O 40: Surface Dynamics

Time: Tuesday 14:00–15:30 Location: H6

O 40.1 Tue 14:00 H6

Manipulation of Optical Phonons in Strained Bi/Si(001) Heterostructures — •Fabian Thiemann and Michael Horn-von Hoegen — University of Duisburg-Essen, Lotharstr. 1, 47057 Duisburg, Germany

Intentionally manipulating phononic properties in materials can cause dramatic changes in their dynamic behavior. A system that responds especially strong to structural changes is Bi due to its strong electron-phonon coupling and its inherent Peierls distortion. This renders the optical A_{1g} mode – oscillating along the [111] direction – sensitive to changes in the electronic system and coherently excitable upon photoexcitation. When grown on Si(001) bismuths lattice matches in the $[1\bar{1}0]$ direction, resulting in a compressive strain in the [110] direction, whereas the films are [111] oriented. The strain is relieved rapidly while increasing the thickness from 10 to 16 BL. This is predicted to influence the Peierls distortion heavily. In this work we extensively studied the impact of strain and confinement on the photoexcited coherent A_{1g} mode's frequency and dephasing in Bi/Si(001) heterostructures by in-situ transient-reflectivity spectroscopy. We observe a dramatic blueshift up to 0.2 THz, that can be preciesely tuned by the film thickness. The frequency however does not follow exactly the strain parameter and behaves differently in two regiemes. We attribute this discrepancy to the confinement in the [111] direction and the influence of the strongly shifting electron surface state.

O 40.2 Tue 14:15 H6

Coherent modulation of the charge density wave gap in 1T-TiSe$_2$ probed by tr-ARPES — •Jan Böhnke, Stephan Schmutzler, Mehul Jotshi, Cornelius Gahl, and Martin Weinelt — Freie Universität Berlin, Fachbereich Physik, Germany

Tuning the charge density wave (CDW) gap in 1T-TiSe$_2$ allows for the de/stabilization of the phase transition. An optical excitation breaks long-range order of the CDW state and results in a semi-metallic state on an ultrafast timescale as free carriers near the Fermi-level enhance Coulomb screening [1,2]. By employing time-resolved ARPES with 1.55 eV pump and 6.2 eV probe pulses, we discover for the first time fluence-dependent coherent modulations of the charge-density wave gap (closing and opening) in 1T-TiSe$_2$ at the Brillouin zone center. While for low excitation fluences, we mainly observe signatures of the CDW connected A^*_{1g} mode (3.45 THz), the optical A_{1g} phonon mode (6.1 THz) modulates the electronic structure near the gap for the high fluence regime. For an intermediate fluence we detect the transition from the A^*_{1g} to the A_{1g} mode. As we additionally find three image-potential states on the surface of 1T-TiSe$_2$, we can confirm the samples' low defect density at the surface after in-vacuum cleavage.

[1] T. Rohwer et al., Nature 471, 490–493 (2011)
[2] M. Huber et al., Sci. Adv. 10, eadl4481(2024)

O 40.3 Tue 14:30 H6

Impact of Coherent Phonons on Time-Resolved Optical Properties of WTe$_2$ — •Francesco Sammartino[1], Manuel Tuniz[1], Wibke Bronsch[2], Fulvio Parmigiani[1,2], and Federico Cilento[2] — [1]Università degli Studi di Trieste — [2]Elettra - Sinctrotrone Trieste

We study the ultrafast dynamics of tungsten ditelluride (WTe$_2$) along both its in-plane axes, by time-resolved reflectivity (TR-R) and time-resolved second-harmonic generation (TR-SHG) experiments with a varying pump fluence.

We identify two phonon modes: the shear phonon mode at 0.24 THz, indicative of uniform in-plane atomic shifts and detected in both TR-R and TR-SHG signals, and one at 2.4 THz, detected solely in the TR-R signal. We observe a large, fluence-dependent shift of up to $\approx 90^\circ$ in the initial phase of the shear mode coherent oscillation, obtained in a narrow fluence range.

