O 77: Focus Session: Structural Dynamics in Nanoscale Materials Probed by Ultrashort Electron Pulses

Time: Thursday 10:30–13:15 Location: MA 005

Topical Talk O 77.1 Thu 10:30 MA 005

Photoinduced phase transitions in vanadium dioxide revealed by ultrafast electron diffraction and broadband spectroscopy — •Bradley Siwick[1], Vance Morrison[1], Robert Chatelain[1], Kunal Tiwari[1], Ali Hendaoui[2], Andrew Bruhacs[1], and Mohamed Chaker[2] — [1]Departments of Physics and Chemistry, Center for the Physics of Materials, McGill University, Montreal, Canada — [2]Institut National de la Recherche Scientifique, Centre Énergie Matériaux et Télécommunications, Varennes, Canada

The complex interplay between strong electron-electron correlations and structural distortions is thought to determine the electronic properties of many oxides, but the respective role of the two contributions is often extremely difficult to determine. Vanadium dioxide is a particularly notorious example. We will report on combined radio-frequency compressed ultrafast electron diffraction (RF-UED) and infrared transmissivity experiments in which we directly watch and separate the lattice and charge density reorganizations that are associated with the optically induced semiconductor-to-metal transition in vanadium dioxide. These studies have uncovered a previously unreported photoinduced transition to a metastable intermediate state with the periodic lattice distortion characteristic of the insulator intact, but differing by a 1D rearrangement of charge density along the octahedrally coordinated vanadium dimer chains and a transition to metal-like mid IR optical properties. The results demonstrate that UED is capable of following details of both lattice and electronic structural dynamics on the ultrafast timescale.

Topical Talk O 77.2 Thu 11:00 MA 005

Spatial and temporal resolution studies on a highly compact ultrafast electron diffractometer and lattice dynamics in few-layer graphene — Christian Gerbig, Arne Senftleben, Silvio Morgenstern, Marlene Adrian, Cristian Sarpe, and •Thomas Baumert — Universität Kassel, Institut für Physik und CINSaT, Heinrich-Plett-Straße 40, D-34132 Kassel

Time-resolved diffraction with femtosecond electron pulses has become a promising technique to directly provide insights into photo induced primary dynamics at the atomic level in molecules and solids. Ultrashort pulse duration as well as extensive spatial coherence are desired, however, space charge effects complicate the bunching of multiple electrons in a single pulse. We experimentally investigate the interplay between spatial and temporal aspects of resolution limits in ultrafast electron diffraction (UED) on our highly compact transmission electron diffractometer. To that end, the initial source size and charge density of electron bunches are systematically manipulated and the resulting bunch properties at the sample position are fully characterized in terms of lateral coherence, temporal width and diffracted intensity. We obtain electron pulse durations down to 120 fs and transversal coherence lengths up to 20 nm. The performance of our compact UED setup at selected electron pulse conditions is finally demonstrated in a time-resolved study of lattice dynamics in few-layer graphene after optical excitation. During the heating process, we observe shearing modes and acoustic breathing modes.

O 77.3 Thu 11:30 MA 005

First principles molecular dynamics simulations of nonthermal structural dynamics in nanoscale materials — Tobias Zier, Bernd Bauerhenne, Benjamin Bartocha, Sergej Krylow, Martin E. Garcia, and •Eeuwe S. Zijlstra — Theoretical Physics, FB10, University of Kassel, Germany

Femtosecond-laser pulses create extreme out-of-hermodynamic-equilibrium conditions in matter with electrons that are orders of magnitude hotter than the ions. We simulated the ensuing nonthermal structural dynamics in Si, Ge, Sb, TiO2, and BN-nanotubes with our ab initio Code for Highly excIted Valence Electron Systems (CHIVES) [1], where the laser-excited potential energy surface is computed on the fly. Our simulations allow us to follow materials through exotic types of motion, for example, fractional diffusion in Si [2]. Computed time-resolved structure factors and structure functions provide a direct link to ultrashort electron diffraction experiments.

[1] E. S. Zijlstra, A. Kalitsov, T. Zier, and M. E. Garcia: "Squeezed thermal phonons precurse nonthermal melting of silicon", Phys. Rev. X 3, 011005 (2013).

[2] E. S. Zijlstra, A. Kalitsov, T. Zier, and M. E. Garcia: "Fractional diffusion in silicon", Adv. Mater. 25, 5605 (2013).

Coffee Break 15min.

O 77.4 Thu 12:00 MA 005

Ultrafast metamorphosis of a complex charge-density wave — Kerstin Haupt[1], Maximilian Eichberger[2], Nicolas Erasmus[1], Andrea Berenike Rohwer[1], Jure Demsar[3], Kai Rossnagel[4], and •Heinrich Schwoerer[1] — [1]Stellenbosch University, Stellenbosch 7600, South Afrika — [2]University of Konstanz, 78457 Konstanz, Germany — [3]University of Mainz, 55128 Mainz, Germany — [4]University of Kiel, 24098 Kiel, Germany

The transitions between commensurately and incommensurately modulated crystalline phases generally involve the formation and rearrangement of domain walls (discommensurations) and is thus thought to be extremely slow. Here, using ultrafast electron diffraction, we directly observe the structural dynamics of a photo-induced transition between a nearly commensurate and an incommensurate charge-density-wave phase in the strongly correlated layer compound 1T-TaS_2. The formation of the incommensurate phase is found to proceed in two steps, an ultrafast (≈1 ps) nucleation and a slower, thermally activated growth of domains. The transition can be fully completed in ≈100 ps, orders of magnitude faster than previously observed for commensurate-to-incommensurate transitions. The mechanism and time scales of this transition may be generic for ultrafast structural transformations between complex phases in which there is no direct path via coherent excitation of specific lattice modes.

