O 86: Surface dynamics

Time: Thursday 15:00–17:45 Location: HSZ/0403

Invited Talk O 86.1 Thu 15:00 HSZ/0403

Ultrafast Dynamics at Surfaces with Machine Learning Surrogates — •Reinhard Maurer — University of Vienna, AT — University of Goettingen, DE — University of Warwick, UK

Ultrafast dynamics at surfaces (driven by light, electrons, or hyperthermal scattering) involve the concerted motion of electrons and atoms at comparable energy and time scales, giving rise to nonadiabatic effects. Excited electrons drive chemical conversions, induce phase transitions, and mediate energy transfer between adsorbates and surfaces. To reliably predict such effects with scalable, state-of-the-art nonadiabatic dynamics simulations requires the use of accurate and data-efficient high-dimensional machine learning (ML) surrogate models. This includes representations of energy landscapes, but also nonadiabatic couplings or excited-state properties that are required for nonadiabatic simulations. I will present recent strategies to construct high-dimensional ML surrogate models of electronic structure, including active learning and fine-tuning of foundation models that allow us to reduce the required electronic structure data to a few hundred data points per gas-surface dynamics model and even transfer learn across density functional approximations. Electronic properties such as electron-phonon coupling tensors or electronic Hamiltonians can be efficiently represented by encoding physical equivariance properties in the model. I will showcase the utility of the introduced models with recent dynamics applications on reactive molecular scattering and light-driven structural dynamics.

O 86.2 Thu 15:30 HSZ/0403

Ultrafast Structural Dynamics of Layered Material Surfaces — •Alp Akbiyik[1,2], Felix Kurtz[1,2], Hannes Böckmann[1,2], and Claus Ropers[1,2] — [1]Department of Ultrafast Dynamics, Max Planck Institute for Multidisciplinary Sciences, D-37077 Göttingen, Germany — [2]University of Göttingen, 4th Physical Institute, D-37077 Göttingen, Germany

Layered materials are promising candidates for studying and applying correlated phenomena, and their carrier dynamics have been extensively investigated, including through ultrafast pump-probe techniques. However, the structural evolution of their surface layers remains poorly accessible. Ultrafast low-energy electron diffraction (ULEED) leverages the surface sensitivity of low-energy electron pulses, with high spatial resolution and temporal resolution down to 1-2 ps. This allows us to monitor the momentum-dependent out-of-plane phonon populations across the Brillouin zone as well as changes in diffraction peak intensities [1,2]. Here, we contrast the phonon equilibration behaviour of transition metal dichalcogenides (TMDCs), such as WTe2, with graphite following electron-phonon coupling.

[1] G. Storeck et al., Structural Dynamics 7, 034304, 2020.

[2] F. Kurtz et al., Nat. Mater. 23, 890-897, 2024.

O 86.3 Thu 15:45 HSZ/0403

Nonadiabatic Energy Loss in Hyperthermal Gas-Surface Scattering: Going Beyond the Mean Field Approximation — •Ash Baldwin[1] and Reinhard J. Maurer[1,2] — [1]University of Vienna, Vienna, Austria — [2]University of Warwick, Coventry, United Kingdom

Nonadiabatic energy dissipation is known to play a crucial role in hyperthermal scattering at surfaces, leading to electron-hole pair excitation and highly inelastic scattering. For projectiles with unpaired spins, such as hydrogen atoms, hybridisation with the metal surface results in strong non-adiabatic effects driven by a phase transition in its spin-polarisation. It is known that traditional mean-field descriptions of the resultant nonadiabatic dynamics using methods like MDEF and Ehrenfest dynamics break down in the presence of the transition, leading to divergent energy transfer rates and unphysical stopping powers at the location of the spin transition [Box et al. *J. Phys. Chem. Lett.* 15, 51 (2024); Lindenblatt et al. *Phys. Rev. Lett.* 97 (2006)].

Here, we model H/Cu(111) and H/Ag(111) scattering in the presence of on-site correlation in the adsorbate state using the Newns-Anderson Hamiltonian. We go beyond previous works by simulating the dynamics using stochastic trajectory surface hopping methods that capture the coupling between the nuclear and electronic degrees of freedom beyond the mean-field. By systematically classifying scattering regimes, we assess the applicability of mean-field and beyond mean-field methods for predicting nonadiabatic energy loss.

