FEIS 2013

Femtosecond Electron Imaging and Spectroscopy

December 9 -12, 2013
Key West, Florida
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FEIS 2013 Schedule

All sessions take place in the Grand Ballroom at Casa Marina

Monday	**9-Dec**		
8:00-8:45			**Registration**
8:45-9:00			**Welcome**
9:00 -	Hrvoje Petek	Pittsburgh	Session Chair
9:00-9:30	Yimei Zhu	BNL	Probing materials' behavior using ultrafast electrons
9:30-10:00	Geoffrey Campbell	LLNL	Quantitative measures of phase transformation kinetics with the dynamic transmission electron microscope
10:00-10:30			**Coffee Break**
10:30-11:00	Nigel Browning	PNNL	Development and application of in-situ gas and liquid stages for quantified TEM, STEM and DTEM observations
11:00-11:30	Sang Tae Park	Caltech	Plasmon charge density probed by ultrafast electron microscopy
11:30-12:00	Xijie Wang	BNL	MeV electron beam for ultrafast electron diffraction and imaging
12:00-2:00			**Lunch Break**
2:00-3:00			**Discussions (UED/UEM & Synergistic development)**
3:00-	James Freericks	Georgetown	Session Chair
3:00-3:30	Hrvoje Petek	Pittsburgh	Ultrafast photoemission electron microscopy: Imaging nonlinear plasmonic phenomena on the femto/nano scale
3:30-4:00	Shuji Sakabe	Kyoto	Intense femtosecond laser accelerated electron pulses for single-shot ultrafast electron diffraction and electron deflectometry
4:00-4:30	Jianming Cao	FSU	Ultrafast structure dynamics in metal films
4:30-5:00	Fabrizio Carbone	EPFL	Time-domain observation of coherent phenomena in solids and nano structures
5:00-5:30	Masaki Hada	Hamburg	Cold ablation driven by localized forces: A femtosecond electron diffraction study
5:30-6:00	Nicholas Matlis	LBNL	Compact laser-plasma accelerators for ultrafast electrons, X-Rays and THz

Tuesday	**10-Dec**		
8:00 -	Yimei Zhu	BNL	Session Chair
8:00-8:30	Thomas Devereaux	Stanford/SLAC	Theoretical understanding of ultrafast electron dynamics in model systems
8:30-9:00	Nuh Gedik	MIT	Observation of Floquet-Bloch states in topological insulators
9:00-9:30	Margaret Murnane	Colorado/JILA	Probing electron dynamics in molecules, quantum dots and materials at the space-time limits using coherent tabletop high harmonic X-rays
9:30:10:00	Shriram Ramanathan	Harvard	Ultrafast phase transitions in correlated oxides and electronics
10:00-10:30			**Coffee Break**
10:30-11:00	James Freericks	Georgetown	Exact theoretical description of pump/probe experiments in charge density wave insulators
11:00:11:20	Simon Schweda	Göttingen	Polymer superstructure dynamics on graphene probed by ultrafast low-energy electron diffraction
11:20-11:40	Luca Piazza	EPFL	Ultrafast structural and electronic dynamics of the metallic phase in a layered manganite
11:40-12:00	Zhensheng Tao	MSU	Ultrafast metal-insulator and charge-ordering transitions in correlated transition metal compounds
12:00-2:00			**Lunch Break**
2:00-3:00			**Discussions (Emerging opportunities & High-flux techniques)**

3:00 -	Margaret Murnane	Colorado/JILA	Session Chair
3:00-3:30	Pietro Musumeci	UCLA	High-brightness beam science
3:30-4:00	Jinfeng Yang	Osaka	RF gun based MeV transmission electron microscope
4:00-4:30	Josef Frisch	SLAC	Laser / RF synchronization
4:30-5:00	Valery Dolgashev	SLAC	X-band deflectors used for fs diagnostics of electron beams
5:00-5:30	Daniele Filippetto	LBNL	The APEX photo-gun: an high brightness MHz repetition rate source
5:30-6:00	Roland Janzen	Mainz	Monochromatizing without filtering using dynamic fields without bunching - a new concept for D-TEM illumination
7:00			**Conference Dinner at Conch Republic Seafood Company**
Wednesday	**11-Dec**		
8:00 -	Valery Dolgashev	SLAC	Session Chair
8:00-8:30	Harald Rose	Ulm	Effect of Coulomb interactions on resolution in ultrafast electron microscopy
8:30-9:00	Martin Berz	MSU	High-order spatio-temporal aberrations under presence of space charge
9:00-9:30	Erik Kieft	FEI	Optimization of pulsed photoemission TEM
9:30:10:00	Weishi Wan	LBNL	Electron bunch compression using static fields
10:00-10:30			**Coffee Break**
10:30-11:00	Brian Reed	LLNL	Electron beam physics and the limits of instrument performance
11:00:11:20	He Zhang	JLab	The multilevel fast multipole method in the differential algebra framework
11:20-11:40	Renkai Li	UCLA	Single-shot picosecond temporal resolution transmission electron microscopy
11:40-12:00	Christian Gerbig	Kassel	Resolution studies on a compact femtosecond transmission electron diffractometer and phonon decay in single crystalline graphite
12:00-2:00			**Lunch Break**
2:00-3:00			**Discussions (Beam optics & Challenging issues)**
3:00 -	Weishi Wan	LBNL	Session Chair
3:00-3:30	Ray Egerton	Alberta	Comparison of X-ray and electron beams for structural studies
3:30-4:00	Frank Merrill	LANL	Multi-GeV electron radiography for MaRIE
4:00-4:30	David Flannigan	Minnesota	Ancillary ultrafast electron microscopy techniques and considerations for studying nanoscale specimen volumes
4:30-5:00	Brett Barwick	Trinity	Imaging at the nm and fs scales with ultrafast electron microscopy (UEM)
5:00-5:30	Wen-Xin Tan	Chongqing	Design of ultrafast spin polarized low energy electron microscopy
5:30-5:50	Alexander Paarmann	FHI Berlin	Femtosecond low-energy diffraction and imaging
5:50-6:10	Cecile Limborg-Deprey	SLAC	Commissioning of SLAC X-Band Photoinjector
6:10-7:20			**Reception at Grand Ballroom Patio (Sponsored by FEI)**
Thursday	**12-Dec**		
8:00 -	Thomas Devereaux	Stanford/SLAC	Session Chair
8:00-8:30	Keiichiro Nasu	KEK	How much time necessary to photo-generate Fermi surface from true electron vacuum
8:30-9:00	Jom Luiten	Eindhoven	Cool beams for ultrafast electron imaging
9:00-9:30	Hyotcherl Ihee	KAIST	Molecular structural dynamics unveiled by time-resolved X-ray solution scattering
9:30-10:00	Ding-Shyue Yang	Houston	Environmental scanning electron microscopy: Probing ultrafast solvation dynamics at interfaces
10:00-11:00			**Roundtable Discussion**
11:00-11:10			**Closing**
11:10			**Transition to Boat (Lunch En Route)**
1:45			**Excursion (Glass Bottom Boat Tour)**

FEIS 2013 – Abstracts

IMAGING AT THE NM AND FS SCALES WITH ULTRAFAST ELECTRON MICROSCOPY (UEM)

Brett Barwick

Trinity College

Investigating ultrafast phenomena with femtosecond (10^{-15} s) and attosecond (10^{-18} s) temporal resolution is pivotal to understanding the dynamic processes that atomic, molecular and condensed matter systems undergo. The time scale for dynamics, at the atomic length scale, ranges from picoseconds to attoseconds for processes such as the heating of a thin metallic crystal and the motion of plasmons in metals. In this talk I will describe ultrafast imaging using single-electron packets as applied to several different nanoscale ultrafast processes. In particular, I will describe a new imaging method that exploits the fact that free electrons (when near a third body) can absorb and emit multiple photons. The physics describing the absorption and emission of photon quanta by free electrons is well known in AMO physics as a free-free transition and is manifested in the laser assisted photoemission effect. We form images by using only electrons that have absorbed photons; allowing us to observe the evanescent electric field created by plasmons that have been excited by an intense ultrafast optical pulse. In describing this imaging technique dubbed, photon-induced near-field electron microscopy (PINEM), I will also discuss future plans to extend the temporal resolution to tens of femtoseconds and possibly even the attosecond regime.

