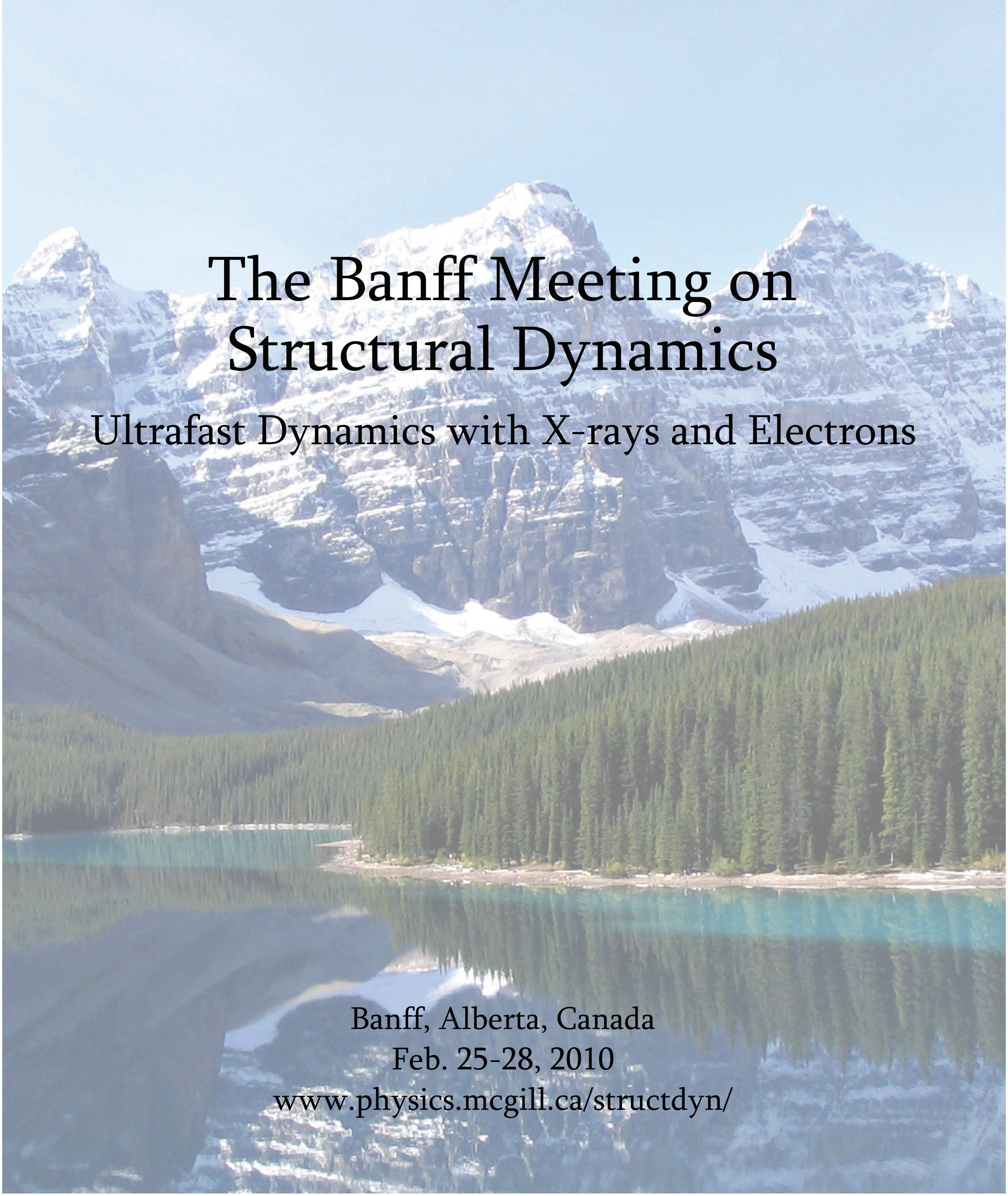

The Banff Meeting on Structural Dynamics

Ultrafast Dynamics with X-rays and Electrons

Banff, Alberta, Canada
Feb. 25-28, 2010
www.physics.mcgill.ca/structdyn/

The Banff Meeting on Structural Dynamics
Ultrafast Dynamics With X-rays and Electrons

February 25-28, 2010
The Banff Centre, Banff, AB, Canada

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Heinrich Schwoerer (U. Stellenbosch)
Michael Wulff (ESRF)

Invited Speakers
Philip Anfinrud (NIH)
John Arthur (LCLS/SLAC)
Andrea Cavalleri (CFEL/Oxford)
Lin Chen (Argonne)
Eric Collet (U. Rennes)
Pieter Glatzel (ESRF)
Michael Horn-von Hoegen (U. Duisberg-Essen)
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Jom Luiten (Eindhoven)
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Keith Moffat (U. Chicago)
German Sciaini (U. Toronto)
John Spence (ASU)
Michael Woerner (MBI)
Linda Young (Argonne)
Jim Zuo (UIUC)

The Banff Meeting on Structural Dynamics

TIME	Thursday, Feb. 25	Friday, Feb. 26	Saturday, Feb. 27	Sunday, Feb. 28
	Location: Max Bell Auditorium			
		Instruments and Techniques for Ultrafast Structural Dynamics	**Structural Dynamics in Materials: Strongly Correlated Materials**	**Structural Dynamics in Molecules**
8:30 AM		P. Glatzel, *Hard X-Ray Emission Spectroscopy*	S. Koshihara, *Dynamics of cooperative lattice-charge (spin) coupled phenomena induced by fs laser light irradiation studied by time-resolved X-ray diffraction*	P. Anfinrud, *Watching proteins function in real time via 150-ps time-resolved X-ray diffraction and solution scattering*
8:45 AM				
9:00 AM		C. Rose-Petruck, *Phase sensitive x-ray imaging and ultrafast chemical dynamics*	A. Cavalleri, *Optical Control in Complex Solids*	K. Moffatt, *But My Crystals Aren't Light-Sensitive....Help!*
9:15 AM				
9:30 AM		K. B. Møller, *Theory and Simulation of Time-Resolved X-ray Diffraction*	M. Eichberger, *Structural dynamics of the nearly commensurate phase in the Charge Density Wave compound 1T-TaS2 probed by ultrafast electron diffraction*	J. Kim, *Molecular Structural Dynamics Visualized by Pump-Probe X-ray Liquidography and Crystallography*
9:45 AM			D. Boschetto, *Coherent Phonon in Iron Pnictide Superconductor Ba(FeCo)As(x=0.06 and x=0.08)*	
10:00 AM		J. Bozek, *First Experiments with the AMO Instrument at LCLS*	*Discussion Break* **Structural Dynamics in Materials: X-rays**	*Discussion Break*
10:15 AM		M. Hoerner, *First results on nonlinear dynamics in diatomic molecules using the LCLS free electron laser*	S. Johnson, *Coherent and incoherent femtosecond structural dynamics in solids studied by x-ray diffraction*	L. Chen, *Molecular Snapshot in Solar Energy Conversion Processes Taken by Ultrafast X-rays*
10:30 AM	Registration (10:30am - 12:30am)	*Discussion Break*		
10:45 AM		N. Browning, *In-situ observation of irreversible reactions in liquids and gases by Dynamic Transmission Electron Microscopy (DTEM)*	A. Lindenberg, *Real-time structural dynamics in materials on femtosecond and picosecond time-scales*	C. Bressler, *Towards Femtosecond X-Ray Spectroscopies*
11:00 AM				
11:15 AM		J. Zuo, *Transient Electric Fields Induced By Ultrafast Pulsed Laser Irradiation and Implications for Time-Resolved Reflection Electron Diffraction*	M. Woerner, *Femtosecond x-ray powder diffraction*	N. Huse, *Photo-Induced Spin-State Conversion in Transition Metal Complexes Probed via Ultrafast Soft X-ray Spectroscopy*
11:30 AM				
11:45 AM	Registration (10:30am - 12:30am)	*Free Afternoon*	E. Collet, *Tracking Consecutive Steps of Photoinduced Switching Dynamics of Spin-Crossover Materials by X-ray Diffraction & Optical Pump-Probe Experiments.*	L. Young, *Observation of multiphoton processes in the x-ray regime: First experiments at LCLS*
12:00 PM				
12:15 PM			K. Sokolowski-Tinten, *Short-pulse laser induced transient structure formation and ablation studied with time-resolved coherent XUV-scattering*	D. Rolles, *Competition between Radiative and Auger Decay after X-FEL Ionization of Atoms and Molecules*
12:30 PM	*Working Lunch*		*Working Lunch*	P. M. Weber, *Structural Dynamics with Bound Electrons: Isomeric and Conformeric Motions of Hot Molecules*

