FEIS-2

Femtosecond Electron Imaging and Spectroscopy

May 6 -9, 2015
Michigan State University
East Lansing, Michigan
www.feis-2.org

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Condensed Matter Physics :	Phillip Duxbury (Michigan State)
Aberrations and	Martin Berz (Michigan State)
Non-Linear Dynamics :	Kyoko Makino (Michigan State)
Electron Diffraction and	Marty Crimp (Michigan State)
Microscopy :	

Sponsors

FEIS-2 Schedule

Wednesday	**6-May**		**Session: UED/UEM and ultrafast X-ray experiments**
12:30-1:25			***Registration***
1:25-1:30			***Welcome***
1:30-2:00	**Geoffrey Campbell**	LLNL	*Experiments and simulations of phase transformations at similar length and time scales*
2:00-2:30	**Xijie Wang**	SLAC	*Development of ultrafast diffraction and imaging at SLAC*
2:30-3:00	**Jason Tenboer**	Wisconsin-Milwaukee	*Making movies of biological macromolecules using time-resolved serial femtosecond crystallography*
3:00-3:30	**German Sciaini**	Waterloo	*Ultrabright femtosecond electron sources for the study of ultrafast structural dynamics*
3:30-4:00			***Coffee break***
4:00-4:30	**Patrik Grychtol**	Colorado/JILA	*Bright circularly polarized soft X-Ray high harmonics for X-Ray magnetic circular dichroism.*
4:30-5:00	**Gilles Doumy**	Argonne	*Applications of laser streaking at X-ray free electron lasers*
5:00-5:15	**Zhaohan He**	Michigan	*A laser-plasma based particle accelerator for ultrafast electron diffraction applications*
5:15-5:45	**Chong-Yu Ruan**	Michigan State	*High-brightness beamlines for ultrafast microdiffraction and imaging*
5:55			***Transition to bus***
6:00-7:30			***Reception at Broad Museum***

Thursday	**7-May**		***Session: Quantum dynamics and phase transitions I***
8:00-8:30	**Keiichiro Nasu**	IMSS,KEK	*Concepts and perspectives on photoinduced structural phase transitions*
8:30-9:00	**Shinichiro Iwai**	Tohoku	*Strong field effects on organic conductors induced by 1.5-cycle infrared light pulse*
9:00-9:30	**Thomas Devereaux**	Stanford/SLAC	*Floquet-Bloch states and photo-Induced chiral edge modes in monolayer TMDCs*
9:30:10:00	**Hermann Durr**	Stanford/SLAC	*Imaging the ultrafast spin-lattice motion during ultrafast demagnetization of ferromagnets*
10:00-10:30			***Coffee break***
10:30-11:00	**Jim Freericks**	Georgetown	*Pump/probe ARPES in electron-phonon coupled metals*
11:00:11:15	**Bin Hwang**	Michigan State	*Optimal ultrafast laser pulse-shaping to direct photo-induced phase transitions*
11:15-11:30	**Faran Zhou**	Michigan State	*Nonequilibrium quantum dynamics in TMD materials*
11:30-11:45	**Junjie Li**	BNL	*Measuring charge and orbital ordering dynamics in layered manganites*
11:45-12:00	**Tatiana Konstantinova**	Stonybrook/BNL	*Ultrafast structural dynamics in Bi2Sr2CaCu2O8+d under polarized photoexcitation*
12:00-2:00			***Lunch break***
			Session: Surfaces and nanosystems
2:00-2:30	**Hrvoje Petek**	Pittsburgh	*Ultrafast photoemission microscopy: Imaging of electromagnetic fields on the femto-nano scale*
2:30-3:00	**Christoph Lienau**	Oldenburg	*Above-threshold ionization and wavepacket dynamics of Rydberg electrons bound to their image potential in a single metallic nanostructure*

3:00-3:30	**Xuan Wang**	IOP, CAS	*Lattice dynamics in Au thin film and nanoparticles, and PbSe quantum dot studied by ultrafast electron diffraction*
3:30-3:45	**Giulia Mancini**	EPFL	*Characterization and light-induced dynamics of alkanethiol-capped gold nanoparticles supracrystals by small-angle ultrafast electron diffraction*
3:45-4:00	**Sebastian Schramm**	Göttingen	*Developing ultrafast low-energy electron diffraction*
4:00-4:30			***Coffee break***
			Session: Quantum dynamics and phase transitions II
4:30-5:00	**Emanuel Gull**	Michigan	*Diagrammatic Monte Carlo for real-time propagation*
5:00-5:15	**Andrey Antipov**	Michigan	*Exact transient dynamics of the Anderson impurity*
5:15-5:30	**Luca Piazza**	EPFL	*Simultaneous observation of the quantization and the interference pattern of a plasmonic near-field*
6:00			***Bus to Banquet, board at front door of Radisson***
6:30-10:00			***Banquet at Michigan Princess***

Friday	**8-May**		***Session: Electron optics and beam dynamics***
8:00-8:30	**Harald Rose**	Ulm	*Holographic imaging and optical sectioning in the aberration-corrected STEM*
8:30-9:00	**Martin Berz**	Michigan State	*End-to-End simulation of nonlinear UEM beam dynamics*
9:00-9:30	**Ludek Frank**	ISI ASCR	*Ultra-low-energy SEM and STEM*
9:30:10:00	**Phil Duxbury**	Michigan State	*Photoemission and beam physics of space charge dominated electron bunches*
10:00-10:30			***Coffee break***
10:30-11:00	**Jie Chen**	SJTU	*Mapping transient electric fields with picosecond electron bunches*
11:00:11:15	**He Zhang**	JLab	*Traceless totally symmetric tensor based fast multipole method for space charge field calculation*
11:15-11:30	**Gregory Hirsch**	Hirsch Scientific	*Bright and durable field emission source derived from refractory-metal Taylor cones*
11:30-11:45	**Andreas Schroeder**	UI-Chicago	*Oriented single-crystal photocathodes: A route to high-quality electron pulses*
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12:00-2:00			***Lunch break***
			Session: UED/UEM development
2:00-2:30	**Pietro Musumeci**	UCLA	*RF photoinjector based MeV electron microscopy*
2:30-3:00	**Dao Xiang**	SJTU	*MeV UED/UEM development at Shanghai Jiao Tong University*
3:00-3:30	**Valery Dolgashev**	SLAC	*Attosecond Diagnostics of Muti-GeV Electron Beams Using W-Band Deflectors*
3:30-4:00	**Sascha Schäfer**	Göttingen	*Ultrafast transmission electron microscopy based on a laser-driven Schottky field emitter*
4:00-4:30			***Coffee break***
			Session: Femtosecond pulse shaping and control
4:30-5:00	**Josef Frisch**	SLAC	*RF Locking of femtosecond lasers*
5:00-5:30	**Marcos Dantus**	Michigan State	*Coherent control of femtosecond pulses and their applications*
5:30-6:00	**Thomas Weinacht**	Stony Brook	*Electronic and nuclear dynamics in strong field ionization*

Saturday	**9-May**		***Session: New horizons***
8:00-8:30	**Jom Luiten**	Eindhoven	*Ultracold and ultrafast electron beams for diffraction and microscopy*
8:30-9:00	**Rick Kirian**	Arizona State	*Coherent diffractive imaging methods at XFELs*
9:00-9:30	**Martin Centurion**	Nebraska	*Ultrafast electron diffraction from molecules in the gas phase*
9:30-10:00	**Renu Sharma**	NIST	*Development of in-situ environment cell for dynamical imaging and chemical analysis of gas-solid interactions*
10:00-10:30			***Coffee break***
10:30-11:00	**Bryan Reed**	IDES	*(Tutorial) Modifying TEMs for high time resolution*
11:00-11:15	**Sergey V. Baryshev**	Euclid TechLabs	*Compact GHz device enabling stroboscopic laser-free ultrafast transmission electron microscopy*
11:15-11:30	**Jasper van Rens**	Eindhoven	*High quality, high repetition rate, ultrashort electron bunches generated with an RF cavity*
11:30-12:00			***Roundtable discussion***
12:00			***Closing***
12:30			***Transition to bus for Ford Museum***

FEIS-2

EXACT TRANSIENT DYNAMICS OF THE ANDERSON IMPURITY

Andrey Antipov, Qiaoyuan Dong, Emanuel Gull

University of Michigan

We study dynamics of a single Anderson impurity model subject to voltage and thermal quenches. We develop a hybridization expansion diagrammatic Monte Carlo algorithm to describe the exact dynamics of the problem. By including the initial correlations into the problem we describe the destruction of the Kondo state and infer the characteristic time scales of the problem. An interplay between different time scales of spin and charge excitations of the strongly correlated setup as visible in the ultra-fast response is in the focus of our study. We compare our results with frequently employed non-crossing and one-crossing approximations.