HIGH-ORDER SPATIO-TEMPORAL ABERRATIONS UNDER PRESENCE OF SPACE CHARGE

Martin Berz, Kyoko Makino

Michigan State University

We review modern differential algebraic (DA) methods for the computation of high order aberrations of electron microscopes, and extend these to systems with time dependent accelerating fields. Since the aberrations are driven by high-order derivatives of on-axis or midplane fields, care has to be taken when the fields arise from numerical computations or measurements. In this case they are best described by data on a closed surface via the Helmholtz theorem, which leads to infinitely often and fully Maxwellian fields thus preserving all Hamiltonian invariants of the motion, and that are furthermore smoothed if measurement or computational errors are present.

The methods are then combined with special versions of the fast multipole method for efficient computation of space charge effects. In the collisionless setting with smoothing by suitable wavelets, the results of the additional nonlinearities arising from space charge are thus combined with the ones due to external fields in a natural way. As a result, DA based high-order time stepping schemes originally developed for the computations of aberrations alone are seamlessly enhanced to incorporate space charge fields as well. In terms of the conventional FMM methods, this provides an additional enhancement going beyond their acclaimed O(N) behavior resulting from local multipole expansions of the fields, by even providing flow expansions in time and thus resulting in more efficient particle pushing.

DEVELOPMENT AND APPLICATION OF IN-SITU GAS AND LIQUID STAGES FOR QUANTIFIED TEM, STEM AND DTEM OBSERVATIONS

N. D. Browning[1], P. Abellan[1], I. Arslan[1], J. E. Evans[2], M. Gu[2], L. Mehdi[1], C. Park[3], L. R. Parent[4], W. D. Ristenpart[4], C. Wang[2], D. Welch[4], T. J. Woehl[4], H. Yang[4]

1. Fundamental and Computational Sciences Directorate, Pacific Northwest National Laboratory ; 2. Environmental Molecular Sciences Laboratory, Pacific Northwest National Laboratory ; 3. Department of Industrial and Manufacturing Engineering, FAMU-FSU College of Engineering ; 4. Department of Chemical Engineering and Materials Science, University of California-Davis

The last ten years have seen a paradigm change in (scanning) transmission electron microscopy ((S)TEM) with unprecedented improvements in spatial, spectroscopic and temporal resolution being realized by aberration correctors, monochromators and pulsed photoemission sources. Spatial resolution now extends to the sub-angstrom level, spectroscopic resolution into the sub-100meV regime and temporal resolution for single shot imaging (with the dynamic TEM or DTEM) is now on the nanosecond timescale (stroboscopic imaging extends this even further to femtoseconds). The challenge now in performing experiments in an (S/D)TEM is to implement the in-situ capabilities that will allow both engineering and biological systems to be studied under realistic environmental conditions. Performing experiments using in-situ stages or full environmental microscopes presents numerous challenges to the traditional means of analyzing samples in an electron microscope we are now dealing with the variability of dynamic process rather than a more straightforward static structure. In this presentation, I will discuss the recent developments in the design and implementation of in-situ stages being pursued at the Pacific Northwest National Laboratory (PNNL). Examples of the use of these capabilities for the direct imaging of oxidation and reduction in metals, ceramics and catalytic systems and to identify the fundamental processes involved in nucleation and growth of nanostructures from solution will be presented. As the in-situ stages have been designed to be incorporated into both high spatial resolution aberration corrected (S)TEM as well as into high temporal resolution DTEM, the potential for future experiments to study dynamics, including those in live biological structures, will also be discussed. The research described in this paper is part of the Chemical Imaging Initiative at Pacific Northwest National Laboratory under Contract DE-AC05-76RL01830 operated for DOE by Battelle. This work is supported in part by the United States Department of Energy, Basic Energy Sciences Grant No. DE-FG02-03ER46057. A portion of the research was performed using EMSL, a national scientific user facility sponsored by the Department of Energy's Office of Biological and Environmental Research and located at Pacific Northwest National Laboratory.

QUANTITATIVE MEASURES OF PHASE TRANSFORMATION KINETICS WITH THE DYNAMIC TRANSMISSION ELECTRON MICROSCOPE

Geoffrey H. Campbell, Thomas LaGrange, Bryan W. Reed, Melissa K. Santala, and Joseph T. McKeown

Lawrence Livermore National Lab

Time resolved transmission electron microscopy in situ observations of phase transformations in materials gives unique quantitative insights into the operative physics and kinetics of transformation process. We have developed a single shot instrument (see companion presentation by Bryan Reed) that allows us to observe the details of individual transformation events with temporal resolution as short as 15 ns and spatial resolution better than 10 nm. We have applied the technique to studies of rapid solidification in aluminum alloys, in which the speed of the liquid/solid interface and the composition of the alloy have strong effects on microstructure formation in the alloy system. We have also studied phase change materials to measure nucleation rates in nucleation-dominated systems such as $Ge_2Sb_2Te_5$ and to measure the growth rate in growth dominated systems such as GeTe. These measurements have been made with high accuracy in the regimes these materials are actually used in their technological applications. Similarly, we have studied the complicated growth morphology of explosively crystallized Ge. Finally, we will show results from the intermixing of pure Al and pure Ni across interfaces and the rate of intermetallic phase formation. All of these studies are based on measurements that are possible by no other technique than the dynamic transmission electron microscope (DTEM). This work performed under the auspices of the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering, by Lawrence Livermore National Laboratory under contract DE-AC52-07NA27344.

ULTRAFAST STRUCTURE DYNAMICS IN METAL FILMS

Jim Cao

Florida State University

Ultrafast electron diffraction is a rapid advancing technique capable of revealing the atomic-detail structural dynamics in real time. Over the past few years, this technique has been used to revolve structure dynamics in a variety of systems, such as phase transitions in physics and materials science, and reactions in chemistry and biology. In this talk, I will focus on its application in probing ultrafast structure dynamics in metal films. The topics will cover the mechanism of coherent phonon generation under the non-equilibrium condition and ultrafast photo-induced demagnetization in ferromagnetic materials.

TIME-DOMAIN OBSERVATION OF COHERENT PHENOMENA IN SOLIDS AND NANO STRUCTURES

Fabrizio Carbone

École Polytechnique Fédérale de Lausanne

Recent advances in ultrafast technology allow both the study and the control of material's properties thanks to the ability to record high temporal resolution movies of their transformations, or the ability to generate new states of matter by selecting ad hoc the excitation that drives a system out of equilibrium. The holy grail of this type of experiments is to combine a high tuneability of the excitation with a wide observation window. In solids, information on the structural degrees of freedom can be obtained in a very direct way via diffraction, while the accompanying dynamics of the electronic structure can be followed by fs optics (at q=0), electron energy loss spectroscopy (as a function of momentum q) or photoemission (also a momentum resolved probe but capable of accessing the very low-energy states close to the Fermi level). In addition, modern time-resolved microscopy also delivers information about the real space morphology of the materials as well as the spatial distribution of charge and spin patterns and domains. In this seminar, we will review the way in which a combination of these tools is used in our laboratory (LUMES at the EPFL) to address specific questions about high-temperature superconductivity, order-disorder transitions and charge/orbital ordering phenomena in solids.

THEORETICAL UNDERSTANDING OF ULTRAFAST ELECTRON DYNAMICS IN MODEL SYSTEMS

Tom Devereaux

SLAC and Stanford University

In this talk I present results from three separate model systems for pump-probe spectroscopy with the goal of understanding a language for non-equilibrium, driven electronic systems. Specifically I will discuss results for pump-probe photoemission across a metal-insulator transition, a coupled electron-lattice system, and driven graphene in an effort to generate a non-equilibrium topological insulator.

X-BAND DEFLECTORS FOR FEMTOSECOND DIAGNOSTICS OF ELECTRON BEAMS

Valery Dolgashev

SLAC National Accelerator Laboratory

Modern tendency to study ultra-fast physical process has led to experimental methods and devices that utilize extremely short, sub-picosecond electron bunches. Examples of such devices are X-ray free electron lasers, rf guns for relativistic electron diffraction, and advanced-accelerator test facilities. Electron energy in such devices varies for a few MeV in rf-electron guns to 20 GeV in SLAC's Facility for Advanced Accelerator Experimental Tests (FACET). Optimization and control of the longitudinal distribution of bunch parameters require diagnostics that have temporal resolution in the order of 10 femtoseconds. This presentation is a review of practical implementations of such diagnostics based on X-band deflecting cavities and waveguides.