	Ultrafast X-ray and Electron Sources I	*Free Afternoon*	**Structural Dynamics in Materials: Electrons**	*Meeting Closes*
1:30 PM	M. Wulff, *Probing molecular dynamics with short X-ray pulses from a synchrotron*		G. Sciaini, *Femtosecond Electron Diffraction: “Making the Molecular Movie”*	
1:45 PM				
2:00 PM	John Arthur, *The LCLS X-Ray FEL Facility*		T. LaGrange, *Studying Nanoscale Material Processes in the Dynamic Transmission Electron Microscope (DTEM)*	
2:15 PM				
2:30 PM	R. Falcone, *A new generation of soft x-ray free electron lasers*		M. Horn von Hoegen, *Ultrafast Electron Diffraction at Surfaces: From Non-Thermal Heat Transport to Strongly Driven Phase Transitions*	
2:45 PM	F. Kaertner, *Key Laser Technologies for Future X-ray Sources*			
3:00 PM	J. Rosenzweig, *Breaking the attosecond, Angstrom and TV/m field barriers with ultra-fast electron beams*		H. Jean-Ruel, *Femtosecond Molecular Photocrystallography*	
3:15 PM	*Discussion Break*		*Discussion Break*	
	Ultrafast X-ray and Electron Sources II		**Diffractive and Dynamic Imaging of Molecules**	
3:30 PM	Jiro Matsuo, *In-air femtosecond X-ray source*		H. Chapman, *First Results of Coherent Diffraction Experiments at the LCLS*	
3:45 PM				
4:00 PM	Heinrich Schwoerer, *Synchrotron Radiation from Laser Accelerated Electrons*		J. Spence, *Fast Protein Nanocrystallography*	
4:15 PM				
4:30 PM	Jom Luiten, *Single-shot Ultrafast Electron Diffraction*		A. Fratalocchi, *Linear and nonlinear imaging with XFEL: results from ab-initio computations*	
4:45 PM			D. Gauthier, *Single shot soft X-ray holography using extended references*	
5:00 PM	E. Vredenbregt, *Experimental realization of an ultracold electron source*		F. Krasniqi, *Ultra-fast, Ångström Scale Structure Determination of Molecules via Photoelectron Holography*	
5:15 PM	P. Musumeci, *RF photoinjector based ultrafast relativistic electron diffraction*		H. Thomas, *Explosions of Xe-Cluster in Intense Soft-X-Ray and X-Ray Pulses*	
5:30 PM	*Dinner*	*Dinner*		
7:00 PM	Panel Discussion: *New Frontiers in Ultrafast X-ray and Electron Sources*	Poster Jam Session: 3 minute talks	*Dinner/Banquet*	
8:00 PM		Posters		
9:00 PM				
10:00 PM				

Ultrafast X-ray and Electron Sources I

Probing molecular dynamics with short X-ray pulses from a synchrotron.

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Abstract
Fast protein nanocrystallography

We have examined the structure of laser excited molecules in solution by X-ray scattering using short pulses of X-rays from the European Synchrotron. The experiments are performed on beamline ID09B, a beamline for pump-probe experiments in physical, chemical and biological systems. Fast reactions are typically triggered by ultrafast optical pulses and the scattering (or diffraction) from delayed 100 ps pulses of X-rays are used to probe that structure of the sample at that time. I will review the latest experiments and beamline techniques, in particular the installation of a fast FReLoN CCD detector that has increased the efficiency of the beamline by a factor 5-10. It is now possible to record around 1000 scattering spectra per hour, which highlights the need for on-line data analyses. Finally we will show our plans for a new advanced pump-probe beamline to be built in 2012 within the framework of the ESRF upgrade program.

The LCLS X-Ray FEL Facility

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Abstract

The Linac Coherent Light Source (LCLS) at the SLAC National Accelerator Laboratory is a free-electron laser based on self-amplified spontaneous emission (SASE) in the wavelength range 1.5 15Å. It includes an FEL undulator about 100m in length, driven by high-brightness electron pulses with energy in the range 4.3-13.6 GeV prepared by a photoelectron gun and about 1km of linac. The FEL x-ray pulses can be directed into any one of 6 experimental stations, which are being optimized for various types of experiments. During 2009, LCLS began commissioning and supported experiments in the first operational experimental station, optimized for studying the interaction of soft-x- ray FEL pulses with atoms, molecules, and clusters in the gas phase. Lasing to saturation was achieved throughout the LCLS design energy range, with every indication that the facility can ultimately provide a significantly wider FEL photon energy range. By adjusting electron parameters, FEL pulse widths were adjusted between about 10 and 300fs. Pump-probe experiments with 50fs resolution were demonstrated using a Ti:sapphire laser pump and LCLS x-ray probe. The overall stability and reliability of the LCLS x-ray source rivals that of synchrotron sources. In summary, during early operation LCLS has proven to be a highly flexible and precisely-controllable x-ray source, and has already exceeded all of its technical design goals. Outfitting of the remaining experimental stations is well underway, with a three new stations expected to be commissioned in 2010. Plans for a major upgrade to the facility have already begun, promising increased photon energy range and experimental capacity by about 2017.

A new generation of soft x-ray free electron lasers

R.W. Falcone, K. Baptiste, J. M. Byrd, J. Corlett, P. Denes, L. Doolittle, H. Gang, J. Kirz, W. McCurdy, H. Padmore, G. Penn, J. Qiang, D. Robin, F. Sannibale, R. Schoenlein, J. Staples, C. Steier, M. Venturini, W. Wan, R. Wells, R. Wilcox, A. Zholents

Lawrence Berkeley National Laboratory Berkeley, CA, USA email: rwf@berkeley.edu

Abstract

Recent reports have identified the scientific requirements for a future soft x-ray light source and a high-repetition-rate free-electron laser (FEL) facility responsive to them is being studied at Lawrence Berkeley National Laboratory. The facility is based on a continuous-wave superconducting linear accelerator with beam supplied by a high-brightness, high-repetition-rate photocathode electron gun operating in CW mode, and on an array of FELs to which the accelerated beam is distributed, each operating at high repetition rate and with even pulse spacing. Dependent on the experimental requirements, the individual FELs may be configured for either self-amplified spontaneous emission, seeded high-gain harmonic generation, echo- enabled harmonic generation, or oscillator mode of operation, and will produce high peak and average brightness x-rays with a flexible pulse format ranging from sub-femtoseconds to hundreds of femtoseconds. This new light source would serve a broad community of scientists in many areas of research, similar to existing utilization of storage ring based light sources.

