIAS WITTE¹, DAVID JANOSCHKA¹, PASCAL DREHER¹, FRANK-J. MEYER ZU HERINGDORF^{1,2}, and MICHAEL HORN-VON HOEGEN¹ — ¹Universität Duisburg-Essen and CENIDE, Germany — ²ICAN, Duisburg, Germany

The prototypical atomic wire system, i. e. the (4 × 1) reconstructed In atomic wires on Si(111) undergoes a first order phase transition to a

Peierls distorted (8 × 2) reconstructed ground state at $T_c = 130$ K. The structural dynamics of these wires upon impulsive excitation by intense fs-IR laser pulses is probed through time resolved electron diffraction and proceeds via an accelerated displacive structural transition to the (4 × 1) state in only 700 fs [Nature 544, 207 (2017)]. An energy barrier for the atoms collective motion from the excited (4 × 1) state to the (8 × 2) state hinders the immediate recovery of the ground state: the In wires remain for many nanoseconds in a super cooled metastable (4 × 1) state. The relaxation happens through the nucleation of the (8 × 2) ground state at step edges which then trigger a recrystallization front. This front propagates 1-dimensionally at a speed of ~ 100 m/s as determined from a correlation of the step morphology (as obtained by PEEM) to the recovery time constant [Struc. Dyn. 6, 045101 (2019)].

O 108.10 Thu 17:15 TRE Ma

Excitation of a CDW Amplitudon Mode in Indium Atomic Wires on Si(111) studied by Femtosecond Surface Electron Diffraction — BERND HAFKE¹, TOBIAS WITTE¹, CHRISTIAN BRAND¹, and ●MICHAEL HORN-VON HOEGEN^{1,2} — ¹Fakultät Physik, Universität Duisburg-Essen — ²Center for Nanointegration Duisburg-Essen, CENIDE

The ground state of the Indium atomic wire system on Si(111) is Peierls distorted with periodicity doubling along and perpendicular to the wires, thus resulting in a (8 × 2) reconstruction. This low-temperature state comes with a metal to insulator transition, opening of a bandgap, and formation of a CDW and exhibits a by 0.1 eV lower energy than the metallic high-symmetry (4 × 1) state. Here, we report on the excitation of an amplitudon mode of the CDW system by impulsive optical excitation through an IR fs laser pulse under conditions of weak laser fluence < 1 mJ/cm². The observed intensity oscillation in ultra-fast time resolved electron diffraction at a period of 1.4 ps can be described by a superposition of the two characteristic phonon modes, namely the soft rotational and soft shear mode at frequencies of 0.82 THz and 0.54 THz, respectively, which at laser fluences > 2 mJ/cm² facilitate the systems non-thermally driven transition to the excited (4 × 1) state [Nature 544 (2017) 207].

O 108.11 Thu 17:30 TRE Ma

Coherent vibrational control of the Peierls transition in atomic Indium wires on Si(111) — ●JAN GERRIT HORSTMANN, HANNES BÖCKMANN, BARELD WIT, FELIX KURTZ, GERO STORECK, and CLAUS ROPERS — IV. Physical Institute, University of Göttingen, Friedrich-Hund-Platz 1, Germany

Exerting active optical control over solids by harnessing the energy stored in the electronic, lattice or spin degrees of freedom is a key objective of ultrafast science. In femtochemistry, the coherences in the electronic and vibrational states of molecules are already employed to steer reactions towards states or products far from equilibrium. However, a transfer of this concept to solids is complicated, e.g. due to a high electronic and vibrational density of states. In this respect, low-dimensional and strongly correlated systems represent a promising intermediate between molecules and solids, with phase transitions assuming the role of a "reaction". Here, we report coherent vibrational control over the phase transition in a quasi-one-dimensional Peierls insulator by manipulating the vibrational amplitudes of decisive lattice modes. Specifically, we employ an optical double-pulse excitation scheme and monitor the structural transformation between the insulating (8 × 2) and the metallic (4 × 1) phase of atomic indium wires on Si(111) by ultrafast low-energy electron diffraction (ULEED). An analysis of the delay-dependent phase transition efficiency proves the distinct roles of specific phonon modes for the transition and provides insight into the underlying control mechanisms and structural pathways.