COMPARISON OF X-RAY AND ELECTRON BEAMS FOR STRUCTURAL STUDIES

R.F. Egerton

University of Alberta

Electron and x-ray beams are used to provide structural and chemical information about organic and inorganic specimens, but both are forms of ionizing radiation and cause radiation damage through inelastic collisions. In addition, electrons have appreciable momentum, leading to displacement damage, and are charged particles, giving rise to electrostatic charging of the specimen and repulsive forces that interfere with the focusing of beams with high current density. However, electrons are diffracted much more strongly than x-rays and this stronger signal is advantageous in term of the spatial resolution of the information obtainable. In the x-ray case, it has proven possible to use sub-100fs pulses to determine molecular structures from diffraction data before an organic specimen damages. We will examine some of the problems involved in trying to do a similar thing with electrons.

HIGH-FLUX ULTRA-FAST ELECTRON DIFFRACTION DEVELOPMENT

D. Filippetto, W. Wan

Lawrence Berkeley National Lab

The APEX gun is a high-brightness high-repetition rate electron source under commissioning at LBNL. It delivers MeV electron pulses at MHz repetition rate, with the transverse brightness needed for driving an FEL. We are investigating the possibility to build a side beamline for high flux UED experiments. First simulations show that sub-100 fs beams with 10^6 particles per bunch and nanometer emittance are achievable. Jitter characterization is underway, with the goal to minimize energy, pointing and time jitters, and the final goal of pump-to-probe time jitters below the 100 fs. Possible proof of principle experiments are being investigated, to be performed whithin the next year.

ANCILLARY ULTRAFAST ELECTRON MICROSCOPY TECHNIQUES AND CONSIDERATIONS FOR STUDYING NANOSCALE SPECIMEN VOLUMES

David J. Flannigan

University of Minnesota

In the first part of my talk, I will provide an overview of photon-induced near-field electron microscopy and outline practical approaches for in situ pulse characterization. For all methods used to study non-equilibrium processes, deconvolution is critical for isolating intrinsic dynamics that occur on timescales commensurate with the instrument response. In order to accomplish this, one must have a way to precisely measure the instrument response function, ideally at the specimen location. Here, I will describe how the response of the low-loss region of the electron energy spectrum during UEM experiments is caused by varying the temporal overlap of the femtosecond photon and electron pulses at the specimen. Relative temporal variation of the pulses causes a modulation of the partitioning of the electrons into quantized virtual states. Variation in the partitioning causes the zero loss peak intensity to fall and rise, with the curve being a convolution of the two interacting pulses, thus revealing the instrument response function in the low-fluence regime. In this way, intrinsic structural dynamics can be isolated from the overall instrument response. In the second part of my talk, I will discuss considerations for achieving high spatial resolutions in UEM experiments. Conceptually, UEM can be thought of as consisting of combined ultrafast spectroscopy and electron microscopy laboratories having all the relevant apparatus and support equipment. To date, much attention has been focused on optimization of photoelectron pulse properties. In contrast, little emphasis has been placed on outlining and quantifying limitations imposed by environmental and specimen stability. Here, I will discuss effects of ambient conditions well-known to both the ultrafast spectroscopy and electron microscopy communities and the impact on observables will be illustrated. I will describe some of the measures we are taking at Minnesota to identify and mitigate such artifacts. In short, the combination of sophisticated experimental techniques, desired high energy-space-time resolutions, and sensitivities to picometer/milli-electron volt changes requires a detailed, quantitative understanding of specific environmental and specimen factors on femtosecond electron-based techniques. The magnitudes of these factors are non-universal and will vary greatly across labs, thus necessitating individualized efforts in identifying and deconvoluting inevitable artifacts from intrinsic dynamics.

EXACT THEORETICAL DESCRIPTION OF PUMP/PROBE EXPERIMENTS IN CHARGE DENSITY WAVE INSULATORS

James Freericks

Georgetown University

In this talk, I will describe a range of different theoretical results for the simplest model of a charge-density-wave insulator, which can be solved exactly in nonequilibrium. I will look at the behavior of the "nonequilibrium melting" of a CDW as seen in time-resolved photomission spectroscopy experiments, I will examine the behavior of quantum excitation, and how it changes from frequency driven to amplitude driven, and finally, I will examine the behavior of high harmonic generation from the solid state. I will discuss how these results compare with those from different experiments, where available, and will also describe what kinds of experiments are interesting to examine in the future. As theoretical treatments of nonequilibrium phenomena develop, we will be able to help understand current experiments and propose new ones. Much of this work is available on the arxiv at preprints numbered 1309.3574, 1309.2723, 1308.6066, and 1308.6060. I want to acknowledge support from the NSF under grants numbered OCI-0904597 and DMR-1006605, from the Department of Energy, Basic Energy Sciences, under grants numbered DE-FG02-08ER46542 and DE-SC-0007091, and the Indo-US Science and Technology Forum under a center grant numbered JC-18-2009 for various different parts of the research and of the collaborations.

LASER RF SYNCHRONIZATOIN

Josef Frisch

SLAC

Studies of ultrafast phenomena require synchronization of electron beams and experiment lasers at the sub-picosecond level. Similar synchronization is required for photocathode drive lasers relative to the RF fields in electron guns. Synchronization is frequently performed by using RF techniques to measure the relative phase of the mode-locked laser oscillator and the RF reference that is used to drive the accelerator, and then feeding back on the phase of the laser oscillator. It is also possible to measure the relative timing with electro-optical techniques, or to treat the laser source as the master oscillator and lock the accelerator RF phase. We discuss the design and engineering issues involved in these locking systems.

OBSERVATION OF FLOQUET-BLOCH STATES IN TOPOLOGICAL INSULATORS

Nuh Gedik

MIT

The topological insulator (TI) is a new phase of matter that exhibits quantum-Hall-like properties, even in the absence of an external magnetic field. Understanding and characterizing unique properties of these materials can lead to many novel applications such as current induced magnetization or extremely robust quantum memory bits. In this talk, I will discuss recent experiments in which we used novel time and angle-resolved photoemission spectroscopy (ARPES) to directly probe and control properties of Dirac Fermions. The unique electronic properties of the surface electrons in a topological insulator are protected by time-reversal symmetry. Breaking such symmetry without the presence of any magnetic ordering may lead to an exotic surface quantum Hall state without Landau levels. Circularly polarized light naturally breaks time-reversal symmetry, but achieving coherent coupling with the surface states is challenging because optical dipole transitions generally dominate. Using time- and angle-resolved photoemission spectroscopy, we show that an intense ultrashort mid-infrared pulse with energy below the bulk band gap hybridizes with the surface Dirac fermions of a topological insulator to form Floquet-Bloch bands. The photon-dressed surface band structure is composed of a manifold of Dirac cones evenly spaced by the photon energy and exhibits polarization-dependent band gaps at the avoided crossings of the Dirac cones. Circularly polarized photons induce an additional gap at the Dirac point, which is a signature of broken time-reversal symmetry on the surface. These observations establish the Floquet-Bloch bands in solids experimentally and pave the way for optical manipulation of topological quantum states of matter.

RESOLUTION STUDIES ON A COMPACT FEMTOSECOND TRANSMISSION ELECTRON DIFFRACTOMETER AND PHONON DECAY IN SINGLE CRYSTALLINE GRAPHITE

C. Gerbig, S. Morgenstern, C. Sarpe, A. Senftleben, M. Wollenhaupt and T. Baumert

University of Kassel, Institute of Physics and Center for Interdisciplinary Nanostructure Science and Technology (CINSaT)

Time-resolved diffraction, using x-ray or electron probes, has become a promising technique to directly provide insights into dynamics at the molecular level with ultrafast precision [1,2]. We study dynamical processes in single crystalline graphite by means of ultrafast electron diffraction in order to expand the understanding of phonon generation and decay mechanisms being essential for future carbon based electronic devices [3,4]. Our highly compact DC electron diffractometer is fully characterized by experiments and N-body simulations. The temporal profile of electron pulses is determined by grating enhanced ponderomotive scattering [2] at multiple charge densities. Spatial resolution and diffraction efficiency analyses are performed for selected electron source sizes. We demonstrate electron pulse durations below 150 fs and a transversal coherence length above 20 nm. At balanced conditions a temporal resolution of 200 fs along with high-definition diffraction is achieved for dynamical studies on graphite single crystals in a maintainable measurement time [5]. We further present generation and decay processes of incoherent as well as coherent phonons in graphite as a function of film thickness down to few-layer graphene [6].