COMPACT GHZ DEVICE ENABLING STROBOSCOPIC LASER-FREE ULTRAFAST TRANSMISSION ELECTRON MICROSCOPY

Gwanghui Ha[1], Jiaqi Qiu[1], Chunguang Jing[1], Sergey Antipov[1], June W. Lau[2], Yimei Zhu[3], and **Sergey V. Baryshev**[1]

1. Euclid TechLabs ; 2. Materials Science and Engineering Division, National Institute of Standards and Technology ; 3. Department of Condensed Matter Physics and Materials Science, Brookhaven National Laboratory

Time-resolved experiments looking at fundamental processes of atomic/molecular motion and interaction set challenging requirements: lateral resolution $\sim$ 1 Å and time resolution $\sim$ 100 fs. A combined resolution metrics called space-time resolution (STR) must reach $\sim 10^{-23}$ m·s or smaller. X-ray and electron based methods are best candidates to push the boundaries of STR due to the smallest wavelength of the primary probe. Currently, x-ray instruments such as free electron lasers and high harmonic generation laser microscopes have STR between 10^{-22} and 10^{-20} m·s. However, these have limited accessibility, either because of exorbitant equipment cost or meager beam time allocations. Transmission electron microscope (TEM), being the gold standard technique in spatially-resolved imaging, cannot resolve dynamic events in its default form. Until recently, TEM has been ruled out as a viable time-resolved technique. Successful integration of lasers with TEM created truly new fields called ultrafast and dynamic electron microscopy (UTEM and DTEM). Nevertheless, laser (repetition rate) and photocathode (quantum efficiency) performance limitations and cost seriously limit applications and ubiquity of UTEM and DTEM. Alternatively, a pre-existing electron direct current (dc) beam can be blanked/unblanked with some periodicity and temporal structure of an electron pulse in the sequence. A device and a method for producing ultrashort electron pulses with GHz repetition rates via pulsing a dc beam will be presented. These are based on an electromagnetic-mechanical pulser (EMMP) that consists of a series of transverse deflecting cavities and magnetic quadrupoles. The EMMP modulates and chops the incoming dc beam and converts it into femto- to pico-second electron pulse sequences at $>$ 1 GHz repetition rates, as well as controllably manipulates the resulting pulses in the phase space. The EMMP is largely tunable, i.e. temporal pulse length and repetition rate are both tunable. A generalized matrix description of the EMMP in thin lens approximation will be presented together with particle tracking results. When applied to TEM, a GHz stroboscopic TEM with STR metrics between 10^{-23}-10^{-20} m·s can be realized. Such stroboscopic microscope can be simply based on a traditional TEM platform with a thermionic or field emission electron source with no additional laser and photocathode needed.

EXPERIMENTS AND SIMULATIONS OF PHASE TRANSFORMATIONS AT SIMILAR LENGTH AND TIME SCALES

Geoffrey H. Campbell, Tae-Wook Heo, Joseph T. McKeown, Linh Nguyen, Robert E. Rudd, and Melissa K. Santala

Lawrence Livermore National Laboratory

We have developed time resolved in situ observation methods based on the Dynamic Transmission Electron Microscope (DTEM) to characterize solid - solid and solid - liquid phase transformations in a range of systems, including phase change materials, semiconductors, and metallic alloys and intermetallics. The DTEM provides data from which phase change kinetics can be precisely measured in the nanosecond and nanometer regimes. We use atomistic simulations to explore nucleation and growth rates of new phases for comparison with the experimental measurements. We also use atomistic simulations and experimental measurements to parameterize phase field simulations to capture microstructure development during the phase transformation and its interaction with pre-existing structure. We will discuss insights brought by the synergy of the experiments with the simulations. This work performed under the auspices of the US Department of Energy, Office of Basic Energy Sciences by Lawrence Livermore National Laboratory under contract DE-AC52- 07NA27344.

ULTRAFAST ELECTRON DIFFRACTION FROM MOLECULES IN THE GAS PHASE

Martin Centurion

University of Nebraska - Lincoln

Ultrafast electron diffraction can in principle be used to observe the structure of a molecule as it undergoes a photoreaction, providing images of the initial, intermediate and final states with atomic resolution. Achieving this goal requires overcoming two major challenges: The first challenge is to retrieve the structure directly from the diffraction pattern for molecules that are in the gas phase, and thus have random orientation, and the second is to achieve sufficient temporal resolution to observe the relevant dynamics. The random orientation of molecules can be overcome by diffracting from laser-aligned molecules. We have shown that by using femtosecond laser pulses to impulsively align the molecules, it is possible to capture a field-free diffraction pattern while the molecules are transiently aligned. First experiment were done on a static molecular structure, and we have recently demonstrated imaging of a short lived excited state, with a resolution of about 1 ps. The second challenge is to improve the temporal resolution to the regime of 100 fs in order to map the structural changes as the molecule transitions from the ground to the excited states. This means not only delivering sufficiently short pulses on the target, but also compensating the effect of the velocity mismatch of electrons and laser as they traverse the sample. We are pursuing two approaches for this, one is to use RF compression of 100 keV electron pulses in combination with a tilted laser pulse excitation to overcome the velocity mismatch, the second is to use relativistic (MeV) electron pulses where the velocity mismatch is negligible.

MAPPING TRANSIENT ELECTRIC FIELDS WITH PICOSECOND ELECTRON BUNCHES

Jie Chen

Shanghai Jiao Tong University

Transient electric field (TEF), which is an important but hardly explored parameter of laser plasmas, can now be diagnosed experimentally with combined ultrafast temporal resolution and field sensitivity using femtosecond to picosecond electron or proton pulses as probes. However, it is challenging to simultaneously record both the global and local TEF features owing to poor high spatial resolution. We present a combined diagnostic of electron schlieren-type radiography and the Abel inversion method to directly measure the 3D TEFs using simultaneous 80-micron spatial and 3.7-picosecod temporal resolutions. We show that the TEFs, which are induced by femtosecond laser pulses illumination on an aluminum foil at 1.9E12 W/cm^2 and are on the order of 1E5 V/m, display a unique "peak-valley" structure along the outgoing direction of the emitted electron cloud, a feature that has never been reported. The field map also naturally leads to a quantitative characterization of the transient density distribution of the electron cloud, which has a charge density of -0.04 C/m^3 and expands at a speed of 4E6 m/s. By combining the micron spatial and picosecond temporal resolutions, this type of radiography using charged particle pulses should enable the mapping of various fast-evolving field structures, such as those found in plasma-based particle accelerators.

COHERENT CONTROL OF FEMTOSECOND PULSES AND THEIR APPLICATIONS

Marcos Dantus

Michigan State University

Femtosecond pulses allow the unprecedented opportunity to probe matter on a timescale that is faster than atomic motion, as recognized by the 1999 Nobel Prize for Femtochemistry. The next frontier is to elucidate and control laser-matter interactions. This talk will present efforts in the Dantus group towards achieving this ambitious goal. The first step in this quest is to control the laser source itself. I will describe how coherent control has led to automated pulse compression down to near single-cycle 3.8 fs pulses. The talk will cover applications related to (a) improving nonlinear optical microscopy by using shaped laser pulses; (b) nanoplasmonic dephasing in single, paired, and dendritic chains of nanoparticles; and (c) standoff detection of explosives.

FLOQUET-BLOCH STATES AND PHOTO-INDUCED CHIRAL EDGE MODES IN MONOLAYER TMDCS

Tom Devereaux

Stanford and SLAC National Accelerator Laboratory

In this talk I will discuss manipulation of electron dynamics via off-resonant application of light, via dynamical breaking of symmetries & dynamical modulation of the electronic band structure. I will present detailed predictions of dynamic band structure manipulation in pump-probe angle-resolved photoemission response in transition metal dichalcogenides. The results show a dynamical breaking of valley degeneracy/TRS via circularly-polarized light, a scheme to achieve a condensed-matter realization of light-induced chiral edge modes for weak pump fields, and the development of topological Wannier Stark physics at strong pump fields. This provides a rich phase diagram of light-induced Floquet topological phases.

