O 42: Electron-driven Processes

Time: Tuesday 14:00–15:30 Location: H11

O 42.1 Tue 14:00 H11

A simple model of nonadiabatic energy loss during hydrogen scattering from a semiconductor — •Xuexun Lu, Nils Hertl, and Reinhard J. Maurer — University of Warwick, Coventry, UK

Experiments on hydrogen atom scattering from Ge(111) show bimodal energy loss distributions with two peaks. The first peak corresponds to low energy loss and can be quantitatively reproduced with classical molecular dynamics (MD) simulations. The second peak lies at energy losses equivalent to or above the band gap of Ge and arises from electronic transitions between the valence band and the conduction band. Here, we develop a simple and interpretable model for H/Ge(111) scattering to inform the development of new mixed quantum-classical dynamics simulation methods suitable for the description of such nonadiabatic effects in gas-surface scattering. Using density functional theory data and experimental quantities, we parametrize an effective one-dimensional analytical model based on the Newns-Anderson Hamiltonian. The model allows us to study the coupled electron-nuclear dynamics and their conjugate energy transfer using nonadiabatic molecular dynamics methods such as independent electron surface hopping (IESH), molecular dynamics with electronic friction (MDEF), and the Ehrenfest method. In particular, IESH dynamics qualitatively reproduce the nonadiabatic energy transfer channel observed in the experiment.

O 42.2 Tue 14:15 H11

Shaping Polarons in Hematite Fe_2O_3: From Creation to Charge Dynamics — •Sreehari Sreekumar[1], Llorenc Albons Caldentey[1], Jesus Redondo Redondo[1], Aji Alexander[1], Sarah Tobisch[2], Michele Riva[2], and Martin Setvin[1] — [1]Department of Surface and Plasma Science, Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic — [2]Institute of Applied Physics, Vienna University of Technology, Austria

The non-contact Atomic Force Microscopy (nc-AFM) technique has enabled breakthroughs in single-electron charge manipulation [1]. Here we use this capability to study polaron dynamics. Polarons are self-localized electrons or holes in ionic lattices that are crucial to material properties like conductivity, catalysis, and exotic phenomena such as high-temperature superconductivity and colossal magnetoresistance [2]. Polarons are studied in hematite at the single quasiparticle limit, focusing on the fundamental mechanisms involved in their injection, formation, migration, and interaction with defects [3].

1. Gross, L., et al., Science, 2009. 324(5933).
2. Franchini, C., et al., Nature Reviews Materials, 2021. 6(7).
3. Redondo, J., et al., Science Advances, 2024. 10(44).

The work was supported by project MSMT LL2324 *PoTr*

O 42.3 Tue 14:30 H11

Observing the directed motion of a single molecule after dissociation on a surface — •Ilias Gazizullin[1], Matthew Timm[1], Matthias Krinninger[2], Friedrich Esch[2], and Leonhard Grill[1] — [1]Physical Chemistry Department, University of Graz, Austria — [2]Faculty of Chemistry, TU München, Germany

Unidirectional motion of single molecules on surface can be achieved via a rare interplay between the surface and the intramolecular chemical reaction [1]. An alternative approach to achieve controllable molecular motion on surface is to induce dissociation of a molecule, resulting in the recoiling motion of the products [2, 3].

Here, we show how orientation of functional groups of an adsorbed molecule can steer its motion after controlled dissociation of these groups. We study single organic molecules with azido groups adsorbed on Au(111) surface by low temperature scanning tunneling microscopy (STM). The azido group of the adsorbed molecule can be oriented in two possible directions. We applied voltage pulses from the STM tip onto the azido group to induce its dissociation and found that the molecule rotates after dissociation in a specific direction. This directionality clearly depends on the initial orientation of the dissociating azido group, opening new possibilities to induce controlled motion of single molecules on surfaces.

