O 70: Structural Dynamics in Nanoscale Materials Probed by Ultrashort Electron Pulses

Time: Wednesday 18:15–21:00 Location: Poster A

O 70.1 Wed 18:15 Poster A

Lattice dynamics in few-layer Molybdenum disulfide investigated by Ultrafast Electron Diffraction — •Marlene Adrian, Christian Gerbig, Silvio Morgenstern, Christian Sarpe, Arne Senftleben, and Thomas Baumert — University of Kassel, Institute of Physics (CINSaT), D-34132 Kassel, Germany

Molybdenum disulfide (MoS2) is a prototype example for transition metal dichalcogenides (TMDs), which form a group of van der Waals bound two-dimensional layered materials [1]. Due to their unique electronic and optical properties such as circular dichroism, a strong spin-orbit coupling and a shift from indirect to direct band gap semiconductor with decreasing film thickness from bulk to monolayer, TMDs are interesting for both fundamental research and industrial applications such as electronic devices [2].

We study dynamical processes following optical excitation in few-layer MoS2 by means of time-resolved Ultrafast Electron Diffraction (UED), which has become a promising technique to directly provide insights into dynamics in crystalline solids at the microscopic level with a sub-picosecond temporal resolution [3, 4]. Our highly compact UED-setup is fully characterized by experiments and many-body simulations [5].

[1] S. Z. Butler et al., ACS Nano 7, 2898 (2013).
[2] G. Berghäuser and E. Malic, arXiv:1311.1045 (2014).
[3] A. H. Zewail, J. Phys. Chem. 98, 2782-2796 (1994).
[4] B. Siwick and D. Miller, Science 302, 1382-1385 (2003).
[5] C. Gerbig et al., in preparation (2014).

O 70.2 Wed 18:15 Poster A

Resolution studies on a compact femtosecond transmission electron diffractometer and phonon decay in single crystalline graphite — Christian Gerbig[1], •Silvio Morgenstern[1], Marlene Adrian[1], Christian Sarpe[1], Arne Senftleben[1], Matthias Wollenhaupt[2], and Thomas Baumert[1] — [1]University of Kassel, Institute of Physics (CINSaT), D-34132 Kassel, Germany — [2]University of Oldenburg, Institute of Physics, D-26111 Oldenburg, Germans

Time-resolved diffraction, using x-ray or electron probes, has become a promising technique to directly provide insights into dynamics at the molecular level with ultrafast precision [1]. We study dynamical processes in single crystalline graphite by means of ultrafast electron diffraction in order to expand the understanding of phonon generation and decay mechanisms being essential for future carbon based electronic devices [2].

Our highly compact DC electron diffractometer is fully characterized by experiments and N-body simulations. At balanced conditions a temporal resolution of 200 fs along with high-definition diffraction is achieved for dynamical studies on graphite single crystals in a maintainable measurement time [3]. We further present generation and decay processes of incoherent as well as coherent phonons in graphite as a function of film thickness down to few-layer graphene.

[1] M. Chergui & A. H. Zewail, Chem. Phys. Chem. 10, 28 (2009).
[2] T. Kampfrath et al., Phys. Rev. Lett. 95, 187403 (2005).
[3] G. Sciaini & R. J. D. Miller, Rep. Prog. Phys. 74, 096101 (2011).

O 70.3 Wed 18:15 Poster A

Laser-induced heating of nano-crystalline graphene monitored by Ultrafast Electron Diffraction — •Silvio Morgenstern, Christian Gerbig, Marlene Adrian, Xaver Holzapfel, Arne Senftleben, Matthias Wollenhaupt, and Thomas Baumert — Universität Kassel, Institut für Physik and Center for Interdisciplinary Nanostructure Science and Technology (CINSaT), D - 34132 Kassel, Germany

Ultrafast Electron Diffraction (UED) has lately become one of the most promising techniques to directly provide insights into fundamental dynamics in solids at the microscopic level and on the pico- to subpicosecond timescale [1,2]. In this contribution we present our UED setup to reach a high spatial and temporal resolution below 200 fs [3]. Additionally we present first results of time-resolved diffraction experiments on nano-crystalline graphene (NC graphen) [4] and discuss the possibility of time-resolved observations of out-of-plane dynamics in such materials [5]. Finally we compare our results to results from CVD graphene [6].