ATTOSECOND DIAGNOSTICS OF MUTI-GEV ELECTRON BEAMS USING W-BAND DEFLECTORS

Valery Dolgashev, Paul Emma, Massimo Dal Forno, Jeffrey Neilson

SLAC National Accelerator Laboratory

Performance of the LCLS free electron laser is determined by the properties of extremely short electron bunches. The multi-GeV electron bunches in LCLS are less than 100 fs long. Optimization of beam properties and understanding of free-electron laser operation require electron beam diagnostics with time resolution of less than 10 fs. These were achieved by using the 2-m-long X-band RF deflector. As for now, the performance and utility of this deflector remains unchallenged. We propose the next generation of this time-resolved beam diagnostic with improvements in time resolution by an order of magnitude, possibly down to a few hundred attoseconds at 15 GeV. We expect that, as with the current X-band deflector, it will allow smooth commissioning, operation and further improvement of future free electron lasers. This 10-fold increase of the timing resolution could, in principal, be achieved by scaling the existing X-band system. In that case, the new deflector system would be $\sim$20 meter long and powered by ten 50 MW X-band klystrons. We see this as an impractical solution and instead propose to increase the operating frequency of the deflector from 11 GHz to $\sim$90 GHz. A 1-meter-long deflector could provide 10-times better temporal resolution down to $\sim$0.4 fs, or less. We envision two practical options: one powered by $\sim$10-MW 90-GHz rf source, and another one, passive: powered by the bunch's self-wake fields. In this talk we will review SLAC's experience with X-band deflectors and 100 GHz high-gradient accelerating structures, then will discuss the potential of 90 GHz rf deflectors.

APPLICATIONS OF LASER STREAKING AT X-RAY FREE ELECTRON LASERS

Gilles Doumy

Argonne National Laboratory

X-ray radiation has been long used to address selectively atoms and to yield structural information with atomic precision. The advent of X-ray Free Electron Lasers (XFEL) is revolutionizing the field of time resolved x-ray techniques. The availability of tunable pulses ranging from the soft to the hard x-ray region, and lasting only few tens of femtoseconds, or perhaps less, is enabling access to unprecedented temporal resolution. However, knowledge of the temporal properties of the x-ray pulses is poor, and synchronization to external sources introduces a timing jitter that dominates the fast dynamics and needs to be corrected for every shot. Manipulating electrons generated by x-ray induced ionization and subsequent relaxation processes represents a powerful concept to gain access to those properties. Specifically, using laser streaking techniques developed by the attosecond community, one can measure the pulse duration, and possibly improve the temporal resolution of pump probe experiments where electrons are collected to follow the processes by use of a self-referencing measurement. Illustration is presented following Auger decay in the time domain.

IMAGING THE ULTRAFAST SPIN-LATTICE MOTION DURING ULTRAFAST ALL-OPTICAL SWITCHING OF FERROMAGNETS

Hermann Durr

SLAC National Accelerator Laboratory

Understanding the ultrafast interplay between charge, magnetic and lattice degrees of freedom is central to gaining control of condensed matter phenomena as diverse as insulator-metal transitions and magnetic switching. Although magnetism in metallic systems is expected to couple only weakly to phonons, the observed ultrafast demagnetization and all-optical magnetic switching demonstrate just the opposite. Femtosecond soft x-ray pulses from the Linac Coherent Light Source, offer the unique opportunity to image in realtime the ultrafast spin dynamics that leads to magnetization reversal. Hard x-rays and fs electron pulses enable first glimpses at the laser-induced lattice motion revealing unexpected spin-lattice relaxation channels. Understanding the evolving spin-lattice motion on the fs time and nm length scales associated with the exchange interaction opens new possibilities of engineering angular momentum relaxation channels in magnetic systems.

[1] E. Beaurepaire, et al., Phys. Rev. Lett, 76, 4250 (1996).
[2] C.D. Stanciu et al., Phys. Rev. Lett, 99, 047601 (2007).
[3] S. Mangin, et al. Nature Materials 13, 286 (2014).
[4] C. E. Graves, A. H. Reid et al., Nature Materials, 12, 293 (2013).

PHOTOEMISSION AND BEAM PHYSICS OF SPACE-CHARGE DOMINATED ELECTRON BUNCHES

Phillip M. Duxbury, Jenni Portman, Chong-Yu Ruan, Martin Berz, Kyoko Makino, He Zhang, Zhensheng Tao

Michigan State University

Efficient computational methods to treat space charge effects are essential to accurate simulation of electron bunches with high electron density. High electron density is typical at the photocathode, in RF cavities and at the focal plane, where N-particle simulations are critical. Stochastic space charge effects occurring at photoemission place a fundamental limitation on UEM and UED resolution. By combining N-particle simulations and analytic Gaussian calculations we provide estimates of the practical limits of UEM and UED resolution as a function of beam energy.

[1] Z. Tao, et al., Journal of Applied Physics 111(4), 044316, pages 1-10 (2012).
[2] J. Portman, et al., Applied Physics Letters 103, 253115 (2013)
[3] J. Portman, et al., J. Appl. Phys. 116, 174302 (2014).
[4] He Zhang, Jenni Portman, Zhensheng Tao, Phillip Duxbury, Chong-Yu Ruan, Kyoko Makino and Martin Berz. "The Differential Algebra Based Multiple Level Fast Multipole Algorithm for 3D Space Charge Field Calculation and Photoemission Simulation". Proceedings of the 9th International Conference on Charged Particle Optics, Brno September, 2014.

ULTRA-LOW-ENERGY SEM AND STEM

Ludek Frank

ISI ASCR, Brno, Czech Republic

The general trend for reducing the energies of primary electrons in electron microscopy, which is promising from the point of view of desirable enhancement of the scattering rate of electrons in targets, had faced a gradual deterioration of the image resolution. Biasing the sample to a high negative voltage and making the electrons arbitrarily slow solely on and inside the sample has shown itself to be far more feasible than originally expected. The most important thing is that the fundamental aberration coefficients (spherical and chromatic) of a combination of an objective lens and an immersion electrostatic lens formed by the biased sample decrease with the decreasing landing energy of the electrons. As a result, the spot size in scanning systems may become nearly independent of the landing energy of the electrons. The requirements placed on samples are strict but feasible: bulk samples observed in the reflection mode have to be flat (but not polished), while thin films for the transmission mode should not exceed 10 nm in thickness. Detection of signal electrons is greatly facilitated by acceleration of both reflected and transmitted electrons in the field of the biased sample and their collimation toward the optical axis. Naturally, the reflected axial ray usually escapes detection through the detector bore, so special arrangements are needed (such as separated primary and signal columns with a beam splitter in LEEM). The interaction of slow electrons is not only more intensive than that at standard energies, but even scattering phenomena appear which are not otherwise available. The wavelength of the electrons approaches the interatomic distances, so diffraction and interference phenomena become sources of image contrasts even in signals acquired with single-channel detectors in the SEM. Moreover, nontrivial distribution of the local density of states close above the vacuum level makes the electron reflectivity dependent on energy in a way specific to a crystallographic system and its orientation, so reflectivity may serve as a fingerprint of the local crystallinity. STEM observation of 2D crystals at units of eV and measurement of their reflectivity and transmissivity provide information supporting calculations of their electronic structure. Biological specimens in the form of ultrathin sections exhibit very high contrasts showing complete structure details without the addition of the heavy metal salts that highlight only some of the details. The benefits of very-low-energy EM are still being uncovered after having been in routine use for several years.

PUMP/PROBE ARPES IN ELECTRON-PHONON COUPLED METALS

Jim Freericks[1], Lex Kemper[2], Brian Moritz[3], Michael Sentef[4] and Tom Devereaux[3]

1. Georgetown University ; 2. Lawrence Berkeley Laboratory ; 3. Stanford University and SLAC ; 4. Hamburg University

In this talk, I will summarize recent work we have done in studying the properties of electron-phonon coupled metals that are examined with pump/probe photoemission studies. In the low fluence regime, one can map time-dependent data within the so-called “phonon window”(where the relaxation has a bottleneck and the system relaxes slowly) onto the equilibrium dynamics. As the fluence is increased, the nonequilibrium driving starts to change the density of states and the scattering, but it does so in a way that preserves the integrated strength of the electron-phonon coupling due to a nonequilibrium sum rule. When one goes into the superconducting state, a moderate fluence can be employed to directly excite the Higgs (or amplitude) mode of the superconductivity, whose dynamic oscillations are correlated with the transiently evolving superconducting gap. We will also discuss implications of this work for experiments.

RF LOCKING OF FEMTOSECOND LASERS

Josef Frisch, Karl Gumerlock, Justin May, Steve Smith

SLAC National Accelerator Laboratory

In RF gun based femtosecond electron imaging systems many experiments require low noise timing control between a pump laser and the electron beam. As the beam time is influenced by both the RF gun photocathode laser and the gun RF fields, precision timing control between the laser and the gun RF fields is needed. We discuss RF based laser timing control, and the system being used in the SLAC ASTA femtosecond electron diffraction system. This system has been used for 100 femtosecond scale timing experiments.