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COLD ABLATION DRIVEN BY LOCALIZED FORCES: A FEMTOSECOND ELECTRON DIFFRACTION STUDY

Masaki Hada and R. J. Dwayne Miller

Max Planck Institute for the Structure and Dynamics of Matter

We will present some results obtained with newly developed femtosecond electron diffraction (FED) setups at Max Planck Institute for the Structure and Dynamics of Matter, Hamburg. The first FED study involves the structural evolution of alkali halide crystals under fs-ultraviolet-laser excitation. Single-shot time-resolved FED, optical reflectivity and ion detection experiments were applied to study the evolution of the ablation process that follows photoexcitation in crystalline NaCl, CsI and KI. The results reveal fast optical and structural changes associated with the development of disordering and electronic stress that would lead to ejection of material (large clusters and/or fragments) and the formation of micron-deep craters. We found evidence for a cold ablation explosion that occurs well below the threshold for plasma formation and the melting point of alkali halides, reflecting the very nature of electron correlations lying right at the onset of the Pauli repulsion well. The second study focuses on the photoinduced structural dynamics associated to charge transfer processes in a large-unit-cell quasi-two dimensional strongly correlated materials. The development of FED setups has reached the point where structural studies of protein dynamics are possible.

MOLECULAR STRUCTURAL DYNAMICS UNVEILED BY TIME-RESOLVED X-RAY SOLUTION SCATTERING

H. Ihee

KAIST and Institute for Basic Science (IBS), Daejeon, South Korea

Time-resolved X-ray solution scattering (liquidography) is an excellent tool for directly probing the structural dynamics of chemical and biological reactions occurring in solution phase. A unique combination of structural sensitivity and superb time resolution of this technique enables characterization of transient structures of reacting molecules in solution. Over the last decade, we have applied this technique to various molecular systems in solution phase and have successfully elucidated the dynamics and structural details of those reactions [1-5]. TRXL (Time-Resolved X-ray Liquidography) can provide direct structural information generally difficult to extract from ultrafast optical spectroscopy such as the temporal progression of bond lengths and angles of all molecular species including short-lived intermediates over a wide range of times, from picoseconds to milliseconds. TRXL elegantly complements ultrafast optical spectroscopy because diffraction signals are sensitive to all chemical species simultaneously and the diffraction signal from each chemical species can be quantitatively calculated from its three-dimensional atomic coordinates and compared with experimental TRXL data. Representative application examples including spatiotemporal kinetics and structural dynamics of small molecules and proteins will be presented.

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MONOCHROMATIZING WITHOUT FILTERING USING DYNAMIC FIELDS WITHOUT BUNCHING - A NEW CONCEPT FOR D-TEM ILLUMINATION

Roland Janzen

Johannes Gutenberg Universität Mainz

Commonly used monochromators are energy filters. The majority of the particles with undesired energies is removed. Thus the output current diminishes with increasing monochromatization rate. Monochromators in the original meaning of the word that would affect the energy of the particles by an energy selective acceleration can only be realized by use of dynamic fields. Dynamic field applications without exception involve a condition between the phase of the dynamic field and the entrance point of time of the particle into the field. If this condition is fulfilled, the dynamic field application works exactly. Unfortunately the width of the time interval where the condition is fulfilled is mathematically zero. Thus up to now dynamic field applications in charged particle optics limit themselves to a small time interval around the periodically recurring point of optimum phase where the condition is fulfilled to a good approximation. For that purpose bunches are formed around the optimum phase point. Unfortunately bunch forming suffers from Liouville's theorem: The product of the bunch length and the energy spread within it is a constant. In this work a concept is presented that circumvents this misery. A circularly polarized standing wave deflects the charged particle beam to a rotating orbital feeding the particles into a propagating wave that rotates within a toroidal wave guide. The circular deflection supplies a time coding. The entrance point of time of any particle is locked to its azimuthal position. Provided that the deflecting field and the rotating propagating wave (denoted by working field in the following) are synchronized correctly the optimum phase condition is fulfilled exactly and constantly. Time uncertainty vanishes within the limits of technical and practical implementing of the concept. The working field may be cascaded. It can be used for a large variety of applications such as monochromatizing, accelerating, spread inversion, time focusing and so on. A monochromator based on circular deflection was demonstrated to work by computer simulation. The energy spread vanishes within the limits of technical and practical implementing of the concept without any loss of current. As a second key aspect in my talk I will present a vision of a dynamic TEM illumination column based on circular deflection yielding equidistant monochromatic one electron pulses. The latter concept makes use of plasmonic field emitters synchronized to the deflection and working fields.

OPTIMIZATION OF PULSED PHOTOEMISSION TEM

Erik Kieft

FEI

Defining, and optimizing, the performance of a pulsed Transmission Electron Microscope (TEM) operating on femtosecond to nanosecond timescales, is an entirely different game from normal, continuous operation. We will look into two aspects. First, pulsed operation, be it in 'stroboscopic' or 'single-shot' mode, necessitates the use of high peak current values. As a consequence, stochastic blur, which is the result of microscopic electron-electron interactions in the imaging section of the column, becomes a limiting factor to image resolution in all but the single-electron-pulse cases. As such, one needs to take it into account when considering the beam parameters (brightness, emittance, and number of pulses if applicable) for a given experiment. To this end, particle tracking simulations that include mutual particle interactions, have been performed on a simplified optical system. The results can be fitted to two different power laws for the blur-limited imaging resolution – one each for the so-called Pencil beam and Holtzmark regimes of stochastic blur. These fits can be combined with other scaling laws in order to guide optimization of the overall resolution (or info limit) as a function of a.o. pulse duration, field of view size, and image contrast type. Note that, contrary to conventional wisdom, it is found that highest brightness is not necessarily always best in pulsed imaging. Second, the electron source needs to be matched to the desired beam parameters as well as possible. It is advantageous to have an electron gun that can be easily modified and optimized for a given task. Again, particle tracking simulations can be employed to provide guidance on optimization of the electron gun. The method is applied to the photoemission gun of the newly introduced Tecnai Femto UEM, a versatile instrument based on the UEM-1 and UEM-2 prototypes at Caltech. Its design is based on a standard LaB6-gun with Wehnelt electrode. Parameter scans show how a few simple degrees of freedom can be exploited to tune performance of the gun and optimize it for varying types of experiments.

SINGLE-SHOT PICOSECOND TEMPORAL RESOLUTION TRANSMISSION ELECTRON MICROSCOPY

Renkai Li and Pietro Musumeci

Department of Physics and Astronomy, UCLA

An active research area to further enhance the capability of state-of-the-art transmission electron microscopies is to dramatically improve their temporal resolving power, which will enable us to visualize in real time many ultrafast dynamic processes in biology, chemistry, and material science. Progress in this direction critically depends on the generation, precise control and advanced diagnosis of electron beams with unprecedented brightness, including ultrasmall emittance, ultralow energy spread, and high beam current. Here we present the feasibility study of a single-shot ultrafast transmission electron microscopy using high brightness electron beams from a high gradient photocathode radio-frequency electron gun aiming at a few picoseconds temporal resolution and a few tens of nanometers spatial resolution. We will discuss several key technical innovations optimized to address the challenging demand on beam quality, including the generation of pico-Coulomb charge ultralow emittance beam in the cigar-aspect-ratio regime, the RF curvature regulation technique to greatly reduce the beam energy spread, and strong electron optics using permanent quadrupole magnets or nano-fabricated micro-quadrupoles. Effects of collective space charge forces and stochastic Coulomb scattering on the achievable spatial resolution will also be discussed.

