

O 63: Poster Session V: Electron-driven processes at surfaces and interfaces

Time: Wednesday 10:30–12:30

Location: P

O 63.1 Wed 10:30 P

Quantifying the breakdown of electronic friction theory during molecular scattering of NO from Au(111) — ●CONNOR L. BOX¹, YAOLONG ZHANG², RONGRONG YIN², BIN JIANG², and REINHARD J. MAURER¹ — ¹Department of Chemistry, University of Warwick, United Kingdom — ²Department of Chemical Physics, University of Science and Technology of China, Hefei, China

The multiquantum vibrational energy loss recorded during molecular scattering from metallic surfaces is a testament to the breakdown of the Born-Oppenheimer approximation. Vibrational state-to-state scattering of NO on Au(111) has been one of the most studied examples in this regard, providing a testing ground for developing various nonadiabatic theories. However, the exact failings compared to experiment and their origin from theory are not established for any system because dynamic properties are affected by many compounding simulation errors of which the quality of nonadiabatic treatment is just one. We use a high dimensional machine learning representation of energy and electronic friction tensor to minimize errors that arise from quantum chemistry.[1,2] This allows us to perform a comprehensive quantitative analysis of the performance of molecular dynamics with electronic friction in describing state-to-state scattering. We find that electronic friction theory accurately predicts elastic and single-quantum energy loss, but underestimates multi-quantum energy loss and overestimates molecular trapping at high vibrational excitation. Our analysis reveals potential remedies to these issues. [1] R. Yin et al, J. Phys. Chem. Lett, 2019 [2] C. L. Box et al, JACS Au, 2020

O 63.2 Wed 10:30 P

Superoxide formation near DMSO/Cu(111) interfaces — ●ANGELIKA DEMLING^{1,2}, SARAH B KING³, and JULIA STÄHLER^{1,2} — ¹Institut für Chemie, Humboldt-Universität zu Berlin, Berlin — ²Fritz-Haber-Institut der MPG, Berlin — ³Department of Chemistry, University of Chicago, Chicago

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]. Under co-adsorption of O₂ another intermediate state arises, whose binding energy is in good agreement with electrochemical data of superoxide [3]. A phenomenological model reproduces the time-dependent data and shows that trapped electrons act as precursors for the superoxide. Further, our data suggests that more superoxide is formed on the surface than in the bulk of DMSO after diffusion.

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] Donald T. Sawyer et al., J. Electroanal. Chem. 12, 90-101 (1966)

O 63.3 Wed 10:30 P

Investigations of polarons in hematite $\alpha - Fe_2O_3(1\bar{1}02)$ by means of nC-AFM and KMC — ●JESÚS REDONDO¹, PAVEL KOCÁN¹, GIADA FRANCESCHI², FLORIAN KRAUSHOFER², MICHELE RIVA², GARETH S. PARKINSON², MICHAEL SCHMID², ULRIKE DIEBOLD², and MARTIN SETVIN^{1,2} — ¹Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic — ²Institute of Applied Physics, TU Wien, Austria

Polarons are known to strongly influence the catalytic activity and the electronic, magnetic and structural properties of transition metal

oxides and halide perovskites. The study of polaron formation and dynamics is fundamental to understanding the actual mechanisms and yields of catalytic reactions in these materials. A new method for the investigation of electron- and hole- polarons is demonstrated. Charge carriers are injected with the atomic force microscope (AFM) tip into the $\alpha - Fe_2O_3(1\bar{1}02)$ surface. The injected charges form a cloud of charged particles trapped in the lattice. This cloud expands due to electrostatic interactions and thermally activated polaron hopping. Controlled annealing the sample and characterization by Kelvin probe force microscopy (KPFM) provides information on polaron dynamics that is compared to kinetic Monte Carlo (KMC) simulations.

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O 63.4 Wed 10:30 P

Ultrafast lattice dynamics and microscopic energy flow in 3d ferromagnets — ●DANIELA ZAHN¹, FLORIAN JAKOBS², YOAV WILLIAM WINDSOR¹, HÉLÈNE SEILER¹, THOMAS VASILEIADIS¹, TIM BUTCHER³, YINGPENG QI¹, DIETER ENGEL⁴, UNAI ATXITIA², JAN VORBERGER³, and RALPH ERNSTORFER¹ — ¹Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany — ²Freie Universität Berlin, Arnimallee 14, 14195 Berlin, Germany — ³Helmholtz-Zentrum Dresden-Rossendorf, Bautzner Landstraße 400, 01328 Dresden, Germany — ⁴Max-Born-Institut, Max-Born-Straße 2A, 12489 Berlin, Germany

The response of ferromagnets to laser excitation is governed by the interplay of electronic, magnetic and lattice degrees of freedom. The lattice plays an important role in the magnetization dynamics, since it drains energy from the electrons and absorbs angular momentum. Here, we study the lattice response of the 3d ferromagnets nickel, iron and cobalt directly using femtosecond electron diffraction. We compare the experimental results to spin-resolved DFT calculations combined with energy flow models. While the conventional two-temperature model is not in agreement with the experimental results, a consistent description of the laser-induced dynamics is achieved with energy-conserving atomistic spin dynamics simulations. Our results provide a clear picture of the microscopic energy flow between electronic, magnetic and lattice degrees of freedom on ultrafast timescales.

O 63.5 Wed 10:30 P

Transient reflectivity in nonequilibrium warm dense gold — ●PASCAL D. NDIONE¹, SEBASTIAN T. WEBER¹, DIRK O. GERICKE², and BAERBEL RETHFELD¹ — ¹Department of Physics and OPTIMAS Research Center, Technische Universität Kaiserslautern — ²CFSA, Department of Physics, University of Warwick

Ultrashort and intense laser pulses can induce strong modification of materials properties such as optical parameters determining the amount of absorbed energy in time. After excitation with lasers of high energy, the electrons thermalize fast to a hot Fermi distribution. Yet, the band occupation numbers are still in strong nonequilibrium as particle exchange between the bands takes longer than energy exchange.

We study excitation of gold with optical photons. Only the 5d and 6sp bands are considered to be active. We present a density-resolved approach coupled with a two-temperature model to follow the nonequilibrium band occupation. A Drude-Lorentz formalism is used to calculate the dielectric function of the heated system. We simulate the temporal evolution of a probe reflectivity up to delay time of 6 ps. Our predictions reveal over time a strong compensation of damping mechanisms for the electrons and phonons. The good agreement between simulation and time-resolved measurements suggests a good understanding of the experimental data and the underlying physical processes, which has been interpreted differently as a lack of dynamics on the subpicosecond timescale.