

O 84: Focus Session: Structural Dynamics in Nanoscale Materials Probed by Ultrashort Electron Pulses

Time: Thursday 15:00–18:15

Location: MA 005

Topical Talk

O 84.1 Thu 15:00 MA 005

Femtosecond electron probes for the investigation of structural dynamics and ultrafast currents in nanomaterials — ●RALPH ERNSTORFER, MELANIE MÜLLER, LUTZ WALDECKER, ROMAN BERTONI, THOMAS VASILEIADIS, and ALEXANDER PAARMANN — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin

We investigate ultrafast structural as well as electronic dynamics in low-dimensional systems such as two-dimensional materials, one-dimensional nanowires, and nanoparticles. Such studies require femtosecond probes strongly interacting with small-volume samples. Electrons with energies ranging from 50 to 1000 eV exhibit extremely large scattering cross sections and high sensitivity to electric fields. Employing a laser-triggered point-like source of either divergent or collimated electron wave packets, we developed a hybrid approach for femtosecond point projection microscopy (fsPPM) and femtosecond low-energy electron diffraction (fsLEED) [1]. We investigate ultrafast electric currents in nanowires with sub-100 femtosecond temporal and few 10 nm spatial resolutions. This new low-energy electron technique is complemented by femtosecond transmission electron diffraction performed with 100 keV electrons. A highly compact diffractometer design allows for delivering 100 fs long pulses containing up to 5000 electrons to the sample [2]. We will discuss structural dynamics and electron-lattice interaction in confined materials such as quasi-2D materials and nanoparticles. References: [1] M. Müller et al., *Nature Communications* 5, 5292 (2014). [2] L. Waldecker, arXiv:1412.1942 (2014).

O 84.2 Thu 15:30 MA 005

Ultrafast transmission electron microscopy with nanoscopic electron sources — ARMIN FEIST, REINER BORMANN, KATHARINA ECHTERNKAMP, JAKOB SCHAUSS, NARA RUBIANO, ●SASCHA SCHÄFER, and CLAUS ROPERS — 4th Physical Institute, University of Göttingen, Göttingen, Germany

Ultrafast transmission electron microscopy (UTEM) is a laser-pump/electron-probe technique, which promises to combine the ultrafast temporal resolution of pump-probe approaches with the spatial resolution of electron microscopy [1]. However, to harness the full capabilities of UTEM, novel laser-triggered electron sources are required, which deliver high-brightness sub-picosecond electron pulses.

Here, we present the development and application of an advanced UTEM instrument based on the custom modification of a commercial electron microscope. Specifically, we employ electron sources based on the localized photoemission [2] from a nanoscale needle photocathode. The enhanced optical field at the tip apex confines nonlinear photoemission to small emitter areas, enabling electron focal spot sizes on the sample of about 10 nm with electron pulse durations of less than 700 fs. First applications are presented, including time-resolved Lorentz microscopy and the nanoscale probing of quantum coherent interactions between free electrons and optical near-fields [3].

[1] A. H. Zewail, *Science* 328, 187-93 (2010). [2] M. Gulde, S. Schweda, G. Storeck, M. Maiti, H. K. Yu, A. M. Wodtke, S. Schäfer, C. Ropers, *Science* 345, 200-204 (2014). [3] A. Feist, K. Echternkamp, J. Schauss, S. V. Yalunin, S. Schäfer, C. Ropers, submitted.

O 84.3 Thu 15:45 MA 005

Simultaneous observation of quantization and interference of a surface plasmon polariton by PINEM — ●YOSHIE MUROOKA¹, TOM LUMMEN¹, LUCA PIAZZA¹, ERIK QUIÑONEZ², BRYAN REED³, BRETT BARWICK², and FABRIZIO CARBONE¹ — ¹École Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland — ²Trinity College, Hartford, CT 06106, USA — ³Lawrence Livermore National Laboratory, Livermore, CA 94551, USA

For applications in photonics and optical data storage, surface plasmon polariton (SPP) is intensively studied because of its high spatial confinement and precise optical control with, for example, its quantum properties. SPP can be photo-generated on a metal surface as a propagating electromagnetic wave, while the vertical confinement is in the sub-wavelength regime. Lately, such SPP fields have been studied in terms of their wave-particle duality. Here, using recently developed Photo-induced near field electron microscopy (PINEM), we have imaged both the quantization and the interference of the confined fields simultaneously on an isolated metal nanowire. PINEM was real-

ized on femtosecond-TEM equipped with an electron energy analyzer. The SPP field was induced by pulsed laser, and probed by electron pulses that were spatio-temporally overlapped with the light. The exchange of energy between the field and the electrons was found to be quantized, and the spatial distribution of the field was synchronously revealed as its interference pattern. This methodology enables to visualize and control SPP fields at nanoscale, and provides a novel tool to understand the fundamental properties of confined electromagnetic fields.