COOL BEAMS FOR ULTRAFAST ELECTRON IMAGING

M.W. van Mourik, W.J. Engelen, E.J.D. Vredenbregt, and **O.J. Luiten**

Eindhoven University of Technology

We have developed a new class of electron source which is based on near-threshold, pulsed photoionization of laser-cooled, trapped rubidium atoms. This allows us to produce intense picosecond electron pulses with large intrinsic coherence length. The ultralow source temperature – three orders of magnitude lower than conventional sources – seems to be in contradiction with the spread in photon energy associated with femtosecond ionization laser pulses. We explain this surprising result by a detailed study of the electron trajectories during photoionization. In addition, we show how laser polarization effects arise and that these can be used to further control the electron beam properties. Recently we have produced the first diffraction patterns from graphite samples, which show that ultrafast nano-diffraction of solid state systems is possible with this new source. Our goal is to demonstrate single-shot electron diffraction of macromolecules and thus enable dynamical studies of biomolecules in an ambient environment.

CONVERTER CHARACTERIZATION FOR USE AT LANL PROTON RADIOGRAPHY (PRAD)

Fesseha G. Mariam

Los Alamos National Laboratory

Proton radiography at LANL has been used for over a decade to explore dynamic processes often driven by explosives. It has also been used for the radiography of quasi dynamic processes such as the observation of phase changes in metals during melting and solidification of alloys. The system offers the acquisition of up to 40 images at fast rates (as fast as 200 ns inter-frame time) or unlimited number of frames for the slow processes such as the melting and solidification of alloys (20 Hz). While fast convertors such as LSO and LYSO are employed for the radiography of the fast processes, slow converters such as CsI(Ta) offer better sensitivity and suppression of prompt background. We report some preliminary results on the comparative advantages of these two classes of converters.

COMPACT LASER-PLASMA ACCELERATORS FOR ULTRAFAST ELECTRONS, X-RAYS AND THZ

Nicholas Matlis, J van Tilborg, GR Plateau, AJ Gonsalves, SV Steinke, CGR Geddes, CB Schroeder, E Esarey, Cs Toth, B Shaw, S Shiraishi, WP Leemans

Lawrence Berkeley National Laboratory

Laser-plasma electron accelerators (LPAs) are emerging as promising tools for ultrafast science due to their ability to simultaneously produce in a compact format a plurality of ultrashort radiation sources including electrons (from MeV to GeV), X-rays (UV to keV) and THz radiation that are intrinsically synchronized to the drive laser system. This diversity of well-timed radiation sources offers new possibilities in ultrafast imaging and probing in a format that is accessible to a wide range of researches. State-of-the-art research in LPAs is aimed at improving control and stability of the accelerator performance to enhance their viability for applications. We discuss generation of electrons, X-rays and THz radiation from an LPA system and present recent results in using control of electron-injection in LPAs to improve the energy spread and stability of the accelerator. Electron pulses with percent-level energy spread in the 200 to 300 MeV energy range from a 2-millimeter accelerator using a sub-10 TW laser are demonstrated. We also introduce a new method for single-shot tomographic imaging which is ideally suited to ultrafast pump-probe experiments using multiple radiation modalities (in particular broad-band betatron X-rays) and demonstrate it's use in the measurement of density profiles of multiple-filament plasma targets.

MULTI-GEV ELECTRON RADIOGRAPHY FOR MARIE

F. E. Merrill, K. N. Borozdin, R. W. Garnett, F. G. Mariam, C. L. Morris, A. Saunders, P. L. Walstrom

Los Alamos National Laboratory

Charged particle radiography was first investigated in the late 1960's when high energy proton accelerators (> 100 MeV) provided the first charged particle probes capable of penetrating relatively thick objects. Because of the image degradation which results from scattering processes within the object, this technique was abandoned by the community. In the mid 1990's, however, Los Alamos National Laboratory developed a magnetic lens imaging technique that has enabled proton radiography for a wide range of applications. Today there are four proton radiography facilities operating in the world and two additional facilities are being designed or fabricated. More recently the technique of charged particle radiography has been extended to utilize high energy electron probes (demonstrated with 30 MeV electrons) and Los Alamos National Laboratory is developing this technique for applications at the future Materials and Radiation in the Extremes facility (MaRIE) with Multi-GeV electrons. The goal for this application is to measure fast dynamic materials properties with spatial resolution < 1 micron and temporal resolution < 1 ps. Through the collection of radiographic movies of dynamic processes a detailed understanding of dynamic materials can be gained. In this presentation we will describe this method of charged particle radiography, present some examples of proton and electron radiographic measurements and summarize our predictions for the performance of a 12 GeV electron radiography facility.

PROBING ELECTRON DYNAMICS IN MOLECULES, QUANTUM DOTS AND MATERIALS AT THE SPACE-TIME LIMITS USING COHERENT TABLETOP HIGH HARMONIC X-RAYS

Margaret Murnane

University of Colorado

Advances in extreme nonlinear optics now make it possible to efficiently upshift femtosecond lasers into the ultraviolet (EUV) and soft X-ray regions of the spectrum, to wavelengths as short as 8 Å. In an optimized geometry, the resultant high harmonics (HHG) emerge as fully spatially coherent beams, with ultra broad bandwidths supporting few-fs to attosecond pulses. This unique light source is ideally suited for capturing and controlling all dynamics relevant to function, from the attosecond timescales characteristic of electrons, to fs timescales characteristic of vibrations and dissociation, to ps timescales characteristic of rotations in molecules. Applications of tabletop ultrafast laser and x-ray sources in materials, molecular and nano-systems will be presented. In recent work, we performed the first photoelectron spectroscopy of quantum dots in the gas phase, and extracted how far the evanescent electron wavefunction extends from different size dots. This is a key property in understanding electronic coupling of nanoscale systems to their environment. We also probed the fastest phase transitions and spin transport in materials using ultrafast HHG x-rays. Finally, we implemented the first reflection-mode full field tabletop coherent diffraction x-ray microscope.

HIGH-BRIGHTNESS BEAM SCIENCE

P. Musumeci

University of California, Los Angeles

In this talk I will review recent results in the physics of high brightness electron beams with relevance to the generation of ultrafast electron scattering probes. Starting with a definition of brightness bridging the accelerator and microscopy community, I'll discuss the different regimes of pancake and cigar beam aspect ratio and show how they respond to the different requirements of ultrafast electron diffraction and time resolved microscopy. The roles of the cathode photoemission properties and injection electric field will be clarified. I will then talk in detail about one particular kind of high brightness electron source, the RF photoinjector, and how it enabled the application of MeV electron beams to ultrafast electron diffraction. To conclude I'll present an overview of our recent efforts to improve the instrument and expand its capabilities to include imaging mode.

HOW MUCH TIME NECESSARY TO PHOTO-GENERATE FERMI SURFACE FROM TRUE ELECTRON VACUUM.

Keiichiro Nasu

IMSS, KEK

Rapid relaxation dynamics of optically excited electrons in metallic systems has already been widely investigated. In most cases, however, only a few electrons are optically excited, while, the main part of electrons is still in the ground state, works as an infinite heat reservoir, resulting in quite rapid relaxation of newly given energy and momentum.

What occurs, if a macroscopic number of electrons are excited into a truly vacant conduction band without electronic heat reservoir, at absolute zero temperature? Two successive laser pulse excitations of GaAs, and subsequent time-resolved photoemission spectrum measurement on the conduction band electrons by Kanazaki-Tanimura can partly answer this most simple, but ultimate photo-induced phase transition problem.

Coulombic inter-electron scatterings within the conduction band, being completely elastic, can give no net energy relaxation. While, the phonon relaxation, according to the Luttinger theorem, becomes infinitely slow as the system approaches to the complete Fermi degeneracy, and hence, it never terminates.