We are developing a design concept for a 10-beamline, coherent, soft xray FEL array powered by a 2.5 GeV superconducting accelerator operating with a 1 MHz bunch repetition rate. Electron bunches of charge 10 pC to 1 nC are fanned out through a spreader, distributing beams to an array of 10 independently configurable undulators and FEL beamlines with nominal bunch rates up to 100 kHz. Additionally, one beamline (the last in the array) could be configured to operate at higher repetition rate of 10 MHz or greater, in a dedicated operating mode, while simultaneously operating the other nine FEL beamlines at 100 kHz. The FELs may be seeded by optical lasers to control the X-ray output characteristics or may use SASE techniques, including generation of low-charge, high-brightness bunches with intrinsically short duration. Users specify the wavelength, pulse duration, and polarization, so that the 10 simultaneously operating beamlines can be individually optimized for specific experiments, including broad spectral coverage and multiple beam capability. The spectral range is from 10 eV to 1 keV, with harmonics to approximately 5 keV at reduced intensity. The beams may also be synchronized with optical lasers or IR and THz sources for pumpprobe experiments. Three principal modes of operation are proposed: ultrashort pulse (300 as-10 fs), short pulse (10 fs-100 fs), and high spectral resolution (requiring pulses from 100-500 fs). The spectral bandwidth in each mode is anticipated to approach fundamental transform limits. Other features include the capability to achieve high peak power ($\sim$1 GW) for nonlinear optics, control of peak power to reduce sample damage, and high average power ($\sim$1-10 W) for low-scattering-rate experiments. With up to 10 FEL beamlines and 20 X-ray beamlines, the facility will be capable of serving $\sim$2000 users per year.

Key Laser Technologies for Future X-ray Sources

Franz X. Kärtner[1], William S. Graves[2] and David E. Moncton[2]

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Abstract

Over the last few years, advances in femtosecond lasers have opened up the possibility to construct fully coherent soft and hard x-ray sources that range from table-top size to kilometer long seeded FELs. The later facilities will be combined laser and accelerator laboratories. In this presentation, we discuss some of the laser technologies and physics central to the development of such sources, including novel concepts for compact x-ray sources based increasingly, and perhaps entirely, on lasers. First, we explain the origin of ultralow timing jitter of femtosecond lasers and discuss the consequences for the control of electron-laser interactions with the precision of a few attoseconds, or potentially better. As an example, long term stable timing distribution for large scale x-ray FELs is shown. Systems designed at MIT now operate with sub- 10 fs precision over multiple days and are currently implemented at the FERMI FEL in Trieste. Timing distribution systems approaching attosecond level precision appear possible. Second, we discuss the production of laser radiation in the EUV and XUV via high harmonic generation. We have derived and experimentally verified closed form analytic expressions for high harmonic conversion efficiencies, verified them experimentally, and predict the possibility of highly efficiency EUV sources, which may achieve 1% conversion of optical power into a single harmonic for wavelengths as short as 13.5 nm. Such sources can stand alone for EUV lithography, for example, or be used for seeding of FELs to reach hard x-ray wavelengths with high longitudinal coherence. Third, we discuss our progress in the development of an energy and power scalable single-cycle waveform synthesizer based on a few-cycle optical parametric chirped pulse amplifier (OPCPA) system delivering synchronized 800 nm and 2 micron pulses for attosecond pulse generation. The pump laser system developed as the OPCPA driver is based on cryogenically cooled Yb:YAG. The large average power capabilities of cryogenically cooled Yb-doped lasers together with advances in superconducting accelerator technology enables ultrafast, bright and intense x-ray sources based on Inverse Compton Scattering (ICS). In particular, we consider sources that are based on high repetition rate (100 MHz), high brilliance electron beams from continuous wave superconducting accelerators operating at 4K. The pulsed electron beam collides with a 1 MW optical beam of pico- or femtosecond laser pulses in an enhancement cavity fed by a kW-class cryogenically cooled Yb-laser. Such a source is suitable for a university or industrial laboratory and can generate quasi monochromatic x-ray beams with average flux and brightness similar to a second generation synchrotron. Furthermore, the ICS-source has a spot size of a few microns (much smaller than a synchrotron beam) enabling improved high resolution phase contrast imaging and protein crystallography using 10-micron sized crystals. Since the source output consists of ultrafast x-ray pulses, time-resolved x-ray diffraction experiments in the sub pico-second regime are possible. Low repetition rate sources generating femtosecond pulses of up to 10^{10} photons appear to be feasible. In the future, attosecond control of the electron emission from nanostructured photocathodes and laser acceleration may produce fully coherent x-rays from sources exploiting the ICS geometry. We discuss the physics and beam properties of such sources as time allows.

Breaking the attosecond, Angstrom and TV/m field barriers with ultra-fast electron beams

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Abstract

Recent initiatives at UCLA concerning ultra-short, GeV electron beam generation have been aimed at achieving sub-fs pulses capable of driving X-ray free-electron lasers (FELs) in single- spike mode. This uses of very low charge beams, which may allow existing FEL injectors to produce few-100 attosecond pulses, with very high brightness. Towards this end, recent experiments at the Stanford X-ray FEL (LCLS, first of its kind, built with essential UCLA leadership) have produced $\sim$2 fs, 20 pC electron pulses. We discuss here extensions of this work, in which we seek to exploit the beam brightness in FELs, in tandem with new developments at UCLA in cryogenic undulator technology, to create compact accelerator/undulator systems that can lase below 0.15 Angstroms, or be used to permit 1.5 Angstrom operation at 4.5 GeV. In addition, we are now developing experiments which use the present LCLS fs pulses to excite plasma wakefields exceeding 1 TV/m, permitting a table-top TeV accelerator for frontier high energy physics applications. In this scenario, one focuses the beam to $\sim$100 nm transverse dimensions, where the surface Coulomb fields are also at the TV/m level. These conditions access a new, novel regime for high field for atomic physics, allowing frontier atomic physics experiments, including sub-fs plasma formation via barrier suppression ionization (BSI) for subsequent wake excitation. Plans for experiments at SLAC based on achieved beam parameters are presented, in which we evaluate the schemes for beam focusing, BSI ionization, TV/m plasma wakefields excitation and ion collapse.

Ultrafast X-ray and Electron Sources II

In-air femtosecond X-ray source

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Abstract

The dynamical behavior of a crystalline structure is not only of scientific interest, but also has technological importance. For instance, the ultra-fast phase transition used in recording materials and laser-induced recrystallization for electrical devices are studied intensively. However, the transitional mechanism of these materials in the femtosecond time scale has to be well understood. In order to develop advanced materials and processing with better performance, fundamental considerations are quite important. To explore various materials, there is a strong need for a compact and easyto-use femtosecond X-ray source.

We have demonstrated that high-reputation rate and low peak power laser can deliver an amount of X-ray similar to that generated with a low reputation rate and high peak power laser. A high reputation rate and low peak power laser is commercially available nowadays. In addition, the new X-ray source can be operated in He ambient. This new compact X-ray source is quite useful for analyzing many different materials.

We will report on the performance of this X-ray source and discuss the possible applications for ultra fast phenomena.

Synchrotron Radiation from Laser Accelerated Electrons

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Abstract

Femtosecond laser pulses have revolutionized the knowledge of intramolecular and microscopic solid state dynamics in the last two decades. This became possible since the duration of the light pulses is on the order of the characteristic microscopic time scales, and the photon energy is in the range of relevant electronic excitations. Transition states of photoinduced molecular and condensed phase dynamics can be observed in real time by applying a pump probe spectroscopy technique with ultrashort laser pulses or even femtosecond laser-generated electron pulses. However, the wavelength regime accessible for femtosecond lasers is limited around the visible spectral range and thereby restricts the interaction with matter to electronic transitions and their coupling to the atomic motion.

Shorter wavelengths down to a few nanometers can be generated using electron storage rings or linear accelerators equipped with undulators. This synchrotron radiation opens a more direct view into intermolecular or solid state dynamics via time-resolved photon diffraction in crystals and recently also of molecules which is of interest for a wide range of interdisciplinary research. If an undulator is operated in the free-electron-laser mode (FEL), extremely brilliant, ultrashort, polarized, and coherent light pulses are produced. FELs promise a wide applicability, spanning from atomic and cluster physics through temporally resolved structural analysis of complex molecules to plasma physics. However, they require km long LINACs producing several GeV electron energies due to the limited energy gain per length of less than 50 MeV/m. Bridging the gaps between femtosecond laser spectroscopy and synchrotron radiation sources may become possible with relativistic laser plasma physics. Femtosecond lasers can be used to generate light intensities exceeding 1020 W/cm^2, providing fields strong enough for electron particle acceleration up to GeV within a few mm, with a few percent bandwidth and within a well-collimated beam [1]. The energy gain per length for a laserplasma accelerator is significantly larger than for radio-frequency accelerators, because the acceleration is based on a plasma.

