O 47: Poster Electron-driven Processes

Time: Tuesday 18:00–20:00 Location: P2

O 47.1 Tue 18:00 P2

Non-equilibrium electron distribution effects in ultrafast light-driven desorption dynamics — •Matthew Larkin, Henry Snowden, and Reinhard J. Maurer — University of Warwick, Coventry, UK

At metal surfaces, light excitation drives electronic states out of the (thermal) Fermi-Dirac distribution. The resulting non-equilibrium electronic distribution can induce coupled electron-nuclear dynamics that lead to ultrafast structural processes such as light-driven desorption of molecules. As non-equilibrium electron distributions in metals are short-lived, their role in the mechanics of light-driven surface dynamics remains insufficiently understood. In this work, we perform mixed quantum-classical dynamics simulations using a variant of the trajectory surface hopping method where electrons and nuclei are propagated simultaneously and electronic transitions between hundreds of electronic states can be captured. We describe light-generated non-thermal electron distributions as initial conditions to those dynamics. For simple 1D and 2D model systems of atomic and molecular desorption from metal surfaces, we study the role of non-thermal electrons by simulating light-driven desorption probabilities for comparable thermal and non-thermal electronic distributions and discuss the future applicability of this approach for high-dimensional dynamics.

O 47.2 Tue 18:00 P2

Spin effects in ultrafast non-equilibrium dynamics at surfaces — •Ash Baldwin and Reinhard J. Maurer — University of Warwick, Coventry, United Kingdom

Energy, charge, and spin transfer are important factors in modelling ultrafast gas-surface dynamics such as hyperthermal scattering and light-driven dynamics at surfaces. In particular, on metal surfaces, nonadiabatic energy dissipation is an important energy transfer channel in atomic and molecular dynamics at surfaces. In cases of adsorbates with unpaired spin, an additional question arises if and under which conditions the spin moment of the adsorbate survives the scattering event. The challenge lies in capturing the explicit electron correlation in the electronic structure and its effect on the coupled electron-nuclear motion, which mean-field dynamics methods cannot fully capture.

Here, we model the fate of adsorbate spin during scattering from a metal surface using the Newns-Anderson model. We go beyond previous works by simulating the nuclear dynamics beyond mean-field Ehrenfest dynamics, using stochastic trajectory surface hopping methods. Our model is applied to study the spin survival probability of hydrogen atoms on copper and platinum as a function of various model parameters and initial conditions such as the adsorbate kinetic energy and the adsorbate-metal coupling strength to identify the regime in which a finite spin survival probability can be measured.

O 47.3 Tue 18:00 P2

Adsorbate motors: toward tuning unidirectionality by chemical functionalisation — •Grant J Simpson[1], Mats Persson[2], and Leonhard Grill[1] — [1]University of Graz, Austria — [2]University of Liverpool, UK

The purpose of a molecular motor is to convert external energy into unidirectional motion. Feringa-type molecular motors, in which double-bond isomerisation leads to large geometric changes in the molecule, are known to undergo rapid unidirectional motion in solution but often have a reduced function when adsorbed on a surface. In contrast, recently discovered adsorbate motors [1] do not exhibit large structural changes and thus their motion is efficient and unidirectional when adsorbed on a metal surface. Using a scanning tunnelling microscope (STM), we track such motion at the single-molecule scale. The motion is triggered by an internal proton transfer reaction. We further show that, by altering the local chemical structure, this tautomerisation can be affected and may result in modulation of the unidirectional molecular translation.

[1] G. J. Simpson, M. Persson, L. Grill, Nature, 621, 82-86 (2023)

O 47.4 Tue 18:00 P2

Creation, Displacement, and Dynamics of Individual Phasons on Si(001) — •Gaël Reecht, Michael Horn-von Hoegen, and Manuel Gruber — Universität Duisburg-Essen, Duisburg, Germany

The Si(001) surface, characterized by buckled dimers resulting from dangling bond pairing, exhibits a variety of structural configurations. The energetically favorable c(4x2) reconstruction can be manipulated into the p(2x2) configuration, differing by a glide of adjacent rows. The interface between these two phases is known as a phason [1]. Despite its significance in structural phase transitions, the mechanisms driving phason propagation remain poorly understood.

In this study, we employ low-temperature scanning tunneling microscopy to explore the diffusion of individual phasons under tip-induced excitation. We systematically examine the effects of tunneling current, bias voltage, and electric field on the formation and displacement of these structural defects. Phason motion below the STM tip results in stochastic jumps in tunneling current, which we analyze through time-resolved measurements. These findings shed light on the displacement dynamics of phasons and enhance our understanding of the underlying processes. We gratefully acknowledge funding from the CRC1242. [1] Y. Pennec et al., Phys. Rev. Lett., 96, 026102 (2006).

O 47.5 Tue 18:00 P2

Triplet exciton dynamics at the tetracene/a-Si/c-Si(111) interface — •Marvin Krenz[1,2], Simone Sanna[1], Wolf Gero Schmidt[2], and Uwe Gerstmann[2] — [1]Institut für Theoretische Physik, Justus-Liebig Universität Gießen, 35392 Gießen, Germany — [2]Lehrstuhl für Theoretische Materialphysik, Universität Paderborn, 33095 Paderborn, Germany

Exciton transfer is highly relevant for many physical, chemical, and biological processes. The transfer of triplet excitons across the tetracene-silicon interface in sensitized solar cells is a recent example in this context [1]: The excitons, generated by singlet fission in tetracene (Tc), promise to increase the cell efficiency from the single-junction limit of 29% to values as high as 35% [2]. In a previous work, we showed the significance of dangling bond (db) defects for the triplet transfer [3] at a model interface, where tetracene was bonded to an ideal c-Si(111) surface. Here, we present ab initio calculations for a more realistic structure, with a thin film of amorphous silicon (a-Si:H) in-between, resulting in a Tc/a-Si/c-Si model structure. We find that dbs are vital for the exciton transfer also in this case. Dbs at the a-Si/c-Si interface or in the a-Si interlayer itself affect the exciton transfer differently. Generally, it is found that the a-Si layer is energetically favourable for excitation transfer. However, the exciton electron tends to be trapped in a-Si conduction band tail states. Thus, the exciton transfer requires an external field to be completed. [1] M. Einzinger et al. Nature 571, 90*94 (2019). [2] A. Rao et al., Nat. Rev. Mater. 2, 17063 (2017). [3] M. Krenz et al., Phys. Rev. Lett. 132, 076201 (2024)