[1] Simpson et al., Nature, 621, 82-87 (2023)
[2] Anggara et al., Sci. Adv., 4 (2018)
[3] Anggara et al., J. Am. Chem. Soc., 138, 7377-7385 (2016)

O 42.4 Tue 14:45 H11

Nonadiabatic quantum dynamics of molecules scattering from metal surfaces — •Riley Preston[1], Yaling Ke[2], Samuel Rudge[1], Nils Hertl[3], Raffaele Borrelli[4], Reinhard Maurer[3], and Michael Thoss[1] — [1]Institute of Physics, University of Freiburg — [2]Department of Chemistry and Applied Biosciences, ETH Zürich — [3]Department of Chemistry and Department of Physics, University of Warwick — [4]DISAFA, University of Torino

Nonadiabatic coupling between electrons and molecular motion at metal surfaces can strongly impact the dynamics of a scattering molecule [1]. We present a theoretical approach based on hierarchical equations of motion (HEOM) [2], which models the scattering of molecules from metal surfaces and incorporates all nonadiabatic and quantum nuclear effects due to the coupling of the molecular degrees of freedom to the electrons in the metal. The approach is exemplified by its application to NO scattering from Au(111), where we observe multi-quantum relaxation of the bond vibrational state due to coupling to electron hole pairs in the surface, in accordance with experiment. The data obtained by the HEOM approach is also used as a rigorous benchmark to assess various mixed quantum-classical methods, from which we derive insights into the validity range of each method [3].

[1] A. M. Wodtke, Chem. Soc. Rev. 45, 3641-3657 (2016).
[2] Y. Ke, R. Borrelli, and M. Thoss, J. Chem. Phys. 156, 194102 (2022).
[3] R. J. Preston, Y. Ke, S. L. Rudge, N. Hertl, R. Borrelli, R. J. Maurer, and M. Thoss, arXiv preprint arXiv:2410.05142 (2024).

O 42.5 Tue 15:00 H11

Nonthermal phonon distributions induced by hot electrons — •Tobias Held, Christopher Seibel, Markus Uehlein, Sebastian T. Weber, and Baerbel Rethfeld — Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau

When an ultrashort laser pulse irradiates a metal, the electrons initially absorb the energy and rapidly establish a hot Fermi distribution. Subsequently, on a picosecond timescale, the electrons transfer energy to the phonon system. Electrons exhibit a stronger coupling to high-wavenumber phonons, leading to nonthermal phonon distributions induced by electron-phonon scattering.

In this study, we use the Boltzmann equation to examine the formation and subsequent relaxation of nonequilibrium phonon distributions. For our model system, we find that the majority of the energy is transferred within 10 ps, while a pronounced phonon nonequilibrium is induced. We observe "hot phonons" at the edge of the Brillouin zone, leading to a collapse of the electron-phonon energy transfer rate. Consequently, a finite temperature difference between electrons and phonons may persist significantly longer than the widely used two-temperature model would predict.

O 42.6 Tue 15:15 H11

Electronic friction simulations of laser-driven hydrogen evolution from copper. Just thermal desorption in a hurry? — •Alexander Spears, Wojciech G Stark, and Reinhard J. Maurer — University of Warwick, Coventry, UK

Ultrafast light pulses can induce energy transfer between light, electrons, and phonons at interfaces, leading to ultrafast dynamics such as light-driven hydrogen evolution from metal surfaces. Whether this energy transfer can drive photocatalysis through selective energy transfer into certain degrees of freedom remains an open question. Molecular dynamics simulations with electronic friction (MDEF) offer a quantum-classical description of electron-phonon coupling and have previously been used to model ultrafast surface dynamics. However, the effect of different electronic friction approximations on the final energy distributions has not been thoroughly investigated. We present MDEF simulations of light-driven hydrogen evolution from different copper surface facets, enabled by machine-learning surrogate models. For various laser fluences, we study desorption probabilities and final state distributions of desorbed molecules. Our results reveal that the choice of electronic friction approximation significantly affects desorption probabilities. However, the magnitude and nature of friction do not seem to affect the final vibrational, rotational, and translational energy distribution of molecular adsorbates. Within the electronic friction approximation, only the shape of the energy landscape determines these properties and no selective energy transfer occurs. This suggests that thermal and laser-driven desorption may yield similar outcomes.