In this paper, we discuss the status of generation of synchrotron radiation from laser-accelerated electrons. A proof of principle experiment was reported by the authors [2,3], but significant improvements in terms of energy, shot-to-shot reproducibility, pointing stability, and spectral width of the driving electron beam have been realized since then. We discuss the potential and the limitations of this novel all-optical synchrotron light source, as it might become an interesting ultrashort pulsed (fs), tuneable VUV to x-ray coherent source, being smaller and more flexible compared to accelerator-based sources.

[1] Leemans et al. Nature Physics, 2, 696 (2006), [2] Schlenvoigt et al. Nature Physics, 4, 130 (2008), [3] Schlenvoigt et al IEEE Trans. Plasma Sci., 36, 1773 (2008).

Single-shot Ultrafast Electron Diffraction

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P.L.E.M. Pasmans, M.P. Reijnders, E.P. Smakman, G. Taban, E.J.D. Vredenbregt

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Abstract

The development in recent years of ultrafast electron diffraction (UED) techniques has enabled the first atomic-level, sub-ps studies of condensed matter phase transition dynamics. UED has also been applied successfully to determine transient molecular structures with 1 ps resolution during a chemical reaction of small molecules in the gas phase. Unfortunately, however, the application of UED up to now has mostly been limited to processes which are sufficiently reproducible, because recording a full diffraction pattern of sufficient quality requires $\sim 10^6$ electrons, corresponding to, typically, at least 100 shots. The number of electrons in a pulse is limited by space-charge forces, which cause rapid expansion of the pulse and therefore loss of temporal resolution. A possible way out is to accelerate the electron bunches to relativistic speeds, which slows down the space-charge expansion and thus allows single-shot UED with sub-ps resolution.

We have developed a method to produce sub-ps electron bunches suitable for single-shot UED at non-relativistic energies. The method relies on the use of radio-frequency (RF) techniques to invert the space-charge expansion. We will report on the first experiments demonstrating RF compression of 0.1 pC, 100 keV electron bunches. We have used these bunches to produce high- quality, single-shot diffraction patterns of poly-crystalline gold.

In all UED experiments up to now electron bunches have been generated by femtosecond photoemission from metal cathodes The transverse coherence length of the ensuing beams is limited to a few nm for crystal samples of ~ 100 μm size, and therefore does not allow the study of, e.g., protein samples. As reported elsewhere, we are developing an ultracold electron source which should enable coherence lengths of a few tens of nm for crystal samples with a size of ~ 100 μm. We will show that 0.1 pC, sub-ps, 100 keV electron bunches can be extracted from such a source, while retaining the transverse beam coherence, by applying RF acceleration and phase-space manipulation techniques. This should enable single-shot studies of macromolecular crystals.

Experimental realization of an ultracold electron source

E.J.D. Vredenbregt, G. Taban, M.P. Reijnders, E.P. Smakman, W.J. Engelen, S.B. van der Geer, and O.J. Luiten

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Abstract

We report on the development of an ultracold electron source, which is based on near-threshold photo- or field-ionization of a cloud of laser-cooled atoms. Such a source offers the unique combination of low emittance and extended size that may be essential for achieving single-shot, ultrafast electron diffraction of macromolecules. As reported elsewhere, a photo-emission electron source that could provide $\approx 10^6$ electrons in a 200 μm rms spot size with a 3 nm coherence length is currently un- der development in our labs. However, sources that provide even larger coherence lengths for a similar amount of electrons in a similar spot size are required to study the dynamics of larger objects such as proteins. An appropriate source must have a small enough emittance in order for all electrons to contribute to the diffraction pattern at the required coherence length. In addition, the local electric accelerating field at the source must be substantially larger than the field due to image charges in order for the pulse not to be lengthened and transversely distorted. The first criterion can be met by a variety of sources as it represents a trade-off between the source size and the effective source temperature. The second criterion, however, favors an extended source, such as the ultra-cold electron source presented in this contribution. Here we present measurements of the effective temperature of such a pulsed electron source employing rubidium atoms that are magneto-optically trapped at the center of an accelerator structure. Transverse source temperatures ranging from 200 K down to 10 K are demonstrated, controllable with the wavelength of the ionization laser. Together with the 50 μm source size, the achievable temperature enables a transverse coherence length of $\approx$20 nm for a 100 μm sample size. On the order of 10^5 electrons are contained in a (calculated) $\approx$50 ps long pulse when the trapped atoms are first converted to a “frozen”Rydberg gas from which electrons are extracted by a fastelectric field pulse.

RF photoinjector based ultrafast relativistic electron diffraction

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Abstract

Electron diffraction holds the promise to yield real time resolution of atomic motion in a easily accessible environment like a university laboratory at a fraction of the cost than 4th generation x- ray sources. Currently the limit in time-resolution for conventional electron diffraction is set by how short an electron pulse can be made. A possible solution to maintain the highest possible beam intensity without excessive pulse broadening from space charge effects is to increase the electron energy to the MeV level where relativistic effects significantly reduce the space charge forces. Rf photoinjectors can in principle deliver up to 10^7 -10^8 electrons packed in bunches of $\sim$100 fs length allowing an unprecedented time resolution and enabling the study of irreversible phenomena by single shot diffraction patterns. The UCLA Pegasus laboratory has recently demonstrated time resolved single shot electron diffraction using a $\sim$200 fs long relativistic beam from an rf photoinjector. We use this novel technique to study the evolution of the laser induced solid-liquid transition in metal foils of different thicknesses. The preliminary results of this experiment and the future directions of ultrafast electron diffraction with relativistic electrons will be discussed.

Instruments and Techniques for Ultrafast Structural Dynamics

Hard X-Ray Emission Spectroscopy

Pieter Glatzel[1], Gyorgy Vanko[2], Christian Bressler[3], Marcin Sikora[4], Amelie Juhin[5], Frank de Groot[5], Simo Huotari[1], Grigory Smolentsev[6]

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Abstract

Inner-shell spectroscopies using hard X-rays provide an element-selective and truly bulk- sensitive probe with great flexibility regarding the sample environment. Analysis of the emitted X-rays (XES) [1, 2] as opposed to scanning the energy of the incident X-ray beam to measure the absorption (XAS) appears to be an attractive option for upcoming 4th generation sources. The instrumentation and theoretical understanding of X-ray emission spectroscopy has made important progress and the technique is nowadays routinely used for the characterization of the local coordination and electronic structure.

XES includes a number of techniques. The least challenging in terms of instrumentation is non-resonant excitation of the sample with an incident beam of large energy bandwidth (tens of eV). Non-resonant XES may provide information on the oxidation and spin-state as well as the ligand orbitals. Resonant XES or resonant inelastic X-ray scattering (RIXS) requires a monochromatic beam that is tunable within a few eV around the Fermi energy. RXES is used to study electron-electron interactions and crystal field splittings in detail. The technique also enables to measure charge-neutral (i.e. non-ionizing) and element-selective excitations within the valence band. The spectral range is thus similar to UV-Vis spectroscopy but with different selection rules for electron transitions and the energy range can be extended well beyond 6 eV that limits standard optical spectroscopies.

The presentation will provide in introduction to the various techniques and discuss their potential for applications at hard X-ray free electron lasers. The instrumentation for single shot experiments and the feasibility will be addressed.