FEMTOSECOND LOW-ENERGY DIFFRACTION AND IMAGING

A. Paarmann, M. Müller, S. Lüneburg, R. Ernstorfer

Fritz-Haber-Institut der Max-Planck-Gesellschaft
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The recent development of femtosecond electron and x-ray diffraction and imaging techniques allows for the direct observation of structural dynamics in the course of photo-induced chemical or physical processes with atomic spatial and femtosecond temporal resolution. Photo-induced structural dynamics are governed by the interplay of electronic and nuclear degrees of freedom, which depend on the dimensionality of the system. While so far these techniques have been predominantly applied to crystalline samples, we aim for the investigation of ultrafast dynamics in low dimensional systems, e.g. two-dimensional materials, surfaces, and nanostructures, which ask for a time-resolved technique with maximal scattering cross section. Expanding ultrafast electron diffraction to low electron energies in the sub-kV range will combine femtosecond temporal resolution with high surface sensitivity. We developed a novel setup for femtosecond low-energy electron diffraction (fsLEED) and imaging in the energy range of 50 to 1000 eV based on a laser-triggered metal nanotip. Owing to the confined emission area due to field enhancement, nanotips are nearly ideal point sources delivering highly coherent ultrashort electron pulses. Besides using single electron pulses at high repetition rates in order to eliminate space charge effects, femtosecond time resolution is achieved by using a compact geometry with sub-mm propagation distances, which minimizes dispersive temporal broadening of the electron wave packets [1]. The instrument is designed for two operation modes: (i) point-projection imaging and (ii) fsLEED, the latter either in transmission or reflection geometry. Time-resolved point projection microscopy (i) utilizes the high sensitivity of low-energy electron to weak electric fields, in order to map transient electric fields and charge distributions in photoexcited nanostructures. Specifically, we investigate charge carrier separation upon above-bandgap excitation in axially doped InP nanowires. This experiment provides a unique possibility to directly image ultrafast currents with nanometer resolution. The diffraction studies (ii), on the other hand, require a collimated electron beam. In order to maintain the short propagation distances in fsLEED experiments, we developed a compact microlens coated directly onto the shaft of the nanotip to collimate the intrinsically divergent electron beam without any optics between tip and sample [2]. We present first experimental data on transmission LEED of free-standing monolayer graphene using our compact approach.

[1] A. Paarmann et al., J. Appl. Phys. 112, 113109 (2012).
[2] S. Lüneburg et al., Appl. Phys. Lett., in print (2013).

PLASMON CHARGE DENSITY PROBED BY ULTRAFAST ELECTRON MICROSCOPY

Sang Tae Park and Ahmed H. Zewail

California Institute of Technology

Ultrafast electron microscopy in space and time domain utilizes an electron pulse to directly probe structural dynamics of nanomaterial, initiated by an optical pump pulse, in imaging, diffraction, spectroscopy, and their combinations. It has demonstrated its capability in the studies of phase transition, mechanical vibration, and chemical reaction. Moreover, electrons can directly interact with photons via near field component of light scattering by nanostructure, and either gain or lose light quanta discretely in energy. By energetically selecting those electrons which exchanged photon energies, one can image the photon-electron interaction, and is termed photon-induced near field electron microscopy (PINEM). Here, we reexamine the physical meaning of the electron-photon interaction, and show that the PINEM image directly maps the optically-driven charge density distribution of nanoparticle plasmons. This insight is applied to various nanostructures and the nature of their plasmon modes is discussed.

ULTRAFAST PHOTOEMISSION ELECTRON MICROSCOPY: IMAGING NONLINEAR PLASMONIC PHENOMENA ON THE FEMTO/NANO SCALE

Hrvoje Petek

University of Pittsburgh

Light interacting with a metal surface can excite both single-particle (e-h pair) and collective (plasmon) excitations. By angle-resolved photoemission spectroscopy and photoemission electron microscopy, we investigate the coherent ultrafast dynamical processes in interaction of light with silver metal surfaces. We employ the two photon photoemission process to image plasmonic phenomena in Ag metal films. By means of interferometric time-resolved photoemission electron microscopy (ITR-PEEM), we can create spatial maps of two-photon photoemission excited in nanostructured Ag films. We fabricate specific nanoscale structures for the coupling of surface plasmon polaritons (SPP), the electromagnetic modes of a metal/dielectric interface, and we image their effect on the coupling, propagation, interference, and focusing of SPP waves. By advancing the delay between identical and collinear pump and probe pulses with interferometric precision, we are able to record movies of SPP wave propagation, and nonlinear interactions with 50 nm spatial resolution and 330 attosecond/frame temporal precision. Based on simple theoretical models, we discuss the imaging process, the optics of SPP wave packets, and the prospects of ultrafast microscopy of plasmonic phenomena.

ULTRAFAST STRUCTURAL AND ELECTRONIC DYNAMICS OF THE METALLIC PHASE IN A LAYERED MANGANITE

L. Piazza, C. Ma, H. Yang, A. Mann, J. Li, F. Carbone

Laboratory for Ultrafast Microscopy and Electron Scattering, ICMP, Switzerland

The transition between different states in manganites can be driven by various external parameters. Controlling these transitions with light opens the possibility to investigate the microscopic path through which they evolve. We performed femtosecond (fs) transmission electron microscopy on a bi-layered manganite to study its response to ultrafast photoexcitation. We show that a photoinduced temperature jump launches a pressure wave that provokes coherent oscillations of the lattice parameters, detected via ultrafast electron diffraction. Their impact on the electronic structure are monitored via broad-band ultrafast electron energy loss spectroscopy (EELS), revealing the dynamics of the different orbitals in response to specific structural distortions.

ULTRA-FAST PHASE TRANSITIONS IN CORRELATED OXIDES AND ELECTRONICS

Shriram Ramanathan

Harvard University

I will discuss our on-going efforts in understanding metal-insulator transitions in correlated oxides in the rutile and perovksite structures (e.g. VO_2, $SmNiO_3$ respectively). The transition can be in ps-ns timescales depending on the triggering mechanism. Scientific questions pertain to the nature of the transition mechanism, i.e. electronic versus structural and whether they components can be de-coupled during actuation. There are a few ways to study this problem from a device physics perspective that I will elaborate on in the presentation, namely two and three-terminal devices that allow dynamic modulation of conductivity, gating the resistance without introducing joule-heating. Complexities arise in interpretation that is due to our limited understanding of the nature of electrically active defects at the correlated oxide interfaces. I will give representative examples of high performance solid state devices that we have realized with such ultra-fast phase transitions. Finally, I will point out some open questions in the field that could be worth visiting in a collaborative setting with this group of researchers.

ELECTRON BEAM PHYSICS AND THE LIMITS OF INSTRUMENT PERFORMANCE

B. W. Reed, T. B. LaGrange, W. J. DeHope, G. Huete, J. T. McKeown, M. K.

Lawrence Livermore National Laboratory

Short-pulse electron probe instruments, including dynamic transmission electron microscopes (DTEM) and various flavors of ultrafast electron diffraction and microscopy (UED/UEM) systems, cover a wide range of operating parameters but have one thing in common: They all aim to use electrons to extract information from a small volume of material in as short a time as possible. While some scientific problems can be studied in the limit of a single electron per pulse, in all other cases we must contend with electron beams with beam currents orders of magnitude higher than is normal for a conventional electron microscope. This introduces unique problems, including space charge effects, stochastic blur, and complex electron-sample interactions, but it also forces us to make the best possible use of the limited brightness of our electron sources. This talk will discuss the physical laws that govern electron beams as they relate to practical applications in DTEM, UED, and UEM, using six-dimensional phase space as a unifying concept. The technology is reaching the point where substantial performance improvements beyond the current state of the art can only be achieved by dramatically reducing the entropy of the electron beam at the source.

EFFECT OF COULOMB INTERACTIONS ON RESOLUTION IN ULTRA-FAST ELECTRON MICROSCOPY

H. Rose

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Ulm University, Germany

Pulsed electron beams are increasingly applied in dynamic electron microscopy for visualizing fast processes such as local changes of magnetization, phase transformations and chemical reactions. Photoemission electron microscopy with pulsed excitation sources like lasers or synchrotron radiation has been evolved as a standard method for observing dynamic processes on surfaces [1, 2]. Recently it has been suggested to employ high-intensity ultra-short electron beams for recording single-shot diffraction patterns or images of radiation-sensitive biological objects [3].

Periodic processes can be observed with low current densities by means of stroboscopic techniques, which are mainly employed for investigating oscillating currents, voltages and magnetic domains in microelectronic devices. However, these techniques cannot be used for visualizing non-periodic processes or for obtaining a single-shot diffraction pattern or image, respectively. In order to achieve high-spatial resolution, the pulse must contain as many electrons as possible. However, due to the repulsive Coulomb interaction between electrons, it is not possible to confine a very large number of electrons in a very small volume. Moreover, these interactions broaden the energy width of the image-forming beam and change the direction of flight of the electrons. As a result, the image spot will be broadened even if chromatic aberration has been corrected [4]. This loss of resolution will also arise for the diffraction pattern.