BRIGHT CIRCULARLY POLARIZED SOFT X-RAY HIGH HARMONICS FOR X-RAY MAGNETIC CIRCULAR DICHROISM

Patrick Grychtol

JILA - University of Colorado

Ultrafast light sources based on the high-harmonic up-conversion of femtosecond laser pulses have been successfully employed to access resonantly enhanced magnetic contrast at the M absorption edges of the 3d ferromagnets Fe, Co and Ni in a table-top experimental setup. Thus, it has been possible to study element-specific dynamics in magnetic materials at femtosecond time scales in a laboratory environment, providing a wealth of opportunities for a greater fundamental understanding of correlated phenomena in solid-state matter. However, these investigations have so far been limited to linear polarized higher harmonics, since most techniques by which circular soft x-rays can be generated are highly inefficient reducing the photon flux to a level unfit for scientific applications. Besides presenting key findings of our ultrafast studies on charge and spin dynamics, we introduce a simple setup which allows for the efficient generation of circular harmonics bright enough for x-ray magnetic circular dichroic. Our work thus represents a critical advance that makes possible element-specific imaging and spectroscopy of multiple elements simultaneously in magnetic and other chiral media with very high spatial and temporal resolution, using tabletop-scale setups.

DIAGRAMMATIC MONTE CARLO FOR REAL-TIME PROPAGATION

Emanuel Gull, Guy Cohen, Andrey Antipov

University of Michigan, Columbia University

We present diagrammatic Monte Carlo methods for the real-time dynamics of quantum impurities, introduce the concept of bold-line Monte Carlo methods, and present a formalism to obtain spectral functions from an auxiliary lead formalism. Using these methods we show results for nonequilitrium systems, including the voltage splitting of the Kondo peak.

A LASER-PLASMA BASED PARTICLE ACCELERATOR FOR ULTRAFAST ELECTRON DIFFRACTION APPLICATIONS

Zhaohan He[1], John Nees[1], Roye Clarke[2], Karl Krushelnick[1], and Alec Thomas[1]

1. Center for Ultrafast Optical Science, University of Michigan ; 2. Department of Physics, School of LSA, University of Michigan

Femtosecond bunches of electrons with relativistic to ultra-relativistic energies can routinely be produced by laser plasma wakefield accelerators (LWFA) driven by 10-100 TW, high peak power laser systems. Scaling the electron energy down to sub-relativistic and MeV level is possible using a sub-TW laser system. Such an electron source can be a potential candidate for ultrafast electron diffraction (UED) applications, due to the intrinsic short bunch duration and perfect synchronization with the optical pump. We will present results of transmission electron diffraction from a crystalline sample, using LWFA electrons driven by 8-mJ, 35-fs laser pulses at 0.5 kHz. The accelerated electrons are collimated with a solenoid magnetic lens. Single-shot diffraction patterns are obtained from a single-crystal gold foil in a proof-of-principle experiment to demonstrate the beam quality and charge. Temporal characterization of the electron bunches using a non-equilibrium plasma reveals the linear energy chirp, which can be utilized for further electron bunch compression or time-resolved measurement in streak mode. Preliminary pump probe studies on gold foil will also be presented.

BRIGHT AND DURABLE FIELD EMISSION SOURCE DERIVED FROM REFRACTORY-METAL TAYLOR CONES

Gregory Hirsch

Hirsch Scientific

A novel method for in-situ creation of field emission tips having desirable operational characteristics is described. In this process, the end of a refractory metal wire is melted in vacuum using a focused laser beam. A high positive potential simultaneously applied to the wire is used to electrostatically form a Taylor cone from the liquid meniscus on the wire end. This experimental configuration can be regarded as an unusual type of Liquid Metal Ion Source (LMIS). Upon cessation of the laser power, the Taylor cone freezes almost instantaneously. Extremely high radiative and conductive cooling assures that solidification of the cone is exceptionally rapid, resulting in a frozen structure having a shape and surface smoothness very closely matching that of the initial liquid Taylor cone. Very uniform and sharp metal cones have been generated from a variety of refractory metals having melting points as high as tungsten. The frozen Taylor cones can be subsequently employed as field emitters by simply reversing the polarity of the applied voltage. This type of field emission source may find widespread application for both continuously operating electron guns, as well as with pulsed sources. A feature of this technology is the ability to rapidly regenerate the frozen Taylor cones after they undergo some degradation during use. When employed for ultrafast applications using laser-assisted field emission, the capability to readily restore the emitter to pristine condition in-situ may permit the attainment of emission currents near or above damage threshold, without undue concern regarding reduced field-emitter lifetimes. The large characteristic opening angle of Taylor cone structures (near 98.6 degrees full angle) results in electron emission being electrostatically directed into a relatively low-divergence forward directed beam. This may be advantageous for achieving low emittance and bright electron sources. The current state of this technology will be described, as well as plans for further development.

OPTIMAL ULTRAFAST LASER PULSE-SHAPING TO DIRECT PHOTO-INDUCED PHASE TRANSITIONS

Bin Hwang, Jenni Portman, Phillip Duxbury

Michigan State University

Photo-induced phase transitions (PIPT) in quantum and/or complex materials are the epitome of challenging non-equilibrium many-body phenomena, that also have a wide range of potential applications. We present a computational approach to finding optimal ultrafast laser pulse shapes to control the outcome of pump-probe PIPT experiments. The Krotov approach for Quantum optimal control theory (QOCT) is combined with a Keldysh Green's function calculation to describe experimental outcomes such as photoemission, transient single particle density of states and optical responses. Results for a simple model charge density wave system will be presented.

STRONG FIELD EFFECTS ON ORGANIC CONDUCTORS INDUCED BY 1.5-CYCLE INFRARED LIGHT PULSE

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Ultrafast control of conduction and magnetic properties in strongly correlated systems has been extensively studied from the perspective of photoinduced insulator-to-metal transitions, or, equivalently, the "melting" of electronic orders in Mott insulators, charge ordered systems, and charge/spin density wave materials. On the other hand, the development of strong electric fields ($>$ MV/cm) of few-cycle optical pulses and recent theoretical studies using dynamical mean-field theory (DMFT) [1] suggest that extreme non-equilibrium electronic states such as charge localization, negative temperatures, and repulsive-attractive conversion can be achieved. Among those highly non-equilibrium phenomena, reducing the intersite transfer integral t by modulating the site energy under high-frequency [$\omega > t/(\hbar)$] continuous-wave (CW) and pulsed AC electric fields E(ω) have attracted attention for $>$ 30 years[2, 3]. Such an intense modulation of the electronic structure driven by a strong electric field, referred to as "dynamical localization", provides a new strategy for controlling charge motion in strongly correlated materials, which have a competing energy balance between the on-site or intersite Coulomb repulsion and t. Here we describe a charge localization induced by the 9.3 MV/cm instantaneous electric field of a 1.5 cycle (7 fs) infrared pulse in an organic conductors alpha-$(ET)_2I_3$ (ET; bis[ethylenedithio]-tetra-thiafulvalene)[4], $(TMTTF)_2AsF_6$ (TMTTF; tetramethyltetrathiafulvalene). In alpha–$(ET)_2I_3$, a large reflectivity change of $> 25\%$ and a coherent charge oscillation along the time axis reflect the opening of the charge ordering gap in the metallic phase. This optical freezing of charges, which is the reverse of the photoinduced melting of electronic orders, is attributed to the 10% reduction of t driven by the strong, high-frequency electric field. In $(TMTTF)_2$ AsF_6, such reduction of t has been demonstrated by the analysis of the transient reflectivity spectra using Drude-model. Dependence on the field intensity and the direction of electric field can be also understood in terms of dynamical localization.

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COHERENT DIFFRACTIVE IMAGING METHODS AT XFELS

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In this talk I will discuss some unique ways in which coherent diffraction data from bioparticles in nanocrystal, solution, and gas phases can be obtained at x-ray free- electron lasers (XFELs). For example, the continuum of diffraction intensities that are observed when protein nanocrystals are illuminated with coherent femtosecond XFEL pulses gives rise to information in regions between the usual Bragg peaks. Such information can be used to determine diffraction phases without prior information, and without resolution restrictions. Similarly, intensity correlations can be observed in what might appear to be isotropic solution scattering, because XFELs effectively freeze atomic motion and capture intensity fluctuations that are weak and persist only for a brief period of time. Intensity correlations contain a wealth of structural information from hydrated biomolecules at room temperature, well beyond the level of information that conventional solution scattering contains. The key to gathering the needed data at XFELs for these methods and others often lies in the sample delivery method. Numerous innovations have been required in order to replenish precious sample rapidly, since in most bioimaging schemes each target is vaporized and must be rapidly replaced with a new one before the next XFEL pulse arrives. Liquid jets, for example, were key to the development of methods such as serial femtosecond crystallography and sub-picosecond time- resolved wide-angle solution scattering. As time permits, I will discuss some of our recent efforts to improve on sample delivery methods, with a look toward the upcoming generation of XFELs that will produce pulses at MHz repetition rates.