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Phase sensitive x-ray imaging and ultrafast chemical dynamics

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Abstract

Recent progress in the area of phase-sensitive x-ray imaging of bio-medical tissues as well as density waves in materials is discussed. Furthermore, recent 2-ps resolution x-ray absorption data from our experiment at the Advanced Photon Source (APS), ID7-C will be presented.

The high transverse coherence of the x-rays produced form laser-driven x-ray sources has been used for in-line holographic hard x-ray imaging of murine livers as well as clathrate hydrate slurries. The employed phase-sensitive x-ray imaging method is fundamentally different from conventional x-ray shadowgraphy because the mechanism of image formation does not rely on differential absorption by matter. Instead, x-ray beams undergo differential phase shifts and subsequently interfere constructively or destructively at the x-ray detector. Hence, material densities are distinguished by the differences between the real parts of their refractive indices rather than their absorptive properties. Example images of cancer bearing livers are presented. The chemical application of x-ray phase contrast imaging aims to observe the melting dynamics of clathrate hydrates in water solutions. These compounds are examples of chemical guest-host systems and are of interest for the capture of CO_2 and contaminant gases from power plant flue gases.

Recently, the first x-ray absorption spectroscopic measurements of the ligand substitution of $Fe(CO)_5$ have been carried out at the APS with 2-ps temporal resolution. This resolution is achieved in a 400-nm pump x-ray probe arrangement by detecting the x-ray pulses transmitted through the sample solution with a steak camera after photo excitation. An ultrafast Fe K-edge shift with subsequent recovery has been observed, which is consistent with impulsive Fe-CO bond elongation and recovery.

Theory and Simulation of Time-Resolved X-ray Diffraction

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Abstract

Pulsed x-ray sources can be used for real-time observation of chemical dynamics [1]. Recently, we derived the basic theoretical formulation for x-ray diffraction with pulsed fields using a fully quantized description of light and matter [2], which is in contrast to previous accounts on time-resolved x-ray diffraction on dynamic non-equilibrium structures, where the pulsed radiation field was treated classically [3-5]. We present some of the key features of our derivation and apply the theory to the laser-induced bond dynamics of simple molecules in particular, in the context of the upcoming free-electron x-ray lasers producing high-intensity x-ray pulses with duration of 100 femtoseconds or less [6,7]. The talk will highlight the differences between the expression for the time-dependent scattering signal we obtain from a first principles treatment and what one gets from just “adding”time to the expression for signal from time-independent scattering theory, and we will argue why the latter may work well for difference scattering images. The talk will also touch upon two issues that become important when moving from the 100 ps time resolution available at current synchrotron sources to 100 fs time resolution, namely laser-induced anisotropy and the importance of taking both vibrational population and vibrational hole dynamics into account.

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First Experiments with the AMO Instrument at LCLS

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Abstract

An instrument has been designed, built and commissioned to take advantage of the unique ultra-fast duration and ultra-intense x-ray beam of the Linac Coherent Light Source (LCLS) for atomic, molecular and optical (AMO) physics experiments. The instrument was commissioned over the summer of 2009 and used for the first set of peer-reviewed and facility approved user experiments at the LCLS in the subsequent months through the end of the year. Without exception the experiments were successful and numerous exciting new results were obtained, some of which are reported separately here at this meeting. The design and performance of the AMO instrument along with the performance of the LCLS will be presented here.

The LCLS is the first of three x-ray free electron lasers (FELs) being built in the U.S., Japan and Germany to begin operations. From the first time electrons were accelerated in the linac and injected into the undulators to the most recent experiments, the LCLS has been fantastically successful. In spite of early reservations among numerous reviewers evaluating the x-ray free electron laser concept, the LCLS lases robustly. The source has been very dependable in its first five months of operation with very few (and short) unscheduled down times. The LCLS x-ray FEL source has also proved to be very versatile, producing pulses ranging in duration from a few fsec to 300 fsec over a photon energy range of 800-2000eV with pulse energies up to 3.5mJ. Currently in a scheduled maintenance period, the LCLS will begin delivering its design goal 0.15nm radiation to the first hard x-ray experiments, when it is started up again in May 2010, satisfying another design goal of the facility.

The AMO instrument was designed to capitalize on the unique properties of the short intense pulses of x-rays generated by the LCLS to study some of the simplest forms of matter; atoms, molecules and clusters. It consists of focusing optics that produce a 1um focus in the interaction region of the first experimental chamber and 5um in the second chamber. Two experimental chambers are located about 1m and 3m downstream of the optics. The first chamber utilizes a skimmed, pulsed supersonic jet to introduce sample into the middle of an ion time-of-flight (TOF) spectrometer and five electron TOF spectrometers. Downstream, in the second chamber, a capillary is used to inject a steady stream of gas into the interaction region of a magnetic bottle spectrometer. IR or higher harmonics from a synchronized optical laser have been used for pump-probe experiments in both chambers. Special attention was paid to the data acquisition system to be able to handle the large amounts of data resulting from measurement of complete spectra from all instruments for each shot of the LCLS.

First results on nonlinear dynamics in diatomic molecules using the LCLS free electron laser

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Abstract

The unprecedented peak power at x-ray wavelengths of the Linac Coherent Light Source (LCLS) at the SLAC National Accelerator Laboratory, was used to study ultra fast, nonlinear and x-ray multiphoton physics in molecules. We report on fundamental questions concerning the creation and decay of multiple core-holes and, in particular, double core-holes in N_2 . We investigated both the Auger and secondary electron relaxation pathways subsequent to multiple core vacancies in molecules, and the fragmentation patterns and charge-state distributions of the resulting ions as function of wavelength, pulse duration and intensity. The new light source allows the characterization of complex molecular ionization and dissociation dynamics and provides new insight into the correlated motion of the electrons remaining in the targets and into fundamental aspects of ultrafast molecular physics and chemistry. In addition our work contributes to the foundation for future imaging experiments on molecules. The LCLS photon beam was focused to about 1μm diameter spot producing an intense x-ray laser beam of up to 10^{18} W/cm^2 , sufficient to investigate multiphoton, multiple core-holes, and multiple-ionization processes.

The experiment was performed at the AMO beamline, which is equipped with an ion time-of-flight spectrometer to determine the charge state and kinetic energy distribution of the ions as well five angle and energy resolving electron time-of-flight spectrometers to detect the emitted photoelectrons and Auger electrons.

This work was supported by the DOE-SC-BES, Chemical Sciences, Geosciences and Biosciences Division.

In-situ observation of irreversible reactions in liquids and gases by Dynamic Transmission Electron Microscopy (DTEM)

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Abstract

In response to a need to be able to observe dynamic phenomena in materials systems with both high spatial (~1nm or better) and high temporal (~1μs or faster) resolution, a dynamic transmission electron microscope (DTEM) has been developed at Lawrence Livermore National Laboratory (LLNL). The high temporal resolution is achieved in the DTEM by using a short pulse laser to create the pulse of electrons through photo-emission (here the duration of the electron pulse is approximately the same as the duration of the laser pulse). This pulse of electrons is propagated down the microscope column in the same way as in a conventional high- resolution TEM. The only difference is that the spatial resolution is limited by the electron- electron interactions in the pulse (a typical 10ns pulse contains ~10^8 electrons). To synchronize this pulse of electrons with a particular dynamic event, a second laser is used to "drive"the sample a defined time interval prior to the arrival of the laser pulse. The important aspect of this dynamic DTEM modification is that one pulse of electrons is used to form the whole image, allowing irreversible transitions and cumulative phenomena such as nucleation and growth, to be studied directly in the microscope. The use of the drive laser for fast heating of the specimen presents differences and several advantages over conventional resistive heating in-situ TEM such as the ability to drive the sample into non-equilibrium states. So far, the drive laser has been used for in-situ processing of nanoscale materials, rapid and high temperature phase transformations, and controlled thermal activation of materials. In this presentation, a summary of the development of in-situ stages for both the existing DTEM at LLNL and a new DTEM being installed at UC-Davis will be described. Particular attention will be paid to the potential for gas stages to study catalytic processes and liquid stages to study biological specimens in their live hydrated state. The potential improvements in spatial and temporal resolution that can be expected through the implementation of upgrades to the lasers, electron optics and detectors used in the new DTEM will also be discussed along with the correlation of dynamic results with conventional high resolution imaging and spectroscopic methods in TEM.

