

O 93: Focus Session: Structural Dynamics in Nanoscale Materials, Probed by Ultrafast Electron Pulses I

Time: Thursday 10:30–13:00

Location: HE 101

Invited Talk

O 93.1 Thu 10:30 HE 101

Ultrafast Electron Diffuse Scattering: Mapping Momentum Dependent Electron-Phonon Coupling and Nonequilibrium Phonon Dynamics in 2D Materials — ●BRADLEY SIWICK^{1,2}, MARTIN OTTO¹, LAURENT RENE DE COTRET¹, MARK STERN¹, and MARK SUTTON¹ — ¹McGill University, Department of Physics, Center for the Physics of Materials, Montreal, Canada — ²McGill University, Department of Chemistry, Montreal, Canada

The nature of the couplings within and between lattice and charge degrees of freedom is central to condensed matter and materials physics. These interactions are determining to phenomena as diverse as superconductivity, charge density waves and carrier mobility in semiconductors and metals. Despite their fundamental role, detailed momentum-dependent information on the strength of electron-phonon and phonon-phonon coupling across the entire Brillouin zone has proved elusive. This talk will describe a new technique, ultrafast electron diffuse scattering (UEDS), that can provide such information. Specific applications of UEDS to 2D materials will be presented.

Invited Talk

O 93.2 Thu 11:00 HE 101

Beyond Debye-Waller Effects in Ultrafast Electron Diffraction — ●XIJIE WANG — SLAC National accelerator Laboratory, Menlo Park, CA 94025, USA

Transient Debye Waller-effects has been used extensively for studying energy relaxation and lattice heating in laser excited materials in time-resolved X-ray or electron diffraction. In this talk, I will discuss the latest developments in MeV ultrafast electron diffraction and its applications for structure dynamics. Taking advantage negligible multiple scattering of MeV electrons in the materials, we successfully extended the transient Debye*Waller-effects to study the energy relaxation and transport in nano-scale heterostructure, ripples and energy relaxation in 2-D materials and phonon-phonon coupling in diffuse scattering. I will also discuss multi-modal studies of quantum materials at SLAC MeV UED.

O 93.3 Thu 11:30 HE 101

Ultrafast nanotectonics with nanocrystalline graphene — SILVIO MORGENSTERN, ●ARNE SENFTLEBEN, MARLENE ADRIAN, CHRISTIAN GERBIG, and THOMAS BAUMERT — Institut für Physik und CIN-SaT, Universität Kassel, Heinrich-Plett-Straße 40, 34132 Kassel

Nanocrystalline graphene consists of nanometer-sized plates with a perfect graphene structure, which are coupled to each other via lattice defects containing sp^3 hybridized carbon atoms. When heated, the plates expand and the sp^3 hybrids act as a hinge. The result is a nano-scale movement that recalls the formation of wrinkled mountains. Here we report on expansion time scales in the 100 fs regime followed by out of plane dynamics on the sub ps time scale on a free standing nanocrystalline graphene sheets with the help of our high resolution ultrafast electron diffraction apparatus.

O 93.4 Thu 11:45 HE 101

Molecular dynamics simulations of a femtosecond laser excitation in antimony — ●BERND BAUERHENNE^{1,2}, FELIPE VALENCIA³, and MARTIN E. GARCIA^{1,2} — ¹Theoretische Physik - Universität Kassel, Heinrich-Plett-Str. 40, D-34132 Kassel, Germany — ²Center for Interdisciplinary Nanostructure Science and Technology (CINSA-T), Heinrich-Plett-Strasse 40, D-34132 Kassel, Germany — ³Physics Department, Universidad Nacional de Colombia, Edificio 404, Ciudad Universitaria, Bogota, Colombia

The goal of this work is to accurately simulate laser induced ultrafast, nonthermal solid-to-solid and solid-to-liquid phase transitions in Sb. In particular we are interested in elucidate the dynamics of the ultrafast transition from the A7 to the simple cubic structure in Sb, and also to investigate how fast nonthermal melting of Sb can occur for extremely high laser fluences. For this purpose, we run ab-initio molecular dynamic simulations of laser excited antimony in thin-film geometry (containing around 400 atoms) using our in house density functional theory code CHIVES. Based on these simulations and in order to allow for a closer comparison with experiments, we developed an analytical interatomic potential describing laser excited Sb. In ad-

dition, we calculated the laser-fluence dependent electron-phonon coupling constant in Sb from ab-initio calculations. Using our potential and including incoherent electron-phonon scattering, we simulated the intense femtosecond laser excitation of antimony on large supercells.