O 86.4 Thu 16:00 HSZ/0403

Manipulation of charge density wave order in 1H-TaS_2 to simulate broken lattice degeneracy of misfit layered chalcogenides — •Laura Pätzold[1], Sandra Sajan[2], Clara Pfister[1,3], Haojie Guo[2], Miguel M. Ugeda[2], and Tim O. Wehling[1,3] — [1]U Hamburg — [2]DIPC San Sebastian — [3]The Hamburg Centre for Ultrafast Imaging

Misfit layer compounds, a class of van der Waals heterostructures comprised of noncommensurate layers, are ideal platforms to explore the influence of broken lattice symmetries and incommensurability on collective electronic phases, such as charge density waves (CDWs). The simulation of these symmetry-broken ground states poses a computational challenge due to large numbers of inequivalent atoms. By employing an electron-lattice downfolding scheme [1] followed by molecular dynamics (MD) simulations, we obtain the (thermo-)dynamics of these systems with the accuracy of full first-principles calculations while reducing the computational cost by several orders of magnitude. Here, we explore CDW control parameters to analyze the influence of the PbS layer in the misfit layer compound $(PbS)_{1.13}TaS_2$, where a shift in CDW order accompanied by symmetry breaking is measured in the 1H-TaS_2 layer. Simulating charge doping, interlayer hybridization effects via external potentials and frustration effects via the supercell size, we show that only a combination of these effects reproduces the characteristics of the measured structure factor while being thermodynamically stable.

[1] A. Schobert et al., *SciPost Phys.* **16**, 046 (2024)

O 86.5 Thu 16:15 HSZ/0403

Control and thermal evolution of commensurate charge-density wave order in 1H-$NbSe_2$ — •Clara Pfister[1,2], Alexander Bäder[3,4], Laura Pätzold[1], Tobias Wichmann[3], Felix Lüpke[3,4], Mariana Rossi[5], and Tim O. Wehling[1,2] — [1]I. Institut für Theoretische Physik, Universität Hamburg — [2]The Hamburg Centre for Ultrafast Imaging — [3]Peter-Grünberg-Institut, Forschungszentrum Jülich — [4]II. Physikalisches Institut, Universität zu Köln — [5]MPI für Struktur und Dynamik der Materie, Hamburg

Charge-density waves (CDWs) are symmetry-broken ground states arising from electron-lattice interactions and offer a platform to study tunable ordered phases in quasi two-dimensional materials. Using *ab initio* calculations and a downfolding approach [1], we investigate how biaxial tensile strain and electron doping affect CDW order in 1H-$NbSe_2$. In both cases, a transition from the native 3×3 to a competing 2×2 CDW ground state is induced. The strain-induced 2×2 CDW is supported by experimental data of monolayer 1H-$NbSe_2$ flakes that are rotated with respect to the underlying bulk $NbSe_2$. By constructing phase diagrams, we are able to identify the strain- and doping-induced boundaries between the competing CDW ground states. Moreover, by performing molecular dynamics (MD) simulations, we find that strain-induced 2×2 order persists up to $T_{\mathrm{CDW}} \approx 10$ K, whereas the doping-induced 2×2 CDW is thermally much less stable. Our MD results furthermore suggest that nuclear entropy is the driving mechanism in the melting process of the CDW.

[1] A. Schobert et al., *SciPost Phys.* **16**, 046 (2024)

O 86.6 Thu 16:30 HSZ/0403

Surface Resonant Raman Scattering of Ag(110) surfaces — Sandhya Chandola[1,2], •Norbert Esser[1], Mohamed Yassine Fatihi[3], and Simone Sanna[3] — [1]Institut für Festkörperphysik,TU Berlin, Hardenbergstr. 36, 10623 Berlin, Germany — [2]Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Schwarzschildstr. 8, 12489 Berlin, Germany — [3]Institut für Theoretische Physik, Heinrich-Buff-Ring 16, 35392 Gießen, Germany

The phonon and electron scattering mechanism on Ag(110) surfaces is investigated using Surface Resonant Raman scattering (SRRS) and Reflectance Anisotropy Spectroscopy (RAS) in ultra-high vacuum (UHV). The surface electronic transition is clearly visible with RAS, with Raman spectra showing surface phonons near the resonant excitation. A low energy peak is also seen, which should be related to the Rayleigh mode at the Brillouin zone edge. This shows that the surface is rough, and that the intensity of this mode depends on the sample treatment. Consequently, RAS together with Raman spectroscopy allows optimization of surface reconstruction and control of surface morphology. By means of first-principles calculations within density functional theory, we demonstrate that the dominant Raman signature is coupled to an electronic transition between localized surface-states (calculated at 1.6 eV). The corresponding surface-localized phonon mode enhances the distance between the topmost surface layers. The combined experimental and theoretical approach to resonant surface Raman spectroscopy establishes a powerful tool for investigating surface vibrational dynamics and electron-phonon interactions.