Aspects of this work are performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory and supported by the Office of Science, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering, of the U.S. Department of Energy under Contract DE-AC52-07NA27344. Aspects of this work at UC-Davis were supported by DOE NNSA-SSAA grant number DE-FG52-06NA26213 and NIH grant number RR025032-01.

Transient Electric Fields Induced By Ultrafast Pulsed Laser Irradiation and Implications for Time-Resolved Reflection Electron Diffraction

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Abstract

Studies of ultrafast processes using time resolved reflection high energy electron diffraction have revealed unusual lattice contraction and expansion and phase transitions in a number of materials. The dynamic processes are initiated by ultrafast laser irradiation. Understanding the interaction of ultrafast pulsed laser with matter is thus critical for understanding these phenomena. It is also important for understanding the physics of laser ablation and the laser induced non- equilibrium carrier dynamics in metals and semiconductors, including plasmonics. When an intense laser pulse of femtoseconds (fs) in duration hits the surface of a targeted matter, it excites a hot electron gas. Part of the hot electrons is emitted from the surface in a way similar to thermionic emission. Electrons can also be emitted through multiphoton photoemission (MPPE) or thermally assisted MPPE. The emitted electrons travel at speeds that create transient electric fields (TEFs). To detect TEFs and study the dynamics of emitted electrons, we have developed a time resolved an electron beam imaging technique that allows us to measure TEFs above a sample surface at picoseconds time resolution. We have also developed a model of the TEFs based on the propagation of emitted electrons and the percentage of electrons escaping from the surface. The results will be reported for silicon and graphite. The measured field strength and direction change with time; at the pump laser fluence of 67.7mJ/cm^2, the maximum field reaches 34 kV/m at 0.29 mm away from the silicon surface. We show that the TEF can induce large deflection of the reflected electron beams and changes in their intensity. The implications of our results for previous reported ultrafast structural studies will be discussed in the talk.

Structural Dynamics in Materials: Strongly Correlated Materials

Dynamics of cooperative lattice-charge (spin) coupled phenomena induced by fs laser light irradiation studied by time-resolved X-ray diffraction

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Abstract

We make a report on the pico-second dynamics of normal and super lattice structures triggered by fs laser irradiation utilizing ps time-resolved x-ray diffraction technique in the thin film of manganite with charge and orbital ordering; $(Nd_{0.5}Sr_{0.5})MnO_3$. The Jahn-Teller distortion becomes weak, i.e. orbital ordering melts, in the photo-induced state leading to the large changes in optical and magnetic properties, though structural coherence is kept even well after excitation. The obtained results shows that the origin of the gigantic photo-response is due to appearance of new state characteristic only for light-induced far-equilibrium condition but not the simple mixture of orbital ordered and disordered states in nanometer scale as like thermally induced phase change

Optical Control in Complex Solids

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Abstract

In this talk I will cover some of our recent work in studying photo-induced dynamics in complex solids. I will focus on the time dependent response of Peierls insulators and on measurements in Mott Insulators on the timescale of hopping and correlations. I will also discuss the case of half doped manganites, where we have combined a variety of time-resolved measurements, spanning THz to soft x-ray wavelengths, to understand how light pulses perturb electronic and lattice structure, as well as magnetic and orbital arrangements on the Ultrafast timescale.

Structural dynamics of the nearly commensurate phase in the Charge Density Wave compound 1T-TaS$_2$ probed by ultrafast electron diffraction

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Abstract

Femtosecond spectroscopy is becoming an important tool for investigation of the so called strongly correlated systems due to its intrinsic ability to determine the interaction strengths between various degrees of freedom which lead to the fascinating phenomena like superconductivity or colossal magnetoresistance. Low dimensional charge density wave (CDW) systems, with their inherently multi-component order parameter (modulation of carrier density is accompanied by the modulation of the underlying lattice) present no exception. In the past decade various one and two dimensional CDWs have been studied by time-resolved optical[1−5] as well as photoemission[6,7] techniques focusing on the dynamics of photoexcited electrons and collective modes. Recently, first systematic studies on the photoinduced melting of the CDW order has been reported, where the results suggest that on the sub-picosecond time scale when melting and subsequent initial recovery of the electronic order takes place the lattice remains unperturbed in its modulated state[8].

Here we report on the first studies of photoinduced CDW transition where the dynamics of the CDW modulation following photoexcitation with an intense optical pulse was probed directly by means of ultrafast electron diffraction. The results demonstrate an extremely fast suppression of the CDW modulation (within ∼200 fs) and the sub-picosecond recovery dynamics. The possible mechanisms of such rapid recovery of the CDW order are going to be discussed.

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Coherent Phonon in Iron Pnictide Superconductor Ba(Fe$_{1-x}$ Cox)$_2$ As$_2$ (x=0.06 and x=0.08)

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Abstract

What's the role of phonon in high temperature superconductivity? The opinions of the scientists on this theme diverge, giving rise to interesting debate and fancy exper- imental essays. The recent discovery of high critical temperature superconductivity in iron pnictide compounds has driven the attention of a large and multidisciplinary community. In these complex materials, the interplay between all the degrees of freedom of the crystal such as spin, charge and lattice, entails the existence of an interesting phase diagram. Here, understanding the role of phonon in the supercon- ducting phase transition is a key point to better understand the occurrence of the superconductivity in these compounds. We will report on the first study of coherent A1g optical phonon mode in the superconductor iron pnictide Ba(Fe$_{1-x}$ Cox)$_2$ As$_2$ (x=0.06 and x=0.08) [1], excited and detected in time domain in a pump and probe scheme by a 40 fs laser pulse. The transient reflectivity was measured for different crystal temperatures and doping. The optical phonon parameters such as amplitude, frequency and damping time, are measured across the superconductivity phase transition. Our results suggest that the A$_{1g}$ optical phonon mode do not participate to the superconductivity phase transition in these compounds.

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Structural Dynamics in Materials: X-rays

Coherent and incoherent femtosecond structural dynamics in solids studied by x-ray diffraction

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Abstract

The fundamental time scales for structural dynamics in a crystalline solid is set by the periods of the normal mode lattice vibrations, typically on the order of 100 fs for the fastest modes. Perturbation of the crystal on time scales comparable to or even shorter than these periods can lead to novel non-equilibrium structural phenomena. X-ray diffraction applied on the femtosecond time scale offers a way to directly study these non-equilibrium structural dynamics. The femtosecond slicing facility at the Swiss Light Source has in this way been able to apply x-ray diffraction to observe several different types of ultrafast structural phenomena in solids. In this talk we discuss examples including coherent phonons, phonon squeezing and the photo-induced melting of charge and orbital order in a manganite.

Real-time structural dynamics in materials on femtosecond and picosecond time-scales

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Abstract

The use of femtosecond x-ray pulses to probe materials opens up new windows into atomic- scale structural and electronic dynamics and the functional properties of materials through both x-ray scattering and x-ray absorption techniques. With the advent of new sources of femtosecond x-rays at synchrotrons and free electron lasers in recent years, the range of accessible time-scales and length-scales that can be probed has been dramatically increased, and provides new methods for elucidating how atoms move in materials in real time. In this talk, I will present recent work probing ultrafast dynamics in the solid and liquid phase, at atomic-scale resolution. We will present hard x-ray diffraction measurements of the first steps in the solid- liquid phase transition and the dynamics of the resulting disordered/liquid phase in both bulk and nanocrystalline systems, including the dynamics of a unique intermediate phase associated with superionicity at the nanoscale. We will show how ultrafast x-ray studies can be used to capture the polarization dynamics associated with perovskite ferroelectrics, leading towards all-optical control of the ferroelectric polarization. Finally we will present recent soft x-ray transmission measurements of ultrafast bond-breaking dynamics in the liquid phase of water.