O 77.5 Thu 12:15 MA 005

Structural dynamics in 2D semiconductors probe by ultrafast electron diffraction. — •Roman Bertoni, Lutz Waldecker, and Ralph Ernstorfer — Structural & Electronic Surface Dynamics, Fritz-Haber-Institut der MPG, Berlin, Germany

Femtosecond electron diffraction is known to be a powerful tool to study the interaction of lattice and electrons in two-dimensional systems during out-of-equilibrium dynamics. Recently, a new class of materials, transition metal dichalcogenide (TMDC), appears promising in order to build nanoscale devices.

We excite a nanometric thin film of WSe2 in resonance with an excitonic band and probe the resulting structural dynamics with ultrafast electron diffraction. We also use complementary technics implying femtosecond optical spectroscopy. By doing so, we address the impact of excitonic excitations onto the lattice and the electron-phonon coupling. Our results reveal an energy transfer from the electronic system to the lattice on picosecond time scale. In parallel, we observe a coherent structural response via the instantaneous activation of coherent phonon.

O 77.6 Thu 12:30 MA 005

Initial Dynamics of the Photo Induced (8×2)↔(4×1) Peierls-like Phase Transition of the In/Si(111) System — •T. Frigge, B. Hafke, B. Krenzer, C. Streubühr, P. Zhou, M. Ligges, D. von der Linde, U. Bovensiepen, and M. Horn-von Hoegen — Department of Physics, University of Duisburg-Essen, Lotharstr. 1, 47057 Duisburg, Germany

The wire-type arrangement of Indium atoms on a Silicon(111) surface serves as a famous prototype for the formation of a quasi one-dimensional charge density wave in the groundstate. We used ultrafast time-resolved surface-sensitive electron diffraction to investigate the transient non-equilibrium dynamics of the impulsively driven $(8 \times 2) \rightarrow (4 \times 1)$ phase transition. Excitation of the (8×2) groundstate with fs-laser pulses revealed the formation of a metastable, supercooled (4×1) excited state at 30 K. Utilizing a tilted pulse front scheme together with a new 30 keV electron gun design we improved the temporal resolution to less than less 400 fs. The high transversal coherence length of 40 nm allows to follow both the initial dynamics in the unit-cell and the kinetics of the phase front on the nano-scale. At fluences of 4 mJ/cm^2 the CDW groundstate is lifted across the entire surface on timescales of less than 400 fs. A transient temperature

rise of the Indium layer was determined through the intensity of the thermal diffuse background intensity utilizing the Debye-Waller effect. The laser induced heating by less than 40 K takes place on timescales 5 times slower than the metal-to-insulator transition which clearly rules out a simple thermal excitation scenario of the phase transition.

O 77.7 Thu 12:45 MA 005

Fluence-Dependence of the Photo Induced (8×2)↔(4×1) Phase Transition in the In/Si(111) System — •B. Hafke, T. Frigge, B. Krenzer, C. Streubühr, P. Zhou, M. Ligges, D. von der Linde, U. Bovensiepen, and M. Horn-von Hoegen — Universität Duisburg-Essen, Lotharstr. 1, 47057 Duisburg, Deutschland

High-energy electron diffraction at grazing incidence (RHEED) in a pump-probe setup is used to study ultrafast surface phenomena on a femtosecond timescale. We employed this technique to follow the initial dynamics of a driven phase transition of Indium on a Silicon(111) substrate. Optical pumping of the Charge Density Wave (8×2) ground state at a base temperature of 30 K triggers an electronically driven structural phase transition to an undercooled metastable (4×1) phase. For fluences above 1.3 mJ/cm^2 the entire surface is excited into the (4 × 1) phase. At lower fluences the surface is excited only partially. We observe fluence-dependent changes of diffraction intensities: For low fluences the transition into the (4 × 1) phase occurs within 11 ps, at higher fluences of 1.8 mJ/cm^2 within 5 ps. We explain this by excitation of free electrons upon laser excitation, which accumulate at specific surface areas, e.g. defects, domain walls or steps. These areas act as nucleation seeds for the (8 × 2) → (4 × 1) transition which propagates linear in time. At higher fluences more nucleation seeds contribute, resulting in a shorter conversion time. For fluences below 1.3 mJ/cm^2 the excited carrier density is not sufficient to induce the conversion of the entire surface.

O 77.8 Thu 13:00 MA 005

Femtosecond-laser-excited silicon surfaces — •Tobias Zier, Eeuwe S. Zijlstra, and Martin E. Garcia — University of Kassel, Germany

A femtosecond-laser irradiation of crystalline silicon leads to interesting ultrafast phenomena, like thermal phonon squeezing [1], fractional diffusion and ultrafast melting [2], because of the highly non-equilibrium conditions created by the excitation. In particular, the energy is deposited mostly in the electronic system, which results in a very high electronic temperature (of the order of 10 000 K), and gives rise to an extreme change in the potential energy surface for the ions and therefore in the bonding character of the crystal. With our in-house code CHIVES we perform MD-simulations of femtosecond-laser excited solids which allows us to analyse the microscopic atomic motions near the surface. We check the possibility of femtosecond-laser-induced structural phenomena at the surface, the existence of coherent vibrational surface states and the influence on the above mentioned phenomena.

[1] E. S. Zijlstra, A. Kalitsov, T. Zier, M. E. Garcia, Phys. Rev. X 3,011005 (2013)

[2] E. S. Zijlstra, A. Kalitsov, T. Zier, M. E. Garcia, Adv. Mater. 25, 5605 (2013)