ULTRAFAST STRUCTURAL DYNAMICS IN $BI_2SR_2CACU_2O_{8+\delta}$ UNDER POLARIZED PHOTOEXCITATION

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We report the structural dynamics of high-T_c superconductor $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi 2212) induced by femtosecond 1.5eV photoexcitation using ultrafast electron diffraction (UED). A pronounced feature of Bi 2212, similar to other high-T_c cuprates, is its ab-plane anisotropy of electronic properties (a superconducting gap below T_c and a pseudogap in the normal state) due to the symmetry of d-wave with nodes near the bisectors of Cu-O bonds. The electron coupling to half-breathing in-plane Cu-O stretching and out-of-plane O-buckling phonon modes is one of the possible explanations of electron glueing in a superconducting state. Thus, the strength of the coupling is predicted to have the d-wave symmetry as well. In current research the use of the pump beam polarized along a- and b- axes and at 45 degree to them allows to investigate the anisotropy of lattice response to the photoexcitation. The lattice dynamics along different crystal axes has been examined while separating signals from the basic crystal structure and from the incommensurate modulation along b-axis. This separation helps us to distinguish between electronic and lattice origin of the structure modulation, which is still an open question. The dynamics was monitored up to 25 ps after excitation with 0.4 ps resolution. We will present the intensity change for the diffraction peaks, corresponding to dynamics of different directionalities of the crystal structure under the photoexcitation with variable polarization, as well as the analysis of the intensity trend. Detailed data analysis and challenges in quantification of our RF-MeV UED experiments will be discussed. Work at Brookhaven National Laboratory was supported by the US DOE, Office of Science, Basic Energy Sciences, under Contract No. DE-SC0012704.

DEVELOPMENTS OF 4D-TRANSMISSION ELECTRON MICROSCOPE AND ANISOTROPIC LATTICE DYNAMICS OF MULTI-WALLED CARBON NANOTUBES

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Institute of Physics, Chinese Academy of Science

Recent advances in the four-dimensional ultrafast transmission electron microscope (4D-UTEM) with combined spatial and temporal resolutions have made it possible to directly visualize structural dynamics of materials at the atomic level. In this paper, we report on our development on a 4D-UTEM which can be operated properly on either the photo-emission or the thermionic mode. We demonstrate its ability to obtain sequences of snapshots with high spatial and temporal resolutions in the study of lattice dynamics of the multi-walled carbon nanotubes (MWCNTs). This investigation provides an atomic level description of remarkable anisotropic lattice dynamics at the picosecond timescales. Moreover, our UTEM measurements clearly reveal that distinguishable lattice relaxations appear in intra-tubular sheets on an ultrafast timescale of a few picoseconds and after then an evident lattice expansion along the radical direction. These anisotropic behaviors in the MWCNTs are considered arising from the variety of chemical bonding, i.e. the weak van der Waals bonding between the tubular planes and the strong covalent sp2-hybridized bonds in the tubular sheets.

MEASURING CHARGE AND ORBITAL ORDERING DYNAMICS IN LAYERED MANGANITES

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1. Brookhaven National Laboratory ; 2. Shanghai JiaoTong University ; 3. SLAC National Accelerator Laboratory

We report a study of the dynamics of electronic order in a photoinduced phase transition in bi-layered $LaSr_2Mn_2O_7$ using time resolved MeV electron diffraction at 77K. We found that melting of the orbital order (OO) takes place with a similar time constant of the charge order (CO). The time constant and order parameter during the phase transition is investigated at various sample temperature and pump fluence. No substantial difference in dynamic behavior is observed. Saturation and threshold fluence are obtained by fitting the order parameter dependence on pump fluence. By quantitative comparison between experimental electron diffraction patterns and simulations based on dynamic diffraction theory, we found that the charge disproportionation on the two Mn ions is very small and the intensity of the CO peaks can be mostly attributed to the secondary harmonics of orbital ordering in our MeV-UED measurements. We further analyzed the influence of different phonon models on diffraction intensities in the system. The results suggest that Jahn-Teller distortion dominates the photo-induced structure phase transition. Our study sheds light on the various and often intertwined degrees of freedom and their responses to external perturbations in strongly correlated electron systems. The authors would like to thank S.-W. Cheong for providing the samples. This research was supported by the US Department of Energy, Basic Energy Sciences, under Contract No: DE-SC0012704.

ABOVE-THRESHOLD IONIZATION AND WAVEPACKET DYNAMICS OF RYDBERG ELECTRONS BOUND TO THEIR IMAGE POTENTIAL IN A SINGLE METALLIC NANOSTRUCTURE

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1. Institute of Physics, Carl von Ossietzky University ; 2. Center of Inferface Science, Carl von Ossietzky University

In atomic and molecular physics, the study of strong-field phenomena has been a major research focus since many years, including a wealth of fundamental processes such as high-harmonic generation, attosecond streaking and the generation of attosecond electron wavepackets for tomographic imaging of molecular orbitals. During the last few years, single metallic nanostructures have developed into a veritable testbed for translating concepts from strong-field physics into the realms of nano-optics and solid state science. In this talk, I will discuss recent experimental of our own group probing the dynamics of photoemission from sharp metallic nanotapers driven by intense few-cycle laser pulses. I will demonstrate how to use to optical near-field at the taper apex to accelerate and steer ultrashort electron pulses [1] and how to manipulate such electron pulses by means of the carrier-envelope phase of the driving laser [2]. I will then discuss recent experimental studies of two-color photoemission from a gold taper in which we observed, for the first time to our knowledge, the coherent dynamics of Rydberg electron wavepackets, bound to their image potentials and nanolocalized to a single nanostructure. When illuminating a sharp gold tip with a an apex radius of 10 nm with a sequence of two time-delayed pulses centered at 600 nm and 1.6 m, we observe pronounced temporal modulations in the photoemission yield. The periodicity of this modulation is in the range of a few tens of fs and depends on the time-ordering of the pulses. The origin of these oscillations is revealed by recording intensity-dependent kinetic energy spectra of the released photoelectrons for optical excitation of the tip with two temporally overlapping visible (600 nm) and near-infrared (1600 nm) pulses. The photoelectron spectra are governed by above-threshold ionization and reveal a rich and surprising fine-structure. Our results provide strong evidence that the field enhancement at the tip apex confines the Rydberg orbits locally in all three dimensions to a volume of a few nm3 only and results in high ATI orders and large kinetic energy photoelectrons for laser intensities well below the destruction level of the metallic nanostructure. Thus they demonstrate a new level of quantum control over the motion of electrons in strong nanolocalized laser fields and open the door to time-resolved photoelectron spectroscopy of single nanostructures with ultrahigh time resolution.

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ULTRACOLD AND ULTRAFAST ELECTRON BEAMS FOR DIFFRACTION AND MICROSCOPY

Jom Luiten

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At Eindhoven University of Technology we are developing new methods for generating beams for ultrafast electron diffraction and microscopy. By femtosecond photoionization of a laser-cooled gas we produce highly charged, highly coherent electron bunches, which ultimately should enable single-shot electron diffraction of macromolecules. Using a simple graphite sample we recently generated the first diffraction patterns with the ultracold electron source, which provided direct evidence that the source has the required coherence. We are currently characterizing the longitudinal phase space of the ultracold electron bunches and we are preparing crystalline samples of the 2D membrane protein hydrophobin. Progress will be reported. By implementing an ultrafast, RF-cavity-based beam blanker in a FEI Tecnai TEM we will realize 'stroboscopic' ultrafast electron microscopy with substantially improved performance in terms of spatial resolution, temporal resolution, and data acquisition time. A specially designed compact and energy efficient RF cavity is currently being prepared for installation in the TEM. Progress will be reported.