O 84.4 Thu 16:00 MA 005

Coherence Properties of Laser-Triggered Field Emitters — ●DOMINIK EBERGER^{1,2}, JAKOB HAMMER^{1,2}, MAX EISELE², MICHAEL KRÜGER^{1,2}, JONATHAN NOE³, ALEXANDER HÖGELE³, and PETER HOMMELHOFF^{1,2,4} — ¹Friedrich Alexander University Erlangen-Nuremberg, Department of Physics, D-91508 Erlangen — ²Max Planck Institute of Quantum Optics, D-85748 Garching — ³Fakultät für Physik and Center for NanoScience (CeNS), Ludwig-Maximilians-Universität München, D-80539 München — ⁴Max Planck Institute for the Science of Light, D-91508 Erlangen

Sharp metal nanotips provide bright and spatially coherent electron beams in DC-field emission. They serve as workhorse in electron imaging and holography. However, the spatial coherence properties, commonly quantified by the effective source radius r_{eff} , are expected to depend strongly on the emission process and have so far not been measured for laser-triggered metal tips.

Here, we present a comparison of r_{eff} for a tungsten tip triggered with near-UV pulses and in DC-field emission. From electron interference patterns obtained by means of a freestanding carbon nanotube biprism an upper bound for r_{eff} is deduced. We find $r_{\text{eff}} \leq (0.80 \pm 0.05)$ nm in laser induced and $r_{\text{eff}} \leq (0.55 \pm 0.02)$ nm in DC-field emission, revealing that the spatial coherence is almost fully preserved in a one-photon emission process.

We expect this finding to have far-reaching ramifications for ultrafast electron imaging applications.

O 84.5 Thu 16:15 MA 005

Deflection of Electron Pulses by THz Fields — ●WALDEMAR SCHNEIDER^{1,2}, ANDREY RYABOV^{1,2}, DANIEL KREIER^{1,2}, FERENC KRAUSZ^{1,2}, and PETER BAUM^{1,2} — ¹Max-Planck-Institute of Quantum Optics — ²Ludwig-Maximilians-University Munich

Pump-probe electron diffraction and microscopy, based on laser excitation and probing with electrons, can provide a four-dimensional visualization of atomic motion in space and time. The time resolution is determined by the temporal and spatial structure of the electron pulse. Here, the temporal information of a single-electron pulse is mapped with THz fields into the transverse position on a screen, realising a THz-driven streaking camera. By spatially sectioning the electron pulse, spatial information was obtained. The deflection trace also revealed the shape of the THz-field at the sample.

The described approach represents a readily improvable method for temporal and spatial characterization of single-electron pulses for ultrafast diffraction applications.

Topical Talk

O 84.6 Thu 16:30 MA 005

Exploring the Spatial and Temporal Resolution Limits of Ultrafast Electron Microscopy — ●DAVID J. FLANNIGAN, DAYNE A. PLEMMONS, DANIEL R. CREMONS, and DAVID T. VALLEY — University of Minnesota, 421 Washington Avenue SE, Minneapolis, MN, 55455, USA

In ultrafast electron microscopy (UEM), the capabilities of transmission electron microscopy are extended into the femtosecond temporal domain. The operating principle of UEM requires spatiotemporal overlap of the photon pulse and electron packet at the specimen; at time zero, significant photon absorption by the freely-propagating electrons can occur. Overlap at the specimen suggests this phenomenon can be used to measure the response function and the electron packet properties. In this talk, I will discuss considerations for isolating the inherent artifacts of the highly non-linear near-field interactions from the actual packet characteristics. Further, I will discuss how temporal cross-sections of peaks in the electron-energy spectra corresponding to

high-order transitions are expected to exhibit the true temporal behavior of the electron packets. In general, the exceedingly small portion of the pump laser pulse capable of initiating such transitions results in temporal widths converging to the electron packet duration. Additionally, population of quantized virtual states occurring for an electron beam focused on a nanostructure suggests that the resulting energy distribution may produce discrete chromatic aberrations arising from the velocity dependence of the Lorentz force. I will discuss the prospect for detecting such phenomena in bright-field images.