This evidence suggests that the excitation density can be used as a powerful knob to control the initial phase of the atomic displacements in a layered material.

O 40.4 Tue 14:45 H6

Controlled formation of thermodynamically unaccessible surface structures — •Simon B. Hollweger, Anna Werkovits, and Oliver T. Hofmann — Institute of Solid State Physics, Graz University of Technology, Austria

It is well known that organic molecules adsorbed on surfaces can form a variety of different surface structures. Which structure is the most favored one at a given temperature and pressure is determined by thermodynamics. However, in this study, we propose the idea of a mechanism with which we can control the formation of a specific metastable surface polymorph that can not be reached thermodynamically. With targeted temperature and pressure changes, a rearrangement process of the adsorbed molecules out of thermodynamic equilibrium is triggered. For specifically designed systems, this rearrangement of the adsorbed molecules leads to a kinetically trapped metastable surface polymorph. As a proof of principle for this proposed mechanism, kinetic Monte Carlo simulations of planar molecules adsorbing on a square lattice are conducted. We show that a metastable upright-standing structure of the planar molecules can be reached with a specific temperature and pressure profile applied to the system.

O 40.5 Tue 15:00 H6

Out of the Crystalline Comfort Zone: Sampling the Initial Oxide Formation at Cu(111) — •Felix Riccius, Nicolas Bergmann, Hendrik H. Heenen, and Karsten Reuter — Fritz-Haber-Institut der MPG, Berlin, Germany

The oxidation of transition metal surfaces is widely recognized as a complex process that still bears many open questions, specifically at the microscopic level. Atomistic simulations could potentially uncover crucial insights, but state-of-the-art approaches are predominantly guided by human chemical intuition, leading to highly idealized surface representations. Here we demonstrate a systematic approach to model high-quality surface-phase diagrams, using the early oxidation of the Cu(111) surface as an example. To this end, we train a MACE machine learning interatomic potential (MLIP) to density functional theory calculations and combine its fast and accurate energetics with replica exchange molecular dynamics. We extensively explore the vast, thermodynamically relevant phase space and further develop surface phase diagrams based on increasingly involved theoretical frameworks. Our approach yields a comprehensive structural ensemble that predicts early Cu(111) oxidation to be characterized by O-Cu-O ring patterns, bearing significant disorder. Within the computed surface evolution, we recover trends in O-Cu-O ring distribution as a function of reaction conditions in line with scanning tunneling microscopy data. Our study illustrates how MLIPs and extensive sampling can be leveraged to rationalize metal surface oxidation fully *in silico* without the need to rely on experimental guidance.

O 40.6 Tue 15:15 H6

Photo-induced carrier and structural dynamics in anatase TiO$_2$ nanosheets — •Zhipeng Huang[1,2], Yan Yan[2], Xinxin Cheng[2,3], R. J. Dwayne Miller[4], and R. Kramer Campen[1] — [1]Faculty of Physics and Center for Nanointegration (CENIDE), University of Duisburg-Essen — [2]Max Planck Institute for the Structure and Dynamics of Matter — [3]SLAC National Accelerator Laboratory — [4]Departments of Chemistry and Physics, University of Toronto

Ultrathin anatase TiO$_2$ nanosheets with {001} facets have significantly higher activity for light-induced H$_2$ evolution than other TiO$_2$-based materials. The mechanism of this enhancement is not understood. Gaining such insight requires understanding the dynamics of charge carriers and their interactions with lattices after optical excitation. Here we characterize structural and charge carrier dynamics in these materials following UV excitation using ultrafast electron diffraction and transient absorption spectroscopy.

We observed an ultrafast lattice expansion and distortion in the nanosheets, occurring earlier than the Debye-Waller effect. Density Functional Theory (DFT) calculations suggest that the lattice expansion and distortion are induced by the trapping of charge carriers and the formation of large electron polarons. Compared to TiO$_2$ nanoparticles, the nanosheets exhibit significantly higher polaron populations, which explains their enhanced photocatalytic properties.