CHARACTERIZATION AND LIGHT-INDUCED DYNAMICS OF ALKANETHIOL-CAPPED GOLD NANOPARTICLES SUPRACRYSTALS BY SMALL-ANGLE ULTRAFAST ELECTRON DIFFRACTION

Giulia Mancini

EPFL, Lausanne, Switzerland

Monodisperse nanoparticles coated with organic ligands form two- and three-dimensional supracrystals, which can be regarded as new complex materials, with applications in optoelectronics and catalysis [1]. The knowledge of the arrangement of the nanoparticles core and their ligands, and its dynamical control upon photo-irradiation, represents the next frontier in the full characterization of these supracrystals. Here we present a femtosecond (fs)-resolved electron diffraction study of a two-dimensional supracrystal of alkanethiol-coated gold nanoparticles with femtosecond time, sub-nanometer spatial resolution and sensitivity down to the carbon and hydrogen atoms in the ligands [2, 3]. Combining dynamical diffraction experiments with static real-space imaging and simulations, we performed ultrafast-electron diffractive imaging obtaining a fs/Å-resolved movie of the evolution of the gold NPs and their organic ligands. We show that both disordering and ordering dynamics of the supracrystal can be achieved upon ultrafast laser excitation, demonstrating that self-organization can be triggered by light. These results reveal the role of thermal disorder in the supracrystal and the ability of ultrafast photo-excitation to induce an ordering transition of the organic ligands in the picosecond (ps) time scale.

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RF PHOTOINJECTOR BASED MEV ELECTRON MICROSCOPY

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Using relativistic electrons from RF photoinjectors offers the possibility to improve temporal resolution of single-shot transmission electron microscopes by three orders of magnitude over current state of the art. Here we discuss our progress in developing a new kind of tool based on this high brightness electron source. In particular we'll present our recent results in the condenser lens stage and our plans to further expand the capabilities of the instrument.

CONCEPTS AND PERSPECTIVES ON PHOTOINDUCED STRUCTURAL PHASE TRANSITIONS

Keiichiro Nasu

IMSS, KEK

We, at first, briefly see the early experimental history of photo-induced structural phase transition study, in connection with the TTF-CA, being the mother material, connotes this new and exciting non-equilibrium critical phenomenon triggered only by the visible lights irradiation. [1-3] We will also see recent developments in the way of observation, not only by the visible light spectroscopy, but also by the electron beam spectroscopy and STM methods in connection with the recent discovery of photo-induced " Diaphite (sp3) " domain in the graphite crystal, which originally has only the sp2 bonds. [4] Finally, we see the electronic excited domain formation, not by the direct visible light excitations, but by the large amplitude lattice displacement pulses (THz pulses) and their nonlinear propagations.

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ULTRAFAST PHOTOEMISSION MICROSCOPY: IMAGING OF ELECTROMAGNETIC FIELDS ON THE FEMTO-NANO SCALE

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By combining ultrafast laser photo excitation with photoelectron emission microscopy we achieve simultaneous femtosecond temporal and nanometer spatial resolution of electromagnetic fields at nanostructured metal-semiconductor surfaces. A variety of nano structured surfaces are formed by deposition of Ag films on Si(001) substrates under UHV conditions. The nanostructured samples can be imaged with $<$10 nm spatial resolution either by photoemission electron microscopy (PEEM) using UV laser or Hg lamp excitation sources, or by low energy electron microscopy (LEEM). Femtosecond laser excitation in the UV-VIS energy range excites coherent electromagnetic modes of the sample, which are dominated by the localized and propagating surface plasmons. The regions of the sample with high field enhancement emit photoelectrons by nonlinear two- or three-photon photoemission process. The photoelectrons are imaged by the PEEM optics to generate spatial maps of the plasmonic field enhancement in the sample. Tuning of the delay between identical femtosecond pump-and probe pulses generates movies of the spatiotemporal evolution of the plasmon fields. I will cover the principles of time-resolved PEEM imaging of surface electromagnetic fields and report on recent measurements with a broadly tunable femtosecond laser source.

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SIMULTANEOUS OBSERVATION OF THE QUANTIZATION AND THE INTERFERENCE PATTERN OF A PLASMONIC NEAR-FIELD

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Surface plasmon polaritons can confine electromagnetic fields in subwavelength spaces and are of interest for photonics, optical data storage devices and biosensing applications. In analogy to photons, they exhibit wave-particle duality, whose different aspects have recently been observed in separate tailored experiments. Here we demonstrate the ability of ultrafast transmission electron microscopy to simultaneously image both the spatial interference and the quantization of such confined plasmonic fields. Our experiments are accomplished by spatiotemporally overlapping electron and light pulses on a single nanowire suspended on a graphene film. The resulting energy exchange between single electrons and the quanta of the photoinduced near-field is imaged synchronously with its spatial interference pattern. This methodology enables the control and visualization of plasmonic fields at the nanoscale, providing a promising tool for understanding the fundamental properties of confined electromagnetic fields and the development of advanced photonic circuits.

(TUTORIAL) MODIFYING TEMS FOR HIGH TIME RESOLUTION

Bryan W. Reed and Daniel J. Masiel

Integrated Dynamic Electron Solutions, Inc.

This tutorial will describe modifications that convert a conventional TEM to a stroboscopic ultrafast TEM (UTEM), single-shot or movie-mode Dynamic TEM (DTEM), or hybrid instrument capable of both modes of operation (UDTEM). Piping pulsed lasers into a microscope is just one step in this process. Experimental demands on reliability, stability, performance, and usability mean that every aspect of the modification-laser ports, cathode selection, column modifications, vibration isolation, laser beam profile and alignment control, data acquisition, electronics, and software-must be optimized not just individually but also in the context of the full integrated system. Without such attention to detail, instrument development time can easily stretch into years, and the resulting instrument can be frustratingly saddled with mundane yet crucial usability issues. Such issues can be avoided with forethought and good system-level design, yielding a complex, flexible, high-performance, yet relatively easy-to-use instrument. This is especially true for movie-mode DTEM, which requires both an advanced arbitrary-waveform laser system and a perfectly synchronized high-speed deflector system. In closing, we will discuss nanosecond/microsecond-scale pulse shaping, complementing the discussion of femtosecond/picosecond-scale pulse shaping also taking place at this workshop.

HIGH-QUALITY, HIGH-REPETITION RATE, ULTRASHORT ELECTRON BUNCHES GENERATED WITH AN RF-CAVITY

Jasper van Rens[1], Wouter Verhoeven[1], Erik Kieft[2], Jim Franssen[1], Peter Mutsaers[1] and Jom Luiten[1]

1. Coherence & Quantum Technology Group, Eindhoven University of Technology ; 2. FEI Company

In collaboration with FEI Company, we are studying the possibility of using microwave TM110 streak cavities in combination with a slit, to chop a continuous electron beam into 100 fs electron bunches. We have shown that this can be done with minimal increase in transverse emittance and longitudinal energy spread. Furthermore, these bunches are created at a repetition rate of 3 GHz. Accurately synchronized to a mode-locked laser system, this allows for high-frequency pump-probe experiments with the beam quality of high-end electron microscopes.

At Eindhoven University of Technology, we will soon implement such a cavity in a 200 keV Tecnai, which should result in high-frequency ultrafast (S)TEM with sub-ps time-resolution while maintaining the atomic spatial resolution of the TEM.

HOLOGRAPHIC IMAGING AND OPTICAL SECTIONING IN THE ABERRATION-CORRECTED STEM

Harald H Rose

Ulm University

The correction of spherical aberration enables efficient holographic imaging in the scanning transmission electron microscope (STEM) if the energy width of the incident electron beam is sufficiently reduced. Holographic imaging implies that the Fourier transform of the image is linearly related with the elastic scattering amplitude of the object. Effective optical sectioning can be realized in the aberration-corrected STEM by employing "holographic" phase-contrast imaging. This imaging mode requires a segmented bright-field detector and a Fresnel phase plate which can be formed with a sufficient degree of accuracy by adjusting appropriately the third-order spherical aberration and the defocus of the corrected objective lens. By subtracting the signals of the annular detector segments covering the region of destructive interference of the scattered wave with the non-scattered wave from that recorded by the annular segments covering the regions of constructive interference, we obtain a pure phase contrast image which may be conceived as a holographic image because the terms of the intensity which depend quadratic on the scattering amplitude cancel out. Theoretical results will be presented which demonstrate the feasibility of the proposed method. In particular, the method enables the transfer of spatial frequencies over a large range which exceeds significantly that of conventional phase contrast imaging.

