O 27: Electron-Driven Processes

Time: Tuesday 10:30–11:45 Location: H6

O 27.1 Tue 10:30 H6

Surprisingly fast adsorbate excitation: CO/Ru(0001) probed with X-ray absorption spectroscopy — •Elias Diesen[1], Hsin-Yi Wang[2], Johannes Voss[1], Alan C. Luntz[1], and Anders Nilsson[2] — [1]SUNCAT Center for Interface Science and Catalysis, SLAC National Accelerator Laboratory, Menlo Park, California, USA — [2]Stockholm University, Sweden

Energy transfer between substrate excitations and adsorbate motion plays an essential role in determining chemical reactivity and selectivity on surfaces. Femtosecond pump-probe experiments, using an X-ray probe pulse from a free electron laser, give unique insights into such processes due to the short pulse duration and species-selective probing. We measure the time evolution of the C K-edge X-ray absorption spectrum from CO/Ru(0001) after excitation by a femtosecond high-intensity optical laser pulse, and use detailed spectrum simulations to distinguish excitations of different modes [1]. We find high excitation of the CO internal stretch and frustrated rotation modes within 200 fs of laser excitation - one order of magnitude faster than theoretical predictions. Consequences for our understanding of ultrafast adsorbate excitation will be discussed.

[1] Diesen et al., Phys. Rev. Lett. 127, 016802 (2021)

O 27.2 Tue 10:45 H6

Are vacancies in field ion microscopy artefacts? A DFT study — •Shyam Katnagallu, Jörg Nuegebauer, and Christoph Freysoldt — Department of computational materials design, Max Planck Institut für Eisenforschung GmbH, Max-Planck- Str. 1, 40237, Düsseldorf, Germany.

Resolving the atomic structure of engineering materials in 3D continues to be an extensive research field. Field ion microscopy under evaporating conditions (3D-FIM) is one of the few techniques capable of delivering such atomic-scale information, allowing to even image vacancies and their interactions with solute atoms in alloys. However, the quantification of the observed vacancies and their origins are still a matter of debate. It was suggested that high electric fields (1-10 V/Å) used in FIM could introduce artefact vacancies. To investigate the possibility of this mechanism, we used density functional theory (DFT) simulations. Stepped Ni surfaces with kinks were modelled in the repeated slab approach with a (971) surface orientation. A field of up to 4 V/Å was introduced on one side of the slab using the generalized dipole correction. Contrary to conventional wisdom, we show that the reaction barrier to form vacancies on the electrified metal surface increases compared to the field-free case. We also find that the electric field can introduce kinetic barriers to a potential "vacancy-killing" mechanism. We compare these findings with field evaporation models proposed in the literature.

O 27.3 Tue 11:00 H6

Modeling electron beam damage in Gold nanoparticles and MoS2 — •Cuauhtemoc Nuñez Valencia[1], Matthew Helmi Leth Larsen[1], William B. Lomholdt[2], Pei Liu[2], Daniel Kelly[2], Thomas W. Hansen[2], and Jakob Schiøtz[1] — [1]DTU Physics, Technical University of Denmark, Kgs. Lyngby, Denmark — [2]DTU Nanolab, Technical University of Denmark, Kgs. Lyngby, Denmark

Beam damage in High-Resolution Transmission Electron Microscopy (HRTEM) is poorly understood theoretically, and not yet well described phenomenologically. The interaction between the specimen and the electron beam changes significantly depending on the material: semiconductors (like MoS2) and metals (like Gold) have different lifetime excitations and beam damage can be triggered in different mechanism.

In this study, we investigate the beam damage in MoS2 and Gold nanoparticles simulating the macroscopic timescale with a Kinetic Monte Carlo like algorithm, using different approaches: long Molecular Dynamics (MD) simulations for Gold and statistical model using different threshold energy (T_D) values for MoS2 considering the thermal vibrations.

The second approach for modeling the beam damage in MoS2 was through ab Initio Molecular dynamics the calculation of the threshold energies for the different scenarios. Where a Kinetic Monte Carlo simulation was implemented for study the dynamics of the defect production and the beam damage for MoS2.Also, we studied the effect in the T_D when we remove or add electrons to the MoS2.

O 27.4 Tue 11:15 H6

Electron-driven mobility of the hydrogen-bonded ammonia clusters — •prashant srivastava[1], Daniel Miller[2], and Karina Morgenstern[3] — [1]Chair of physical chemsitry-I, Ruhr University Bochum, Germany — [2]Department of Chemistry, Hofstra University, 106 Berliner Hall, Hempstead, New York 11549, United States — [3]Chair of physical chemsitry-I, Ruhr University Bochum, Germany

Electron-driven processes in polar solvents are of great interest to study, e.g., electron solvation in ammonia plays a vital role in ozone layer depletion. We study the impact of electrons on the ammonia clusters adsorbed on a copper surface using a combination of low-temperature scanning tunneling microscopy, femtosecond laser pulses, and ab-initio calculations. Photo-injected electrons from the copper surface lead to diffusion or desorption of the second layer of the clusters. Upward mass transport (UMT) and downward mass transport (DMT) also play an important role in modifying the hydrogen-bonded network of these clusters. Theoretical calculations confirm electron solvation into the second layer. We present a molecular-scale insight into the interactions of photo-injected electrons with the ammonia clusters in the second layer. Our results show that this interaction can modify an ammonia cluster and enhance its mobility.

O 27.5 Tue 11:30 H6

Distribution of Charge and Lattice Defects via Machine Learning. — Viktor Birschitzky[1], Michael Prezzi[1], Marco Corrias[1], Lorenzo Papa[1], Igor Sokolovic[2], Alexander Gorfer[1], Martin Setvin[2,3], Michael Schmid[2], Ulrike Diebold[2], Cesare Franchini[1,4], and •Michele Reticcioli[1] — [1]University of Vienna (Austria) — [2]Institute of Applied Physics, TU Wien (Austria) — [3]Charles University, Prague (Czech Republic) — [4]University of Bologna (Italy)

Lattice defects and localized charge on oxide surfaces impact the properties of the material to a different degree depending on their spatial distribution. However, the high number of possible defect configurations poses practical challenges to first-principles studies. Here, we propose a machine-learning-accelerated approach to explore in the framework of density functional theory the spatial configurations of charge and lattice point defects. We apply this approach to analyze the distribution of surface oxygen vacancies on rutile TiO_2(110). The attractive interaction with small polarons (electrons localized on the Ti atoms) are revealed to weaken the repulsion between oxygen vacancies, favoring particular arrangements of the vacancies. The resulting distribution can be compared with the patterns identified by computer vision algorithms on scanning-probe microscopy images.