[1] A. H. Zewail, J. Phys. Chem. 98, 2782-2796 (1994), [2] B. Siwick & D. Miller, Science 302, (5649), 1382-1385 (2003), [3] C. T. Hebeisen, Opt. Letters Vol. 31, No. 23, 3571 (2006) [4] A. Truchanin, ACS Nano Vol. 5, No. 5, 3896 (2011), [5] J. C. Meyer, Nature 446, 60-63 (2007), [6] M. Schäfer, New J. Phys. 13, 063030 (2011)

O 70.4 Wed 18:15 Poster A

Laser-induced ultrafast phenomena in Ge — •Tobias Zier, Eeuwe S. Zijlstra, and Martin E. Garcia — University of Kassel, Germany

The extreme non-equilibrium state in a solid induced by an intense femtosecond laser pulse excitation, in which the electrons have a temperature of several 10 000 K and the atoms remain nearly unaffected, allows to access pathways that are not accessible in thermodynamic equilibrium and therefore gives rise to interesting effects. Prominent phenomena are thermal phonon squeezing, nonthermal melting and solid-to-solid phase transitions. We performed ab initio Molecular-Dynamics simulations of laser-excited germanium in order to study the structural response of this semiconducting material as a function of the fluence and compared our results to recent experimental findings [1].

[1] Trigo, M et al., Nature Phys. 9, 790 (2013)

O 70.5 Wed 18:15 Poster A

Delayed surface phonon excitation in Bi(111) films — Verena N. Tinnemann, Tim Frigge, •Boris Krenzer, Bernd Hafke, Annika Kalus, Carla Streubühr, Ping Zhou, Manuel Ligges, Dietrich von der Linde, Uwe Bovensiepen, and Michael Horn-von Hoegen — Universität Duisburg-Essen, Lotharstr. 1, 47057 Duisburg

We used ultra-fast reflection high energy electron diffraction to follow the excitation of surface phonons on an epitaxial Bi(111) film on Si(111) upon impulsive excitation with a femtosecond laserpulse. The thermal motion of the atoms is determined from the transient intensity drop described through the Debye-Waller effect. The excitation of vibrational motion of the surface atoms occurs with a time constant of 12 ps. For strained Bi(111) films grown on Si(001) we observe a bi-exponential behaviour with a fast ($<$ 6 ps) and slow, temperature dependent (12 to 60 ps) component for the vibrational excitation of the surface. We attribute the different response to the presence of an electronic surface state for relaxed 4.5 nm thick Bi films on Si(111) while this state is absent on the strained 4.5 nm thick Bi films grown on Si(001). In the first case the excited carriers populate the surface state without heating the bulk and weak electron phonon coupling excite surface phonons on a timescale of 12 ps. On Si(001) the excited carriers remain in the bulk of the film, couple to phonons on a much faster time scale $<$ 6 ps, and surface phonons are then excited only by anharmonic coupling between bulk and surface phonon modes.

O 70.6 Wed 18:15 Poster A

Sub 400 fs temporal resolution in reflection high energy electrondiffraction at surfaces — •Tim Frigge, Bernd Hafke, Boris Krenzer, Carla Streubühr, Ping Zhou, Manuel Ligges, Dietrich von der Linde, Uwe Bovensiepen, and Michael Horn-von Hoegen — Universität Duisburg-Essen, Lotharstr. 1, 47057 Duisburg

In time resolved reflection high energy electron diffraction at surfaces an ultra short electron pulse probes the surface at grazing incidence. At energies of 30 keV the electrons travel at 1/3 of the speed of light and require 20 ps to traverse a typical sample width of 2 mm. For pump-pulses at normal incidence the sample surface is excited simultaneously. This resulting geometric velocity mismatch limits the overall experimental temporal resolution. Employing a tilted pulse front scheme for the laser excitation we are able to overcome this limitation and obtain a temporal resolution of 600 fs when probing the entire sample width. Reduction of the electron number per pulse and probing only a narrow part of the sample further improves the temporal resolution to less than 400 fs without electron pulse compression. Temporal broadening due to vaccum dispersion on the 8 cm distance between photocathode and sample is minimized by the narrow initial electron energy spread of only 100 meV of the back illuminated Au photocathode. Additionally, a new electron gun design ensures a vertical coherence length of more than 30 nm. An integration time of 10 s is sufficient for good statistics for one diffraction image, i.e. a diffraction movie with 100

frames at 50 fs delay steps is recorded in less than 30 min.