O 93.5 Thu 12:00 HE 101

Electron-Phonon coupling in thin Pb films on Si(111) from the lattice's perspective — ●TOBIAS WITTE, TIM FRIGGE, BERND HAFKE, BORIS KRENZER, and MICHAEL HORN-VON HOEGEN — Universität Duisburg-Essen, 47048 Duisburg, Germany

Employing time resolved reflection high energy electron diffraction (tr-RHEED) the non-equilibrium dynamics of the phonon system after optical excitation of ultrathin Pb films and islands on Si(111) has been investigated. The sample is pumped by fs-IR-laserpulses, while the transient response is probed by 30 keV electron pulses. Surface sensitivity is achieved through grazing incidence of the high energy electrons. fs-laserpulses cause impulsive excitation of the electron system of the Pb films and islands, respectively. The hot electron system thermalizes and heats the lattice system by electron-phonon coupling. The thermal motion of the (surface-) atoms is observed through the transient Debye-Waller effect. As Pb is known to exhibit strong electron-phonon coupling in the bulk the time constant for the energy transfer to the lattice system is - in the framework of the two temperature model - expected to be in the sub-ps regime. In contrast, a rather slow time constant of $\tau = 3.2$ ps was observed at the surface. Moreover, incoherent lattice excitation of the islands occurs even slower at a time constant of $\tau = 4.6$ ps. These results demonstrate how low-dimensionality and quantum-size effects reduce the energy flow mediated by electron-phonon coupling.

O 93.6 Thu 12:15 HE 101

Efficient simulation scheme for propagation of energetic electrons in solids. — ●BEATA ZIAJA-MOTYKA — CFEL, DESY Hamburg, Germany — INP PAS Krakow, Poland

Intense X-rays pulses, while irradiating solids, excite energetic photoelectrons. In case of inner-shell photoionization, frequent with hard X-rays, the core holes left after the photoionization relax predominantly through Auger decays, accompanied by emission of further electrons. Both photo- and Auger electrons ionize the sample through impact ionization, releasing further electrons. Those collisional processes then trigger secondary electron cascades which significantly contribute to the progressing damage of the sample.

Here we report on a dedicated Monte Carlo code able to accurately follow spatial and temporal evolution of electron cascades in solids under low-fluence X-ray pulse irradiation (implying moderate density of excited electrons). This computationally efficient scheme uses an 'independent cascade' approximation. We discuss an extension of the code to simulate propagation of excited electrons in a bulk material under intense X-ray irradiation - preserving its high computational efficiency. If successful, the tool can be applied for radiation damage studies of X-ray diffraction as well as of electron diffraction, as relaxation of a material excited by a highly energetic electron projectile is similar to that after an X-ray excitation.

O 93.7 Thu 12:30 HE 101

Direct Observation of Reaction Dynamics by Femtosecond Electron Diffraction — ●HENRIKE MÜLLER-WERKMEISTER^{1,2}, YIFENG JIANG¹, LAI CHUNG LIU³, and R. J. DWAYNE MILLER^{1,3} — ¹Max Planck Institute for the Structure and Dynamics of Matter, 22607 Hamburg, Germany — ²Institute of Chemistry, Physical Chemistry, University of Potsdam, 14476 Potsdam, Germany — ³Departments of Chemistry and Physics, University of Toronto, Toronto, M5S 3H6, Canada

Femtosecond electron diffraction (FED) is a sensitive tool to directly study molecular dynamics in structural detail. We have studied the photoinduced spin crossover (SCO) in the coordination compound [FeII(PM-AzA)2](NCS)2 (or AZA). The unique capability of FED allows us to directly probe atomic motions and to track ultrafast structural changes within the crystal lattice of AZA. The data and refinement calculations indicate the global structural reorganization within 2.3 ps, as the metal-to-ligand bond distribution narrows dur-

ing intramolecular vibrational energy redistribution (IVR), driving the molecular rearrangement. Three independent dynamical groups are identified to model the structural dynamics upon photoinduced SCO.

Y. Jiang, L. Liu, H. M. Müller-Werkmeister, C. Lu, D. Zhang, R. L. Field, A. Sarracini, G. Moriena, E. Collet, and R. J. D. Miller, *Angew. Chemie* 129, 7236 (2017).

O 93.8 Thu 12:45 HE 101

Ultrafast pathways of the photo-induced insulator-metal transition in an organic molecular crystal — BART SMIT¹, FLORIAN HÜWE², NANCY PAYNE¹, OLUFEMI OLAOYE¹, IRENE BAUER³, JENS PFLAUM², MARKUS SCHWOERER³, and •HEINRICH SCHWOERER^{1,4} — ¹Stellenbosch University, South Africa — ²Universität Würzburg, Deutschland — ³Universität Bayreuth, Deutschland — ⁴Max-Planck-Institut für Struktur und Dynamik der Materie, Hamburg, Deutschland

Low dimensional molecular metals represent an intriguing class of solid functional organic materials due to their tunable electronic, dielectric, magnetic and structural ground states. We report on the collective atomic motion at the insulator-to-metal Peierls transition of the radical ion salt Cu(Me,Br-DCNQI)₂ (DCNQI: Dicyanoquinonediimine), detected by ultrafast electron diffraction. We observe the photo-induced lifting of the insulating phase within 2 ps in the entire crystal volume, which enables high-speed optical sensors of outstanding signal response. We uncover that the phase transition is initiated by specific collective bending mode within the organic molecule, however, only an additional internal pressure relief, manifested by a volume growth of rest groups, allows to optically lock the metallic state for long times. The disclosing of the microscopic pathways to optically drive the structural Peierls transition in Cu(DCNQI)₂ points out the tailored response to external stimuli available in this unique class of complex functional materials.