The mean space charge acts like a change of the focal length and of the aberrations of the electron lenses. Therefore, this stationary effect can be compensated in an aberration-corrected microscope. Unfortunately, this correction is not possible for the effects resulting from the stochastic interactions. Ray deflections arising within the lenses from stochastic interactions are most deleterious, because even small angular deviations may result in appreciable displacements at the image plane owing to the large distance between the lenses and the image plane. This deleterious effect also prevails for a lens-less system, where it reduces the resolution of the diffraction pattern. The broadening of the diffraction or image spots resulting from stochastic interactions is inversely proportional to $U^{*3/4}$. Here $U^* = U(1 + eU/2E_e)$ defines the relativistic modified acceleration voltage and $E_e \approx 0.51MeV$ is the rest energy of the electron. Hence, the resolution increases significantly by going to higher voltages if the geometry of the system remains fixed. However, this condition cannot be met for an imaging system because the focal length of the magnetic lenses increases in proportion to U^* whenever their magnetic field has reached its maximum. Therefore, going to high voltages will only improve the resolution limited by stochastic interactions if superconducting lenses with short focal lengths are employed. Projector systems composed of quadrupoles are another alternative because quadrupoles are strong-focusing elements and can form astigmatic intermediate images of the source, thus reducing the current density and the resulting stochastic ray deflections.

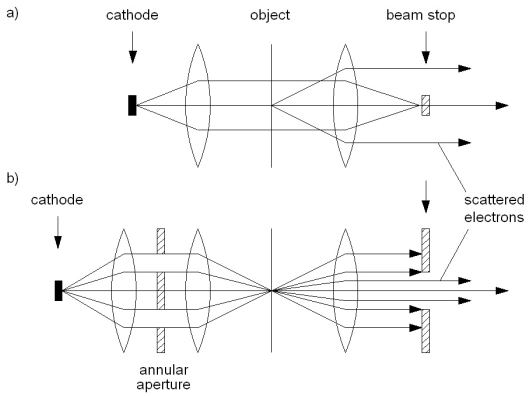

Figure 1: Schematic arrangement of apertures and lenses for (a) central beam stop and (b) hollow-cone dark-field imaging

Single elastic scattering prevails in amorphous objects whose thickness t is smaller than the elastic mean free path length. For simplicity we assume that the object is composed of atoms with the same atomic number Z. In this case the relation $l_{el} = 1/n_A\sigma_{el}$ holds. Here n_A is the atom density of the object and $\sigma_{el} \approx (E_e/eU^*)(1 + eU/E_e)^2 Z^{4/3}$ pm the elastic scattering cross-section of the atom. This cross-section is inversely proportional to U in the non-relativistic regime and adopts the constant value $2Z^{4/3}\text{pm}^2$ in the relativistic case $U >> 2E_e \approx 1$ MeV. For imaging thin ($t < l_{el}$) amorphous objects, it is advantageous to operate at lower voltages due to the larger fraction of scattered electrons. In addition, one can reduce the number of interactions in the regime between object and image by removing the non-scattered electrons in the back-focal plane of the objective lens by means of a beam stop. Two modes of dark-field imaging exist, as depicted in Fig.1. The first mode uses parallel illumination and a central beam stop, the other hollow cone illumination and a circular aperture. Compared to parallel illumination this mode reduces the effect of the Coulomb interactions due to the large angles of illumination which shorten the regions of high current density.

Instead of employing an annular aperture and a standard cathode for producing hollow cone illumination, it may be possible to develop a pulsed high-intensity annular cathode. We achieve hollow-cone Koehler illumination by imaging this cathode into the back-focal plane of the objective lens. In order to minimize the Coulomb interactions and to utilize as many scattered electrons as possible large usable angles are required for the illumination hollow cone and the objective aperture. Such angles can only be realized by correcting the resolution-limiting aberrations.

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[2] A. Krasyuk et al., Appl. Phys. A 76 (2003) 863.
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INTENSE FEMTOSECOND LASER ACCELERATED ELECTRON PULSES FOR SINGLE-SHOT ULTRAFAST ELECTRON DIFFRACTION AND ELECTRON DEFLECTOMETRY

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To observe ultrafast changes of atomic-scale structure in matters and electromagnetic fields near matters during phenomena, time-resolved electron diffraction and electron deflectometry using femtosecond electron pulses are useful, respectively. The key issue to realize single-shot ultrafast electron diffraction (UED) or electron deflectometry is to develop intense short electron-pulse sources. With conventional UED instruments, an electron pulse is generated at a photocathode irradiated by a femtosecond laser pulse and accelerated in an additional external static electric field. The amount of electrons in the pulse is limited because the electron pulse expands during its flight by space-charge forces in the pulse. Electrons accelerated by intense femtosecond laser pulses have potential for intense electron pulse sources. It is featured by pulse, point source, rather broad momentum, unnecessity of external accelerators, and perfect synchronization with other radiation (x-ray, ions, white light, THz, etc.) generated by the same laser pulse. We have been studying the physics of electron emission during/after laser-plasma interaction to develop higher-intensity electron pulses. Using the characteristics of broad momentum, we have demonstrated femtosecond pulse compression of a laser-accelerated electron beam with energy of around 350keV [1, 2]. The electron pulses are generated by irradiating a tightly focused terawatt femtosecond laser pulse on a polyethylene foil target, then, the pulse is compressed with an achromatic bending magnet system. It has been demonstrated to take a single-shot diffraction pattern using these femtosecond electron pulses. For an aluminum foil target, much more electrons are emitted than for the polyethylene foil target, but the electrons are emitted along the target surface while in the laser direction for the polyethylene foil target. Using this characteristics, it has been demonstrated to guide electrons along a metal wire target, resulting in directional emission with several tens higher intensity than metal foil target. To observe the electric fields near the laser plasma produced by the interaction of intense femtosecond laser pulse with a solid target, we have made the electron deflectometry using laser accelerated electron pulses with an electron lens [3,4,5]. Using femtosecond electron deflectometry with electron pulses accelerated by intense laser pulses mentioned above, it has been successfully demonstrated to observe the electromagnetic surface (Sommerfeld) wave propagating along a metal wire irradiated by an intense femtosecond laser pulse [6,7].

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[7] Phys. Rev. Lett. 110, 155001(2013).

POLYMER SUPERSTRUCTURE DYNAMICS ON GRAPHENE PROBED BY ULTRAFAST LOW-ENERGY ELECTRON DIFFRACTION

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Atomic-scale microscopy and diffraction techniques with high temporal resolution are of particular interest for the understanding of fundamental transient phenomena such as structural reorganizations and phase transitions. The past years have shown a substantial development in the time-resolved analysis of bulk media utilizing ultrashort x-ray or electron probes. However, the time-resolved investigation of structural dynamics at surfaces remains challenging, despite recent accomplishments, e.g., in ultrafast reflection high-energy electron diffraction. Implementing low-energy electron diffraction (LEED) with ultrafast temporal resolution in an optical pump / electron probe scheme is a natural approach to observing surface structures, due to the high surface sensitivity of slow electrons. So far, ultrafast LEED (ULEED) has not been demonstrated, owing to the technical difficulties in realizing suitable pulsed low-energy electron sources. In particular, in this regime, electron pulses are very sensitive to spatio-temporal broadening due to Coulomb-repulsion and velocity dispersion. In this contribution, we present the implementation of ultrafast LEED based on a nanometric photocathode, achieving a temporal resolution of few picoseconds. The setup allows us to investigate the structural dynamics of an ultrathin bilayer consisting of an ordered polymethylmethacrylate (PMMA) layer on freestanding, single-layer graphene. Specifically, we temporally resolve the ultrafast melting of a stripe-like PMMA superstructure and the formation of a disordered expanded phase. Our experiments illustrate the potential of time-resolved low-energy electron diffraction as a powerful tool to study ultrafast atomic-scale phenomena at surfaces.

DESIGN OF ULTRAFAST SPIN POLARIZED LOW ENERGY ELECTRON MICROSCOPY

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With the support of Chongqing University, we have procured a state-of-the-art aberration-corrected LEEM/PEEM with high temperature Scanning Tunneling Microscopy from SPECS. A spatial resolution of 1.7 nm has been achieved in LEEM mode during on-site commissioning in Germany. The main feature of this microscope is that it contains three magnetic prism arrays (MPAs), as oppose to two in their standard configuration, allowing the possibility of adding a second electron gun to the system to achieve spin polarized LEEM (SPLEEM). With an aberration corrector, the electron transmission flux of AC-SPLEEM is around one order of magnitude higher than non-AC case which is critical for developing time resolved SPLEEM. Therefore, we are currently developing an ultrafast spin-polarized electron gun, ultra-fast laser system and electronics with the support of the National Science Foundation of China (NSFC). Together with the necessary optics, the future electron gun will realize time resolved aberration-corrected spin-polarized LEEM (TR-AC-SPLEEM) mode at a time resolution of ps. We will report the concept of our electron optical design of the ultrafast spin-polarized electron gun and the whole column leading to the AC-TR-SPLEEM for low dimensional spin dynamics and ultrafast surface dynamics research.