O 70.7 Wed 18:15 Poster A

Quantum coherent interaction of electrons with optical near-fields in an ultrafast electron microscope — Armin Feist, •Katharina E. Echternkamp, Jakob Schauss, Sergey V. Yalunin, Sascha Schäfer, and Claus Ropers — 4th Physical Institute, University of Göttingen, Göttingen, Germany

Harnessing the coherent interaction of light and matter is expected to play a key role for future quantum computation, communication and metrology. Here, we present the coherent quantum state manipulation of free electrons in an ultrafast electron microscope [1]. Ultrafast transmission electron microscopy (UTEM) is a laser pump/electron probe technique, enabling the investigation of ultrafast processes on the nanometer length scale [2]. We have recently implemented an UTEM by modifying a commercial Schottky field emission TEM (JEOL JEM-2100F). In our experiments, the electron beam is focused to a spot close to a surface of a conical gold tip. The high spatial confinement of the optically excited near-field of the nanostructure allows for an otherwise forbidden dipolar coupling between the free electrons and photons. In the electron kinetic energy spectra, we observe the creation of spectral sidebands, stemming from the absorption and emission of multiple photons [3]. The field dependent sideband populations reveal the quantum coherence of the process.

[1] A. Feist *et al.*, submitted (2014).
[2] A.H. Zewail, Science, **328**, 187 (2010).
[3] B. Barwick *et al.*, Nature, **462**,902 (2009).

O 70.8 Wed 18:15 Poster A

Classical potential for femtosecond-laser excited silicon — •Bernd Bauerhenne, Tobias Zier, Eeuwe S. Zijlstra, and Martin E. Garcia — Theoretische Physik - Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany

Using femtosecond-laser pulses it is possible to manipulate solids in unconventional ways, which can, for example, be used to write structures on surfaces. In order to understand and predict such processes an effective classical potential is a key tool. Here we developed a new potential for laser-excited silicon starting from known classical potentials by fitting realistic density functional theory simulations of nonthermal melting of silicon [1]. We also optimized the potential function. As first results, we found, that our new potential describes accurately the fractional diffusion during ultrafast melting of silicon and that we can reproduce both the time-resolved pair correlation function and the angular distribution function.

[1] E. S. Zijlstra, A. Kalitsov, T. Zier, and M. E. Garcia, Fractional diffusion in silicon, Adv. Mater. 25, pp. 5605-5608, 2013.

O 70.9 Wed 18:15 Poster A

Imaging, diffraction, and interferometry with ultrabright electron pulses — •Robert Bücker[1], Albert Casandruc[1], Chiwon Lee[1], Philipp Pelz[1], Haider Zia[1], Günther Kassier[1], and R. J. Dwayne Miller[1,2] — [1]Max Planck Institute for the Structure and Dynamics of Matter, CFEL, Luruper Chaussee 149, 22761 Hamburg, Germany — [2]Departments of Chemistry and Physics, University of Toronto, Toronto Ontario, M5S 3H6, Canada

High-brightness electron pulses in the ns to μs regime promise to be the optimal probe for determination of structure and irreversible dynamics in biological systems. Combined with the favorable elastic-to-inelastic ratio of electron scattering, the large transverse coherence length achievable with nanometric emitters allows maximizing the information content per scattering event, hence keeping radiation damage at a minimum and allowing to extract sufficient structural information even from small samples, such as nanocrystals. In this contribution, we present the concept, implementation, and first results of experiments tailored to these goals. This comprises both field emission-based electron diffraction beam lines in the range from few to hundreds of kilovolts, as well as transmission electron microscopes enhanced with pulsed sources and high-speed imaging detectors.