Femtosecond x-ray powder diffraction

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Abstract
Fast protein nanocrystallography

We report on the first femtosecond x-ray powder diffraction experiment in which we directly map the transient electronic charge density in the unit cell of a crystalline solid with 30 picometer spatial and 100 femtosecond temporal resolution. X-ray diffraction from polycrystalline powder samples, the Debye Scherrer diffraction technique, is a standard method for determining equilibrium structures. The intensity of the Debye Scherrer rings is determined by the respective x-ray structure factor which represents the Fourier transform of the spatial electron density.

In our experiments, the transient intensity and angular positions of up to 20 Debye Scherrer reflections from a polycrystalline powder are measured and unravel for the first time a concerted electron and proton transfer in hydrogen-bonded $(NH4)_2$ SO_4 crystals. Photoexcitation of ammonium sulfate induces a sub-100 fs electron transfer from the sulfate groups into a highly confined electron channel along the z-axis of the unit cell. The latter geometry is stabilized by transferring protons from the adjacent ammonium groups into the channel. Time-dependent charge density maps derived from the diffraction data display a periodic modulation of the channels charge density by low-frequency lattice motions with a concerted electron and proton motion between the channel and the initial proton binding site. A deeper insight into the underlying microscopic mechanisms is gained by quantum chemical calculations with the result that the photo-excited electron from the sulfate groups triggers up 15 proton transfer events along the reaction trajectory $NH^+ + SO^{2-} \leftrightarrow NH_3 + HSO^-$.

Our results set the stage for femtosecond structure studies in a wide class of (bio)molecular materials.

Tracking Consecutive Steps of Photoinduced Switching Dynamics of Spin-Crossover Materials by X-ray Diffraction & Optical Pump-Probe Experiments.

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Abstract

Light may direct the functionality of a material through spectacular collective and/or cooperative photoinduced phenomena in the solid state. This can trigger the transformation of the material towards another macroscopic state of different electronic and/or structural order, for instance from non magnetic to magnetic or from insulator to conductor. This addresses photosteady instabilities as well as light pulse driven transformations. The increase of sophisticated instrumentation, including ultra-fast time-resolved diffraction [1], gives fascinating capabilities not only to observe and understand the elementary dynamic processes in materials but also to watch how matter works and can be directed to a desired outcome. We present here detailed investigation of the out-of-equilibrium spin-state switching dynamics of a molecular $\mathrm{Fe}^{(III)}$ spin-crossover solid triggered by a femtosecond laser flash. The time-resolved x-ray diffraction and optical results [2-4] show that the dynamics span from sub- picosecond local photo-switching followed by volume expansion on nanosecond time scale and thermal switching on microsecond) time-scale. We discuss a physical picture of the consecutive steps in the out-of-equilibrium dynamics associated with the photo-switching of such molecular materials.

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Short-pulse laser induced transient structure formation and ablation studied with time-resolved coherent XUV-scattering

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Abstract

XUV- and X-ray free-electron-lasers (FEL) combine short wavelength, ultrashort pulse duration, spatial coherence and high intensity. This unique combination of properties opens up new possibilities to study the dynamics of non-reversible phenomena with ultrafast temporal and nano- to atomic-scale spatial resolution. This contribution discusses results of time-resolved experiments performed at the XUV-FEL FLASH (HASYLAB/Hamburg) aimed to investigate the nano-scale structural dynamics of laser-irradiated materials. Thin films and fabricated nano- structures, deposited on Si_3N_4-membranes, have been excited with ultrashort optical laser pulses. The dynamics of the non-reversible structural evolution of the irradiated samples during laser- induced melting and ablation has been studied in an optical pump XUV-probe configuration by means of single-shot coherent scattering techniques.

In a first set of experiments we investigated the formation of laser induced periodic surface structures (LIPSS) on the surface of thin Si-films. Time-resolved scattering using femtosecond XUV-pulses at 13.5 nm and 7 nm allowed us to directly follow the LIPSS evolution on an ultrafast time-scale and with better than 40 nm spatial resolution. The observed scattering patterns show almost quantitative agreement with theoretical predictions and reveal that the LIPSS start to form already during the 12 ps pump pulse.

In the second set of measurements we studied picosecond and femtosecond laser induced ablation and disintegration of fabricated nano-structures. Time-dependent auto-correlation functions were obtained from the coherent diffraction patterns measured at various pump-probe time delays and reveal the expansion dynamics of the irraditated samples. Under certain circumstances (e.g. adequate sampling) it became also possible to reconstruct real-space images of the object as it evolves over time [1].

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Structural Dynamics in Materials: Electrons

Femtosecond Electron Diffraction: “Making the Molecular Movie”

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Abstract

Imagine one being able to follow chemical reactions and phase transformations with atomic spatial and temporal resolution. This dreamed experiment has been entitled “Making the Molecular Movie”(1). Recent advances in ultrafast time-resolved X-ray (2) and electron diffraction (1, 2) techniques have shown that such a dream became real. Femtosecond Electron Diffraction (FED) is very promising table-top technique that holds a great potential for the study of ultrafast structural phenomena of matter. In FED a femtosecond laser pulse excites the sample and the photoinduced structural changes are probed by an ultrashort electron pulse that scatters off the irradiated area to generate a diffraction pattern downstream. By varying the time delay between the excitation and the electron pulses atomic-level movies can be reconstructed after Fourier analysis. We have fully characterized our electrons pulses employing transient optical gratings in order to scatter electrons off via ponderomotive forces (3). With the development of our 4^{th} generation electron gun, we were able to reduce the electron pulse duration to 200 fs to provide enough time resolution and brightness to study structural changes of matter occurring under strongly driven nonreversible conditions. Nonthermal melting in Si caused by the promotion of 10% of its valence electrons to the conduction band (4), bond hardening in warm dense Au (5) and strongly accelerated atomic motions in Bi (6, 7) are some examples of the very different phenomena that were observed by FED. During my talk I will present an overview of the ongoing efforts put forward the development of ultrafast X-ray and electron diffraction techniques for the study of structural dynamics of matter and show some recent result obtained by FED at University of Toronto.

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Studying Nanoscale Material Processes in the Dynamic Transmission Electron Microscope (DTEM)

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Abstract

Often materials macroscopic properties and behavior under external stimuli can be described through observation of its microstructural features and dynamical behavior. Materials models and computer simulations that are used to predict material behavior in different environments, e.g., phase transformation kinetics under high pressure loading, typically require experimental data for validation or interpretation of simulated quantities. However, most materials dynamics are extremely rapid, making it difficult to capture transient, fine-scale features of the material process, especially on the length and time scale relevant for most simulations. In effort to meet the need for studying fast dynamics in material processes, we have constructed a nanosecond dynamic transmission electron microscope (DTEM) at Lawrence Livermore National Laboratory to improve the temporal resolution of in-situ TEM observations.

The DTEM consists of a modified JEOL 2000FX transmission electron microscope that provides access for two pulsed laser beams. One laser drives the photocathode (which replaces the standard thermionic cathode) to produce the brief electron pulse. The other strikes the sample, initiating the process to be studied. A series of pump-probe experiments with varying time delays enable, for example, the reconstruction of the typical sequence of events occurring during rapid phase transformations. This presentation will discuss the core aspects of the DTEM instrument citing specific examples for which the DTEM has been used to elucidate the kinetics of rapid martensitic phase transformations, the morphologies rapid solidification and chemical reaction fronts and high temperature crystallization processes in amorphous metallic films.

Work was performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore National Laboratory and supported by the Office of Science, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering, of the U.S. Department of Energy under contract No. DE-AC52-07NA27344.