O 86.7 Thu 16:45 HSZ/0403

Spatiotemporal control of Peierls heterostructures by valley-selective photoswitching — •Hannes Böckmann[1], Jan Gerrit Horstmann[3], Felix Kurtz[1], Manuel Buriks[4], Karun Gadge[4], Salvatore Manmana[4], Stefan Wippermann[5], and Claus Ropers[1,2] — [1]Max Planck Institute for Multidisciplinary Sciences, Göttingen, Germany — [2]4th Physical Institute, Solids and Nanostructures, University of Göttingen, Germany — [3]Department of Materials, ETH Zurich, Zürich, Switzerland — [4]Institute for Theoretical Physics, University of Göttingen, Germany — [5]Faculty of Physics, Philipps University of Marburg, Germany

Phase competition and coexistence are hallmarks of strongly-correlated materials, promising tunable functionality on the nanoscale. Light allows for tilting the balance between distinct correlated states and phases with a prominent example given by the optical switching of an insulator to a metal. However, optical excitation generally lacks the specificity to select sub-wavelength domains. In this work, we employ valley-selective photodoping of correlated electronic states to drive the domain-specific quench of a textured Peierls insulator. Specifically, we exploit the anisotropic absorption of nanowire domains by tuning both the photon energy and the polarization to the transition matrix elements most strongly coupled to the structural transformation. We find that averting dissipation facilitates domain-specific carrier confinement, control over nanotextured phases, and a prolonged lifetime of the metastable metallic state.

O 86.8 Thu 17:00 HSZ/0403

Surface resonant Raman scattering from coinage metal surfaces — •Mohamed Yassine Fatihi[1], Sandhya Chandola[2,3], Norbert Esser[2,3], and Simone Sanna[1] — [1]Institut für Theoretische Physik and Center for Materials Research (LaMa), Justus-Liebig-Universität Gießen, 35392 Gießen, Germany — [2]Helmholtz Zentrum Berlin für Materialien und Energie GmbH, 12489 Berlin, Germany — [3]Institut für Festkörperphysik, TU Berlin, 10623 Berlin, Germany

The vibrational characterization of clean metal surfaces has remained largely inaccessible to optical spectroscopy, as Raman scattering from pristine metals is extremely weak and dominated by bulk phonon backgrounds. However, the first direct Raman observation of surface phonons on a Cu(110) single crystal has been recently reported[1]. We explain this Raman activity and predict it for the related Ag(110) surface from first principles, demonstrating by means of atomistic models how tuning the incident photon energy to the surface-state transition induces a striking resonant enhancement of the surface phonon signal. This mechanism selectively amplifies the surface response with respect to the bulk contribution. Our resonant surface phonon Raman approach establishes a powerful probe for investigating surface vibrational dynamics and electron-phonon interactions, creating new opportunities for real-time studies of metallic surfaces under diverse conditions, from ultra-high vacuum to catalytic and electrochemical environments

[1] Denk, M., et al. Phys. Rev. Lett. 128, 216101 (2022).

O 86.9 Thu 17:15 HSZ/0403

Manipulation of Optical Phonons by Thermal Expansion Mismatch in Bi/Si(111) Heterostructures — •Fabian Thiemann, Maja Penkl, and Michael Horn-von Hoegen — University of Duisburg-Essen, Lotharstr. 1, 47057 Duisburg, Germany

We investigate how substrate-induced biaxial strain controls the temperature behaviour of the A_{1g} phonon frequency $\Omega(T)$ in epitaxial Bi(111) films on Si(111) in the time domain. Bismuth's anisotropic A7 structure, strong electron-phonon coupling and its Peierls' distortion make the out-of-plane c-axis vibrations highly sensitive to strain. Using *in situ* SPA-LEED and broadband femtosecond transient reflectivity on the same samples, we measured temperature-dependent lattice parameters and coherent-phonon frequencies. Strained Bi films on Si(111) exhibit a larger thermal redshift $\partial\Omega/\partial T$ compared to relaxed bulk or polycrystalline Bi. Applying an extended phonon-shift model reveals that the anharmonic multi-phonon term depends solely on the value of the c-axis lattice parameter. This identifies c-axis expansion as the key structural factor governing the A_{1g} phonon, enabling targeted strain-engineering of phonon properties in bismuth and related anisotropic materials.

O 86.10 Thu 17:30 HSZ/0403

Nested Sampling Simulations of Oxygen Induced Surface Reconstructions on Cu(100) — •Felix Riccius[1,2], Hendrik H. Heenen[1], Jutta Rogal[2], and Karsten Reuter[1] — [1]Fritz-Haber-Institut der MPG , Berlin — [2]Flatiron Institute, New York

Under operating conditions, catalysts can undergo structural, compositional, and morphological changes, which ultimately dictate their activity and stability. However, extensive sampling of surface configurations at finite temperature and pressure is computationally expensive and non-trivial. To this end, nested sampling simulations can be used to explore the full configuration space, estimate the partition function, and directly access the desired thermodynamic ensemble averages. We here render such simulations tractable by using them in combination with a machine-learning interatomic potential, optimize the code to run efficiently on GPUs, and tailor system specific nested sampling moves. Applied to the early oxidation of Cu(100), the approach correctly captures clean surface configurations at various oxygen coverages and the relative stability of the O adsorption sites as a function of temperature. Moreover, nested sampling successfully predicts the experimentally observed and highly complex $(2\sqrt{2}\times\sqrt{2})$ R45°-O missing-row surface reconstruction and their defect states without any human bias.