HIGH-BRIGHTNESS BEAMS FOR ULTRAFAST MICRODIFFRACTION AND IMAGING

Chong-Yu Ruan

Department of Physics and Astronomy, Michigan State University

Currently the ultrafast electron diffraction (UED) with 10^3-10^5 electrons per pulse has achieved sub-picosecond temporal resolution and atomic resolution. However, direct ultrafast imaging of a nanometer scale specimen through coherent single-particle diffraction has not been achieved largely due to insufficient intensity when tuned to a coherence length that matches the size of the specimen under the projected phase space density. A source-limited performance can be delivered by proper and flexible electron optical designs to rotate the phase space so as to optimize the performance limited only to the Liouvilles theorem constraint, which is ultimately subject to the brightness of the electron sources preserved during the production of the electron beam. Utilizing a recently implemented high-brightness electron source under a DC-gun linear acceleration field, we test the performance of such a beamline for ultrafast electron microdiffraction and coherence imaging. We demonstrate the feasibilities of single-shot mirodiffraction on a single micrometer-sized domain in Highly Ordered Pyrolytic Graphite (HOPG) and coherent diffractive imaging of 10 nm scale charge-ordered domain structures in single-crystal complex materials, as qualified by the measured brightness at the sample plane. These initial results show that source-limited performance even from a sub-relativistic electron beamline can drastically improve the current performance of ultrafast electron imaging and diffraction.

ULTRAFAST TRANSMISSION ELECTRON MICROSCOPY BASED ON A LASER-DRIVEN SCHOTTKY FIELD EMITTER

Armin Feist, Katharina E. Echternkamp, Reiner Bormann, Nara Rubiano da Silva, Marcel Möller, Jakob Schauss, Sergey V. Yalunin, **Sascha Schäfer**, and Claus Ropers

4th Physical Institute, University of Göttingen

Ultrafast transmission electron microscopy (UTEM) is a promising technique, which utilizes sub-picosecond electron pulses within an electron microscope to resolve ultrafast laser-induced processes on a nanometer length scale [1]. The potential of this approach crucially depends on the implementation of advanced laser-driven electron sources which make full use of the high-quality electron optics within a TEM. Here, we present recent developments and first applications of the Göttingen UTEM instrument, which is based on the custom modification of a JEOL 2100F Schottky field emission TEM. The laser-triggered nanoscopic electron source employs localized single-photon photoemission from a tip-shaped ZrO/W(100) emitter. Highly coherent ultrashort electron pulses with a normalized emittance of 5 nm·mrad are generated, enabling ultrafast electron imaging with phase contrast and time-resolved local probing. Specifically, at the sample position, we obtain electron focal spot diameters down to 3 nm with a temporal pulse width of 300 fs (full-width-at-half-maximum) and a spectral bandwidth of 0.6 eV. In a first application, we utilize the nanoscopic probing capabilities of our microscope to investigate the inelastic scattering of free electrons with confined light [2,3]. Specifically, the energy spectrum of electrons traversing an optical near-field develops into a comb of spectral side-bands, corresponding to the absorption and emission of multiple photons with the interaction strength governed by the local near-field amplitude. We experimentally demonstrate the quantum coherence of this process by observing Rabi oscillations in the population of spectral side-bands as a function of the amplitude of the optical driving field [4]. Such an interaction constitutes a coherent optical phase-modulation of the incident electron wave function, and we discuss its application for the longitudinal manipulation of electron densities on attosecond time scales.

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DEVELOPING ULTRAFAST LOW-ENERGY ELECTRON DIFFRACTION

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Time-resolved high-energy electron diffraction experiments have unraveled ultrafast structural and electronic processes in bulk media [1]. Ultrafast diffraction with low-energy electrons would be ideal to investigate the structural dynamics of quasi-two-dimensional systems such as ultrathin films and surfaces. However, the realization of such experiments remains demanding, due to the challenges in creating suitable electron pulses [2]. Here, we discuss the development of ultrafast low-energy electron diffraction (ULEED) with first applications in transmission and reflection geometries [3]. Low-energy electron pulses of few-picoseconds duration are generated with a laser-driven tungsten needle emitter and are employed in a laser-pump/electron probe scheme. In a first application, we study the structural dynamics of an ordered polymer adsorbate on free-standing graphene by ULEED in transmission [3]. We give a detailed account of the characteristic time scales of the superstructure dynamics, including the energy transfer from the graphene to the adsorbate layer, the loss crystalline order in the polymer, and the formation of amorphous components at extended spatial frequencies. Furthermore, we extend ULEED to a backscattering geometry by developing a miniaturized laser-driven photoelectron source. At low electron energies around 100 eV, we can resolve the nearly commensurate charge density wave (CDW) [4] at a 1T-TaS_2 surface, which demonstrates the capability of ULEED to access complex electronic and structural dynamics at surfaces.

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ORIENTED SINGLE-CRYSTAL PHOTOCATHODES: A ROUTE TO HIGH-QUALITY ELECTRON PULSES

W. Andreas Schroeder, T. Li, and B.L. Rickman

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The generation of ultrashort electron pulses with a high spatial quality (i.e., high transverse brightness) from laser-driven photocathode electron guns is primarily dependent upon two factors: (i) areal pulse charge density limitations due to screening of the acceleration field (the short-pulse Child's law), and (ii) the rms transverse momentum of the emitted electrons. While the former is mainly influenced by the photo-gun's operational parameters, a density functional theory (DFT) based analysis of photoemission has revealed the fundamental role that the band structure of a planar photocathode material has on momentum distribution of its emitted electrons. This DFT-based analysis is in good agreement with our solenoid-scan measurements of the rms transverse momentum of electrons emitted from ten elemental metal photocathodes (Ag, Be, Cr, Cu, Mo, Nb, Sn, Ta, V, and W) - measurements which deviate significantly from prior theoretical predictions for many of the metals. Importantly, our analysis indicates that photoemission from hole-like (negative dispersion) electronic states with a low transverse effective mass is preferred for the generation of electron pulses with intrinsically low divergence. Moreover, the rms transverse momentum of electrons emitted from such hole-like states is not strongly dependent upon temperature. As a result, Group Vb (Nb, Ta, and V) metal photocathodes are preferred over Ag, Be, Cu and Sn, and those from Group VIb (Cr, Mo, and W). Further and in agreement with prior experimental work, the DFT-based analysis also shows that many elemental metals have strong work function anisotropy with respect to crystal orientation, which should lead to the photoemission of spatially inhomogeneous electron pulses from planar polycrystalline metal photocathodes as each micro-crystalline face will generally have different emission characteristics (i.e., efficiency and rms transverse momentum). Consequently, significant improvements in the homogeneity and transverse brightness of ultrashort electron pulses for diffraction and imaging applications appear to be possible through the use of single crystal photocathodes with a selected orientation

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ULTRABRIGHT FEMTOSECOND ELECTRON SOURCES FOR THE STUDY OF ULTRAFAST STRUCTURAL DYNAMICS

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Current ultrafast structural techniques provide the temporal and spatial resolutions required for the stroboscopic observation of atoms in motion. In regards to femtosecond electron sources, different compression approaches have made it possible the generation of ultrashort and ultrabright electron pulses. With an effective brightness only one hundredfold below that of fs-hard X-ray Free Electron Lasers, ultrabright femtosecond electron sources have reveal unprecedented results in the study of photoinduced ultrafast structural dynamics [1, 2]. I will present a brief overview of field along with a recent femtosecond electron diffraction (FED) study of the photoinduced insulator-to-metal phase transition of organic charge-transfer salt (EDO-TTF)2PF6 [3]. Here, we implemented a low repetition rate (10 Hz) and ultra-bright femtosecond electron source in order to avoid cumulative heating and photo degradation effects and obtain a movie of the relevant molecular motions driving this photo-induced insulator-to-metal phase transition. We were able to record time-delayed diffraction patterns that allow us to identify time-dependent changes over hundreds of Bragg peaks. Model refinement calculations indicate the formation of a transient intermediate structure (TIS) in the early stage of charge delocalization (during the initial 2 ps). The molecular motions driving the formation of TIS were found to be distinct from those that, assisted by thermal relaxation, convert the system into a metallic-like state on the 100-ps timescale. These findings illustrate the potential of ultrabright femtosecond electron sources for capturing the primary processes governing structural dynamics with atomic resolution in labile systems relevant to chemistry and biology.