ULTRAFAST METAL-INSULATOR AND CHARGE-ORDERING TRANSITIONS IN CORRELATED TRANSITION METAL COMPOUNDS

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Competitions between lattice, spin and charge-ordered states in correlated transition metal compounds can lead to phase transitions that are highly tunable by applying heat, temperature, electrical and optical excitations. These attributes are ideal for constructing electronic and photonic devices promising ultrafast high-fidelity switching with less energy than the conventional semiconductor-based devices. Nonetheless, the successful implementation of these devices is hindered by our current lack of understanding in key phase transition mechanisms under strong electron-electron correlation and electron-phonon coupling. Here, using optical reflectivity, TEM, and ultrafast electron crystallography, we investigate vanadium dioxide (VO_2) and tantalum disulfide (1T-TaS_2), which are two prototypical correlated electron systems exhibiting strongly first-order metal-insulator transitions due to subtle interplay between the underlying Mott and Peierls physics. In the VO_2 case, we find that for single-crystal nanobeams gently placed on substrates the structural and metal-insulator transitions can be tuned from cooperative to non-cooperative conditions by simply changing the substrates from insulating to metallic. In contrast, for polycrystalline thin film influenced by the inter-domain strain fields, the phase transitions are often cooperative at steady states, but exhibit step-wise atomic movements and strong anisotropic lattice responses under ultrafast perturbations. These clear-cut nonequilibrium features can be attributed to the competitive Mott and Peierls physics in directing the phase transitions. In the 1T-TaS_2 case, the relevant Mott and Peierls-driven processes are displayed in the distinct charge ordering and domain dynamics revealed at the ultrafast timescales. We will describe the various collective state switching phenomena from both the steady-state and ultrafast perspectives, and discuss the prospects of advancing such studies with the development of an RF-enabled ultrafast electron microscope system at MSU.

ELECTRON BUNCH COMPRESSION USING STATIC FIELDS

Weishi Wan

LBNL

It is well known that a short electron bunch (e.g. 100 fs) with reasonably high charge (e.g. 10000 electrons), would quickly expands to a few and, eventually, to hundreds of picoseconds due to space charge. In addition to limiting the number of electrons per bunch to the single digit, RF cavity have been used and successfully compressed high charge electron bunches (0.2 pC) to about 0.5 ps. Another possible way of bunch compression which may potentially simplify the technology and improve stability is using static electrostatic or magnetic fields. The idea is that an achromatic beam transport line with positive R56 can make the head of the bunch, which is more energetic due to space charge, to travel a longer distance thus compressing the bunch. This talk will review the ideas that have been proposed and discuss the effect of Coulomb scattering on the final bunch length.

MEV ELECTRON BEAM FOR ULTRAFAST ELECTRON DIFFRACTION AND IMAGING

X.J. Wang

Brookhaven National Laboratory

Recent years we have witnessed tremendous progress in our understanding of the ultrafast and ultra-small world thanks to the X-ray Free Electron Laser (XFEL). The development of photoelectron source directly led to the success of XFEL; and in the same time, such high-brightness electron source could also open the door to the next generation electron scattering instrumentation: MeV Ultrafast Electron Diffraction (UED) and Ultrafast Electron Imaging (UEM)[1]. MeV UED and UEM not only has the potential of a higher temporal resolution [2], but also has the larger the scattering signal and less sample-damage. After a brief review the history of MeV UED and UEM, I will discuss the latest developments and technical challenges in MeV UED and UEM. I will also discuss the potential scientific opportunities enabled by MeV UED and UEM.

[1] X.J. Wang et. al., FEMTO-SECONDS ELECTRON BEAM DIFFRACTION USING PHOTOCATHODE RF GUN, Proceedings of 2003 Particle Accelerator Conference, pp. 420 (2003).
[2] X.J. Wang et al, Experimental observation of high-brightness microbunching in a photocathode rf electron gun, Phys. Rev. E, 54,R3121 -3124 (1996).

ENVIRONMENTAL SCANNING ELECTRON MICROSCOPY: PROBING ULTRAFAST SOLVATION DYNAMICS AT INTERFACES

Ding-Shyue Yang

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Probing interfacial dynamics and heterogeneous catalysis at the nanoscale requires experimental methods that incorporate simultaneous resolutions in both space and time into environmental techniques. However, dynamics investigations of such surface-molecule structures are limited in time resolution, being controlled by the video camera rate of recording at (sub)milliseconds. In this talk, I will describe our development of environmental scanning ultrafast electron microscopy, whose orders-of-magnitude improvement in temporal resolution allows in situ detection of solid-gas interaction dynamics at materials surfaces. Using CdSe surfaces as a prototype, we investigated the spatiotemporal characteristics of solvation (by water, ammonia, and acetonitrile) in the recorded images. The ultrafast reorganization and relaxation dynamics of polar molecules in the adsorbate layers, observed on different crystalline surfaces, are striking in their temporal behavior and in the dependence on the surface structure, (0001) vs. (101bar0)/(112bar0). The present development paves the way to other applications, e.g., those involving charge flow on heterogeneous surfaces and particles, catalysis, and hydration of biological systems.

RF GUN BASED MEV TRANSMISSION ELECTRON MICROSCOPE

Jinfeng Yang

Osaka University

The first prototype of RF gun based relativistic-energy electron microscopy has been constructed at Osaka University to study ultrafast structural dynamic processes in matter. The RF gun driven by a femtosecond laser has generated a 100-fs-pulse MeV electron beam with emittance of 0.2 mm-mrad and energy spread of 10^{-4}. Both the electron diffraction and image measurements have been succeeded in the prototype using the femtosecond electron beam. In the diffraction measurement, an excellent quality of diffraction pattern was acquired with electron number of 10^6. The single-shot measurement is available in the prototype. In the image measurement, the TEM image was acquired with a total electron number of 10^8. The magnification was 3,000 times. In the next step, we will reduce further the emittance to increase the beam brightness on the sample, and then improve the spatial resolution to < 10 nm.

THE MULTIPLE LEVEL FAST MULTIPOLE METHOD IN THE DIFFERENTIAL ALGEBRA FRAMEWORK

He Zhang[1], Zhensheng Tao[2]
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1. Jefferson Laboratory ; 2. Michigan State University

Collective effects become more and more important as accelerator physicists try to provide higher and higher intensity beams. Numerical simulation is a very useful tool for collective effect study, and it is inevitable when the collective effect is strong and highly nonlinear. In this paper, we present a grid-free algorithm to calculate the electrostatic field between charged particles. The algorithm is based on the fast multipole method (FMM) and the differential algebra (DA). FMM allows us to calcualte the field of charged particles of any arbitrary distribution with an efficiency that scales linearly with the number of particles. DA considerably simplifies the math and allows us to calculate not only the field but also its high order derivativies. The algorithm can be parallelized to further improve its efficiency. It has been used in a simulation code for photoemission process. Some results about the changes of the electron bunch profile in the first 100ps of the photoemission process are presented.

PROBING MATERIALS BEHAVIOR USING ULTRAFAST ELECTRONS

Yimei Zhu

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In this presentation I will give a brief overview on the frontiers of electron microscopy, including atomic imaging, quantitative electron diffraction, energy-loss spectroscopy, off-axis electron holography and in-situ microscopy to understand materials functionalities. Examples of various probing methods will be given to reveal the behavior of electrons, spins, orbitals and lattice and their correlations. The advantages and drawbacks of the methods and their limitation on spatial and time resolution will be deliberated. Challenges and future opportunities for electron scattering will be discussed. The author would like to acknowledge the collaborations with Advanced Electron Microscopy and Nanostructure Group at BNL. The research was supported by the US Department of Energy under Contract no. DE-AC02-98CH10886.

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