Particular attention will be paid to a compact 12 kV apparatus, designed for interferometric characterization of various kinds of pulsed electron sources, as well as proof-of-principle demonstrations of coherent diffractive schemes with bright many-electron pulses.

O 70.10 Wed 18:15 Poster A

Characterization of a picosecond electron gun for diffraction experiments — •Nele L. M. Müller[1], Sebastian Trippel[1], Terence G. Mullins[1], Karol Długołecki[1], and Jochen Küpper[1,2,3] — [1]Center for Free-Electron Laser Science, DESY, Hamburg — [2]The Hamburg Center for Ultrafast Imaging, Hamburg — [3]Department of Physics, University of Hamburg

The aim of the presented work is to investigate the structure and dynamics of molecules in the gas-phase by electron diffraction experiments. The contribution presents our newly set-up electron gun that will be combined with an existing controlled-molecules apparatus [1]. The developed DC electron gun can produce up to 10 million electrons per pulse and uses an electro-static lens for focusing. The expected pulse durations are tens of picoseconds. The focusing electrodes are arranged in a configuration similar to a Velocity Map Imaging spectrometer. Besides focusing this can be used to measure the spatial and velocity distribution of the electron pulse when emitting from the cathode. In combination with electron trajectory simulations this allows for further characterization of the electron beam, as for example the determination of pulse duration and coherence length. Electron diffraction data from solid state and gaseous samples will be presented.

[1]Trippel et al., *Mol. Phys.* **111**, 1738-1743 (2013)

O 70.11 Wed 18:15 Poster A

UED@SLAC: Structural dynamics in laser-excited solids studied by MeV electron diffraction — •Klaus Sokolowski-Tinten[1], Renkai Li[2], Alex H. Reid[2], Stephen P. Weathersby[2], Garth Brown[2], Martin Centurion[3], Tyler Chase[2], Ryan Coffee[2], Jeff Corbett[2], Josef C. Frisch[2], Markus Guehr[2], Nick Hartmann[2], Carsten Hast[2], Ling Ho[2], Michael Horn von Hoegen[1], David Janoschka[1], Keith Jobe[2], Erik Jongewaard[2], James R. Lewandowski[2], Justin E. May[2], Dough McCormick[2], Frank Meyer zu Heringdorf[1], Xiaozhe Shen[2], Christian Witt[1], Juhao Wu[2], Jie Yang[2], Dürr Hermann[2], and Xijie Wang[2] — [1]Faculty of Physics and Centre for Nanointegration Duisburg-Essen, University of Duisburg-Essen, Lotharstraße 1, 47048 Duisburg, Germany — [2]SLAC National Accelerator Laboratory, Menlo Park, USA — [3]University of Nebraska-Lincoln, Lincoln, USA

With the aim to provide synergistic and complementary experimental capabilities for the study of ultrafast processes at their fundamental length and time scales SLAC National Laboratory, operator of the world's first hard X-ray free electron laser, the Linear Coherent Light Source, has recently started an initiative for ultrafast electron scattering and microscopy. This contribution will discuss the setup for femtosecond time-resolved MeV electron diffraction, which has been brought into operation over the last few months at SLAC's Accelerator Structure Test Area (ASTA), as well as results from first user experiments addressing the ultrafast lattice response in laser-excited thin Bismuth-films.

O 70.12 Wed 18:15 Poster A

Miniaturized photoelectron gun for ultrafast low-energy electron diffraction — •Gero Storeck, Simon Schweda, Max Gulde, Sebastian Schramm, Sascha Schäfer, and Claus Ropers — IV. Physical Institute, University of Göttingen, 37077 Göttingen, Germany

Probing structural dynamics at surfaces with high temporal resolution provides insights into a rich class of phenomena, which are unique to quasi-two-dimensional systems [1,2]. Utilizing a laser-pump/electron-probe scheme, ultrafast low-energy electron diffraction (ULEED) promises direct access to such processes, provided that the generation of well-collimated ultrashort low-energy electron pulses is achieved. Recently, nanometric photocathodes were shown to minimize spatial and temporal pulse broadening at low electron energies, resulting in a first ULEED experiment [3]. Using this experimental setup, laser-driven dynamics in a polymer/graphene bilayer were resolved with a temporal resolution of 2 ps in a transmission geometry. In a further development, we present the design of a compact pulsed electron gun allowing for ultrafast low-energy electron experiments in backscattering diffraction. First applications to graphene on silicon carbide are shown.