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DEVELOPMENT OF IN-SITU ENVIRONMENT CELL FOR DYNAMICAL IMAGING AND CHEMICAL ANALYSIS OF GAS-SOLID INTERACTIONS

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In recent years the environmental transmission scanning electron microscope (ESTEM) has been successfully employed to elucidate the structural and chemical changes occurring in the catalyst nanoparticles under reactive environments. While atomic-resolution images and the combination of high spatial and energy resolution is ideally suited to distinguish between active and inactive catalyst particles and identify active surfaces for gas adsorption, unambiguous data can only obtained from the area under observation. This lack of global information available from TEM measurements is generally compensated for by using other, ensemble measurement techniques such as x-ray or neutron diffraction, x-ray photoelectron spectroscopy, infrared spectroscopy, Raman spectroscopy etc. However, it is almost impossible to create identical experimental conditions in two separate instruments to make measurements that can be directly compared. Moreover, ambiguities in ESTEM studies may arise from the unknown effects of the incident electron beam and uncertainty of the sample temperature. We have designed and built a unique platform that allows us to concurrently measure atomic-scale and micro-scale changes occurring in samples subjected to identical reactive environmental conditions by incorporating a Raman Spectrometer on the ESTEM. We have used this correlative microscopy platform i) to measure the temperature from 60 μm^2 area using Raman shifts, ii) to investigate light/matter interactions iii) as a heating source, iii) for concurrent optical and electron spectroscopy such as cathodoluminescence, EELS and Raman. Details of the design, function, and capabilities will be illustrated with results obtained from in situ combinatorial measurements.

MAKING MOVIES OF BIOLOGICAL MACROMOLECULES USING TIME-RESOLVED SERIAL FEMTOSECOND CRYSTALLOGRAPHY

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The method of time-resolved serial femtosecond crystallography (TR-SFX) provides a technique to record reaction intermediates at atomic spatial resolution and femtosecond time resolution, enabling the construction of an ultrafast movie of biological macromolecules in action. We present results from the model system photoactive yellow protein (a bacterial blue light photoreceptor), demonstrating that TR-SFX can produce difference electron density maps of exquisite quality to 1.6 angstroms resolution with a level of reaction initiation not possible using conventional 3rd generation synchrotron X-ray sources (Tenboer et al., 2014). This high level of reaction initiation stems from the ability of an X-ray free electron laser to obtain diffraction images from single microcrystals. These results pave the way toward probing exciting femtosecond intermediates, such as the cis to trans isomerization that is characteristic of many photo-reactive proteins.

DEVELOPMENT OF ULTRAFAST DIFFRACTION AND IMAGING AT SLAC

X.J. Wang

SLAC National Accelerator Laboratory

SLAC launched the Ultrafast Electron Diffraction and Imaging (UED&UEM) initiative with the objective of developing the world leading ultrafast electron scattering instrumentation, complementary to the X-ray Free Electron Laser - Linac Coherent Light Source (LCLS). The objective of the SLAC initiative is to develop a UED&UEM facility will possess unique capabilities that enable Grand Challenge science in chemistry, material science, physics and biology. In addition, the ability to couple the UED&UEM measurements with linac-based intense THz and X-ray FEL pump pulses will open new scientific opportunities. The SLAC UED&UEM facility will take advantage of the recent developments in high-brightness ultrafast electron sources, high-field magnets and electron detection. It will provide direct access to atomic coordinates with temporal resolution down to 100 fs and even below in the diffraction (UED) mode. The ultrafast imaging capabilities of the SLAC UEM will represent a paradigm shift compared to present day facilities, and it can achieve 10-nanometer and 10-picosecond resolution in single-shot mode. To realize high temporal resolution required for the SLAC UED&UEM facility, a MeV high-brightness electron beam generated by a photocathode RF gun will be employed. This allows more electrons to be packed into each bunch, offering single-shot capabilities similar to those of x-rays from LCLS. A further important advantage of relativistic beams is that they eliminate the velocity mismatch between the electromagnetic pump pulses and the electron probe beam. This mismatch limits the time resolution of ultrafast dynamics for dilute samples, such as gas and liquid samples. In addition to the higher temporal resolution, MeV electrons can penetrate thicker samples. Finally, the higher electron beam energy leads to a larger elastic scattering cross section and a decrease in the inelastic scattering cross sections, increasing the diffraction signal and reducing inelastic scattering.

LATTICE DYNAMICS IN AU THIN FILM AND NANOPARTICLES, AND PBSE QUANTUM DOT STUDIED BY ULTRAFAST ELECTRON DIFFRACTION

Xuan Wang[1]], Jun Zhou[2], Matthew Gorfien[2], Dong Li[2], Jim Cao[2]

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Ultrafast electron diffraction (UED) can simultaneously monitor both coherent and random motion of lattice, and therefore provides a power tool to study lattice dynamics with sub-picosecond temporal resolution. In this talk, I will present several such examples in nanometer scale devices. In Au thin film and nanoparticles, we found that the thermal expansion was always ahead of the thermalization process of the lattice, indicating the contribution from the electron thermal stress. By carefully fitting the vibration in Au thin film in picosecond time scale, the electronic Grüneisen constant of Au was deduced. In PbSe quantum dot (QD), the thermalization process of lattice followed by heat diffusion to the substrate was observed. The thermalization process of lattice could be well fitted by a mono-exponential curve with a time-constant about 2ps showing no sign of phonon bottleneck effect. The heat diffusion process was also very fast with a time-constant about 30ps, which was due to the big surface to volume ratio and good contact between QD and the substrate.

ELECTRONIC AND NUCLEAR DYNAMICS IN STRONG FIELD IONIZATION

Peter Sandor, Arthur Zhao, Vincent Tagliamonti, Tamas Rozgonyi and **Thomas Weinacht**

Stony Brook University

Strong field ionization is an important tool for studying molecular structure and dynamics. This talk will focus on experiments that study strong field ionization with few cycle laser pulses, using velocity map imaging of electrons and ions in coincidence to understand the coupled electron and nuclear dynamics during the ionization process.

MEV UED/UEM DEVELOPMENT AT SHANGHAI JIAO TONG UNIVERSITY

Dao Xiang

Shanghai Jiao Tong University

Historically particle accelerators are instrumental for high energy physics (accelerator based colliders) and photon science (accelerator based synchrotron light sources and free electron lasers). Now there is growing interest in applying accelerator technology to solve the grand challenge in probing matter at ultrafast temporal and ultrasmall spatial scales. In this talk I will present the status and future plans for the MeV ultrafast electron diffraction and microscopy facility being constructed at Shanghai Jiao Tong University.

TRACELESS TOTALLY SYMMETRIC TENSOR BASED FAST MULTIPOLE METHOD FOR SPACE CHARGE FIELD CALCULATION

He Zhang[1], He Huang[2], Rui Li[1], Jie Chen[1], Li-Shi Luo[2]

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The fast multipole method (FMM) is widely used to calculate the Coulomb interaction between a huge amount of charged particles. The efficiency of FMM scales with $O(N)$ for N particles with any arbitrary distribution. Hence it is apposite for problems with complicated charge distribution or geometry. We developed a new FMM code for Coulomb interaction. In the new code the Coulomb potential is expressed as traceless totally symmetric tensors in Cartesian coordinates, in which the number of independent element is only $2n + 1$ for the n^{th} rank, while it is 3^n for a general Cartesian tensor. The significant reduce of independent element number in Cartesian tensor helps to achieve high efficiency in the new algorithm. Result of test runs for space charge field calculation and multi-particle tracking using the new algorithm will be presented.

NONEQUILIBRIUM QUANTUM DYNAMICS IN TMD MATERIALS

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Characterizing and understanding the emergence of multiple macroscopically ordered electronic phases over subtle tuning of temperature, pressure and chemical doping has been a long-standing central issue for complex materials research. Unlike classical systems, phase transition of many- body quantum systems can be driven nonthermally by tuning the electron-phonon and electron- electron interactions without involving excessive lattice entropy, which may be enhanced by low-dimensionality. Identifying the origins for the competitive or cooperative emergence of various interaction-driven functional states and their tuning by doping, temperature, strain, or electrostatic and magnetic fields is of vital importance for elucidating the basic physics and in order to leverage their enormous technological potential. We study transition-metal dichalcogenide (TMD) materials, which host an assortment of intriguing electronic phases, including various textured charge-density orders, as well as recently reported superconducting phase under chemical doping, and pressure. Using femtosecond infrared pulses a succession of electronic phase transitions are successfully triggered in 1T-TaS_2, and a temperature-optical-density phase diagram is constructed, and substantiated with the dynamics of metastable states highlighting the cooperation and competition through which the macroscopic quantum orders emerge. The optical-doping-induced changes highlight the previously undisclosed interaction-driven dynamics and hidden states, in contrast to thermally driven phase transformations. The methodology introduced here can be generally applied to survey the complex energy landscape in strongly correlated electron systems, avoiding the difficulty of electrostatic gating or confounding effects due to defects and/or disorder. In particular, the observation of robust non-thermal switching at meso-scales and at ultrafast timescales, provides a platform for designing high-speed low-energy consumption nano-photonics and electronics devices.

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