O 84.7 Thu 17:00 MA 005

Developments in ultrafast electron microscopy and diffraction — ALEXANDER BAINBRIDGE and ●WILLIAM BRYAN — Department of Physics, Swansea University, Singleton Park, Swansea SA2 8PP, UK

A progress report on recent ultrafast electron microscopy and diffraction studies at Swansea University (UK) will be presented. Facilities to time-resolve charge migration on nanometre to micron scales are to be discussed, and are currently propagating sub-100fs electron pulses from a nanoscale metal tip (NSMT) to thin solid state samples mounted on a TEM substrate. This work employs the 20fs 2uJ output from a Light Conversion Orpheus-N OPA, which is split into a pump pulse illuminating the target and probe driving electron emission from the NSMT. Laser delivery includes an external compressor, FROG and pointing stabilisation. Field emission from this system indicates imaging to sub-hundred nm is currently possible.

Recent simulations investigating the ultimate time resolution of our PPM instrument will also be presented, and we will discuss balancing the requirements for field of view over time resolution. Modelling of electron pulse collimation with an electrostatic microlens will be presented, which will facilitate coherent diffractive imaging with time resolution, opening the door to tracking charge motions on the atomic scale. Clearly instrument design and implementation is just the start, and to illustrate our broad interests in this field, research directions involving graphene liquid cells, coupling nanorods to 2D crystalline materials and tracking charge flow through plasmonic nanoparticle strings will be discussed.

O 84.8 Thu 17:15 MA 005

Topical Talk
Ultrafast single-electron diffraction and its perspectives — ●PETER BAUM — Ludwig-Maximilians-Universität München — Max-Planck-Institut für Quantenoptik

Matter transformations are basically defined by atomic and electronic motions from initial to final conformations. Ultrafast electron diffraction and microscopy are good at seeing the atoms, but purely electronic motion is still mostly hidden. Here we report our recent progress with single-electron wave packets without space charge [1-2] for advancing 4D diffractive imaging into novel resolution and application regimes. Specifically, we report our shortest pulses so far (28 fs) and first diffraction applications on graphite and carbon nanotubes [3].

- [1] Peter Baum, J. Phys. B 47, 124005 (2014).
- [2] Gliserin, Apolonski, Krausz, Baum, NJP 14, 073055 (2012).
- [3] Lahme, Kealhofer, Krausz, Baum, Struct. Dyn. 1, 034303 (2014).

O 84.9 Thu 17:45 MA 005

Radiation-induced transitions in solids — NIKITA MEDVEDEV, ZHENG LI, and ●BEATA ZIAJA — CFEL DESY, Notkestrasse 85, 22607 Hamburg

Femtosecond intense light pulses from free-electron lasers can trigger structural transitions in solids. Their theoretical description is a challenge, as it has also to include contributing non-equilibrium processes. In order to account both for thermally and nonthermally triggered transitions, we extended our recently developed hybrid model by including non-adiabatic electron-phonon coupling. In this way the heating of a material due to the electron-phonon coupling could also be treated. We show model application for laser-induced transitions in carbon and silicon. The developed scheme is general and can be used in any molecular dynamics model, also with an implementation to describe structural transitions induced by electron pulses.

O 84.10 Thu 18:00 MA 005

Ultrafast Electron Diffraction on nano-crystalline Graphene — ●SILVIO MORGENSTERN, CHRISTIAN GERBIG, MARLENE ADRIAN, CRISTIAN SARPE, ARNE SENFTLEBEN, and THOMAS BAUMERT — Universität Kassel, Institut für Physik und CINSaT, D - 34132 Kassel, Germany

In carbon layered materials the electronic subsystem, stimulated by high currents or optical excitations, is strongly coupled to a small set of optical phonons which limits the ballistic conductance. A detailed understanding of phonon decay mechanism is thus essential in improving the performance of carbon based future devices [1,2]. Time-resolved diffracton experiments using x-rays or electrons probes, has become a promising technique to directly provide insights into fundamental dynamics in solids at the microscopic level and on the pico- to subpicosecond timescale [3,4]. In this contribution we present results on photo-induced structural dynamics in single layer nano-crystalline graphene [5] obtained with our compact and well characterized Ultrafast Electron Diffractometer [6] and discuss the influence of the main structural properties for our results[7].

- [1]T.Kampfprath et.al.,Phys.Rev.Lett.95,187403(2005)
- [2]S.Schäfer et.al.,New J.Phys.13,063030(2011)
- [3]A.H.Zewail,J.Phys.Chem.98,2782-2796(1994)
- [4]B.Siwick et.al.,Science Vol.302,No.5649,1382-1385(2003)
- [5]A.Turchanin,ACS Nano Vol.5,No.5,3896(2001)
- [6]C.Gerbig et.al.,in submission
- [7]S.Morgenstern et.al.,in preparation

Coffee Break, 15min.