[1] A. Hanisch-Blicharski et al., Ultramicroscopy 127, 2-8 (2013). [2] J. M. Kosterlitz et al., J. Phys. Chem. 6, 1181-1203 (1973). [3] M. Gulde et al., Science 354, 200 (2014).

O 70.13 Wed 18:15 Poster A

Lattice response with respect to the orientation of the crystal to the femtosecond laser excitation observed by time-resolved electron diffraction — Carla Streubühr, •Ping Zhou, Manuel Ligges, Klaus Sokolowski-Tinten, Thomas Payer,

Frank Meyer zu Heringdorf, and Michael Horn-von Hoegen — Fakultät für Physik und Zentrum für Nanointegration, Universität Duisburg-Essen

One of the fundamental questions in the field of ultrafast structural dynamics is the time scale for the energy transfer between the electron and phonon subsystem of a solid after femtosecond optical excitation. For crystalline materials, especially for layered materials like graphite or most of superconductors, the material properties depend strongly on the symmetries of the crystals. The excitation and relaxation of the lattice cannot be treated as symmetrical. The advantage of electron diffraction is the ability to map many diffraction orders at the same time and, thus, obtain rich structural information. We present here results of time resolved electron diffraction experiments on Bismuth and Nickel membranes to demonstrate the difference of the energy transfer of hot electrons to the lattice depending on the direction of the displacements. Besides the disordered thermal displacement, strain waves can be generated. They propagate between the surfaces and influence on the diffraction intensity in different way depending on the materials and the diffraction geometries. By using a simple model we could extracted strain wave information from the disordered thermal displacement and get the frequency and damping of the strain wave.

O 70.14 Wed 18:15 Poster A

Observation of heat transport by time-resolved x-ray diffraction using a conventional microfocus x-rax tube — •Mathias Sander[1], Peter Gaal[2], and Matias Bargheer[1] — [1]Institut für Physik und Astronomie, Universität Potsdam, Karl-Liebknecht-Str. 24-25,14476 Potsdam,Germany — [2]Institut für Nanostrukturen und Festkörperphysik, Universität Hamburg, Jungiusstr. 11 20355 Hamburg, Germany

We demonstrate a table top setup for real-time observation of heat transport in crystalline layered structures via time-resolved x-ray diffraction on timescales from nanoseconds to milliseconds. The sample is excited either by an electronically heated microchip or by short laser pulses depending on the relevant timescale. The x-rays are generated by a conventional microfocus x-ray tube, focused by a polycapillary x-ray optic and monochromatized by a single reflection, e.g. from a Si or HAPG crystal. X-rays diffracted by the sample are detected in a fast x-ray phosphor and a photomultiplier and fed into a time-correlated single-photon counting module, which records histograms of events along the entire pump-probe delay. The resulting data shows time-dependent angular shifts of Bragg reflections which are interpreted as thermal diffusion dynamics on nano- and micrometer length scales.

O 70.15 Wed 18:15 Poster A

Inelastic scattering of low-energy electrons by optical near-fields — •Lars Drögemüller, Sascha Schäfer, and Claus Ropers — IV. Physical Institute, University of Göttingen, 37077 Göttingen, Germany

Ultrafast low-energy electron diffraction (ULEED) is a new tool in ultrafast surface science [1], which utilizes a laser-pump/electron-probe scheme to study the structural dynamics at surfaces and ultrathin films. For the future development of ULEED, a reliable and quantitative temporal characterization of the employed low-energy electron pulses is required. While inelastic near-field scattering was successfully applied for pulse characterization at high-energies [2, 3], its extension to the low-energy regime is challenging.

In this study, numerical simulations based on the finite-element-method were performed to investigate near-field electron scattering at low energies. Plasmonic effects and optical resonances in tailored nanostructures were considered to optimize the electron-photon scattering cross section.

[1] M. Gulde et al., Science 345, 200-4 (2014). [2] B. Barwick, D. J. Flannigan A. H. Zewail, Nature 462, 902-6 (2009). [3] F. O. Kirchner, A. Gliserin, F. Krausz, P. Baum, Nature Photonics 8, 52-57 (2013).

O 70.16 Wed 18:15 Poster A

Cooperative atomic motion probed by femtosecond electron diffraction — Maximilian Eichberger[1] and •Jure Demsar[2] — [1]Department of Physics, University of Konstanz — [2]Institute of Physics, Johannes Gutenberg-University Mainz

Recently, several studies of light-induced suppression or quenching of charge density wave order were performed using ultrafast diffraction methods. This coherent process, which takes place on a 100 femtosecond timescale (a fraction of a period of the corresponding amplitude mode), is accompanied by a rapid sub-picosecond energy transfer to the lattice via strong electron-phonon and phonon-phonon scattering. Both processes, the coherent order parameter dynamics and the incoherent redistribution of energy among different subsystems, affect the diffraction pattern. For their comparable timescales they are hard to distinguish based on the dynamics alone.

Using ultrafast electron diffraction in transmission we show, that by simultaneous tracking the intensities of lattice and super-lattice diffraction peaks for multiple diffraction orders (this being one of the main advantages of ultrafast electron diffraction against femtosecond X-ray methods) the two processes can be effectively disentangled.

O 70.17 Wed 18:15 Poster A

Coherent and incoherent electron-phonon coupling in graphite observed with radio-frequency compressed ultrafast electron diffraction — •Bradley Siwick, Robert Chatelain, Vance Morrison, Bart Klarenaar, and Jean-Phillipe Boisvert — McGill University, Center for the Physics of Materials, Montreal, Canada

Radio-frequency compressed ultrafast electron diffraction has been used to probe the coherent and incoherent coupling of impulsive electronic excitation at 1.55 eV (800 nm) to optical and acoustic phonon modes directly from the perspective of the lattice degrees of freedom. A bi-exponential suppression of diffracted intensity due to relaxation of the electronic system into incoherent phonons is observed, with the 250 fs fast contribution dominated by coupling to the E2g2 optical phonon mode at the Γ-point (Γ-E2g2) and A1 optical phonon mode at the K-point (K-A1). Both modes have Kohn anomalies at these points in the Brillouin zone. In addition, electronic excitation leads to both in-plane and out-of-plane coherent lattice responses in graphite whose character we are able to fully determine based on spot positions and intensity modulations in the femtosecond electron diffraction data. The in-plane motion is specifically a Γ-point shearing mode of the graphene planes with an amplitude of approximately 0.06 pm and the out-of-plane motion an acoustic breathing mode response of the film.

O 70.18 Wed 18:15 Poster A

Structure and Dynamics with Ultrafast Electron Microscopes — •Bradley Siwick[1], Mark Stern[1], Lily Nikolova[2], Federico Rosei[2], Jennifer McLeod[2], Tom Lagrange[3], and Bryan Reed[3] — [1]McGill University, Center for the Physics of Materials, Montreal, Canada — [2]Institut National de la Recherche Scientifique, Centre Énergie, Matériaux, Télécommunications, Varennes, Canada — [3]Condensed Matter and Materials Division, Physical and Life Sciences Directorate, Lawrence Livermore National Laboratory, Livermore, USA

This poster will describe a series of recent experiments on explosive crystallization in amorphous germanium, where we have directly "watched" the nano-micro structural evolution in the material using dynamic transmission electron microscopy (DTEM). Direct visualization of the crystallization front allows for time-resolved snapshots of the initiation and roughening of dendrites on sub-microsecond time scales and a rapid transition to a ledge-like growth mechanism at longer times. Direct observations of the speed of the explosive crystallization front as it evolves along a laser-imprinted temperature gradient have been used to experimentally determine the complete interface response function (i.e., the temperature-dependent front propagation speed) for this process, which reaches a peak of 16m/s. These result suggests a modification to the liquid-mediated mechanism commonly used to describe this process that replaces the phase change at the leading amorphous-liquid interface with a change in bonding character (from covalent to metallic) occurring in the hot amorphous material.