Ultrafast Electron Diffraction at Surfaces: From Non-Thermal Heat Transport to Strongly Driven Phase Transitions

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Abstract

The multitude of possible processes that can occur at surfaces cover many orders of magnitude in the time domain. While large scale growth and structure formation, for instance, happens on a timescale of minutes and seconds, diffusion processes are already much faster. Energy transfer processes take place on the femto- and picosecond timescale and are important for electron excitation and relaxation, chemical reactions, phonon dynamics, nanoscale heat transport, or even phase transitions.

In order to study such ultrafast processes at surfaces we have combined modern surface science techniques with fs laser pulses in a pump probe scheme. We use a reflection high energy electron diffraction (RHEED) setup with grazing incident electrons of 7 - 30 keV to ensure surface sensitivity [1,2]. Utilizing the Debye Waller effect the cooling of vibrational excitations in monolayer adsorbate systems or the nanoscale heat transport through a heterofilm interface is studied on the lower ps-time scale [3-5]: the heat transport of ultrathin Bi(111) films on Si(001) is dominated by a pronounced non-equilibrium distribution in the phonon system resulting in a much slower cooling rate.

In order to demonstrate the huge potential of this technique I will shortly present examples for the dynamics of strongly driven structural phase transitions at surfaces upon excitation with a fs-laser pulse: the famous order-disorder phase transition from c(4x2) to (2x1) on Si(001) at 200 K and the Indium induced Peierls-like transition from c(8x2) to (4x1) on Si(111) at 80 K which is additionally accompanied by the formation of a charge density wave [6].

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Femtosecond Molecular Photocrystallography

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Abstract

Diarylethenes are a class of photochromic compounds which undergo well documented conformational changes in both the solution and crystal phase [1]. The photoreversible isomerization involves ring-closing and -opening of the molecular system, which leads to distinct absorptive features in the visible and UV spectral regions respectively. Of particular interest is the recent development of diarylethene derivatives that exhibit not only pronounced thermal stability of the open and closed-ring isomers, but also a high degree of fatigue resistance in the crystal phase suggesting the potential for optical switching and memory applications. Here we present preliminary results of a femtosecond electron diffraction (FED) study on such a derivative.

FED will provide a direct observation of the structural dynamics involved in the conformational changes of diarylethene with femtosecond time resolution and atomic level details [2]. Among other studies, FED has now been successfully used to study ultrafast structural dynamics in the order-to-disorder phase transition of strongly driven melting in gold [3], and the electronically driven melting of silicon [4]. In FED, an ultrashort laser pulse initiates the reaction in the sample under study and an electron bunch probes its structure via diffraction; by varying the time delay between the laser and electron pulses, the recorded diffraction patterns temporally resolves changes in the molecular structure. In the case of diarylethene, a third beam is required to bring the sample back to its initial state before the next pump-probe event. To complement the electron diffraction study, an optical pump-probe absorption measurement will first be performed to characterize the required experimental parameters and insure complete reversion to the initial conditions.

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Diffractive and Dynamic Imaging of Molecules

First Results of Coherent Diffraction Experiments at LCLS

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Abstract

The ultrafast pulses from X-ray free-electron lasers may enable the determination of structures of proteins that cannot be crystallized. The specimen would be completely destroyed by the pulse, but that destruction will ideally only happen after the termination of the pulse. In order to address the many challenges that we face in attempting molecular diffraction, we have carried out experiments in coherent diffraction from protein nanocrystals at the Linac Coherent Light Source (LCLS) at SLAC. The periodicity of these objects gives us much higher scattering signals in order to determine the effects of pulse duration and fluence on the high-resolution structure of single objects. The crystals are filtered to sizes less than 2 micron, and are delivered to the pulsed X-ray beam in a liquid jet. Diffraction patterns are recorded at the LCLS repetition rate with pnCCD detectors. Preliminary results will be presented on our first LCLS experiments. This work was carried out as part of a collaboration, for which Henry Chapman is the spokesperson. The collaboration consists of CFEL DESY, Arizona State University, SLAC, Uppsala University, LLNL, The University of Melbourne, LBNL, the Max Planck Institute for Medical Research, and the Max Planck Advanced Study Group (ASG) at the CFEL. The names and addresses of all do not fit on one page. The experiments were carried out using the CAMP apparatus, which was designed and built by the Max Planck ASG at CFEL. The LCLS is operated by Stanford University on behalf of the U.S. Department of Energy, Office of Basic Energy Sciences.

Fast protein nanocrystallography

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Abstract

The invention of the hard X-ray laser has opened the way for a new form of protein microcrystallography under an entirely new regime of radiation-damage conditions (1, 2). When combined with pump-probe methods, this "diffract-and-destroy"mode, in which an X-ray pulse terminates before damage begins, promises dramatic advances in the study of protein dynamics, and of structures which have never been seen at high resolution because of their radiation sensitivity. In this talk our recent diffraction data obtained at LCLS from individual sub-micron crystallites of Photosystem 1 membrane protein will be discussed, where femtosecond pulses (with repetition rate of 30 Hz) were used at 2 kV with a 3 micron X-ray beam diameter to obtain tens of thousands of patterns from individual crystallites fired in single-file across the beam by a protein-beam injector. Previous work at Flash (2), and simulations (3), have indicated the difficulties in phasing and orientation determination for single non-periodic bioparticles (such as viruses or single macromolecules) due to the very low counts at high angle (much less than unity). The Bragg amplification of coherent scattering in "stills"(snap-shot diffraction) from nanocrystals increases counts greatly, providing high resolution information, but requiring a new form of data analysis. Additionally, since Miller indices are coordinates in reciprocal space, the ability to index these stills solves the molecular orientation problem.

This talk will focus mainly on data analysis methods (4), in which, following indexing, we achieve a Monte-Carlo integration over particle size and orientation by adding together all "spots"(partials) with the same index from different crystals. Our crystals are roughly sorted by size, but are not identical particles. This makes whole-particle phasing a challenging excercise (5) since it requires sorting by both size and orientation - if that can be done, by selecting phases only on lattice points, a new method of phasing would be possible for protein crystallography. The method of aperture photometry (as used in Astronomy) is used to integrate over the crystal shape-transform on each pattern. Simulations showing the convergence of these orientation and size summations to yield wanted structure factors will be discussed. These address the question of how many patterns are needed for a required accuracy, with a given photon count per pulse. Details of the indexing method, of the protein-beam injector (6,7), of hit rates, and membrane protein hydration will also be discussed.

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Linear and nonlinear imaging with XFEL: results from ab-initio computations

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Abstract

The ultimate frontier of single molecule imaging with XFEL sources is currently hampered by several challenging questions concerning sample damage, time-gating imaging and the role of nonlinearity. By employing an original *ab-initio* approach, as well as exceptional resources of parallel computing, we provide a decisive answer to them. Our model, directly stemming from the quantum-mechanical equations governing the dynamics of atoms subjected to electromagnetic fields, try to definitively settle down the theoretical grounds for present and future *ab-initio* researches on XFEL science. More specifically, our approach combines classical molecular dynamics, nonlinear Scrhödinger and Maxwell's equations into a single efficient parallel environment, which features original second order propagators designed with state-of-the-art methods and algorithms. By analyzing a selection of atoms and molecules, we address the problem of sample radiation damage, thus predicting a large sample photoionization in a few of femtoseconds (with external electron emission in hundreds of attoseconds). We then deeply analyze the the scattered far field, highlighting the role of nonlinearity and anticipating the possibility to spread out the XFEL application domain to nonlinear coherent imaging. We finally investigate the coherent imaging capabilities of XFEL sources, collecting snapshots of integrated far field (as retrieved by a standard camera), reporting *ab-initio* molecular images and discussing image blurring versus XFEL pulse length.

Single shot soft X-ray holography using extended references

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Abstract

X-ray lensless imaging is demonstrating a very high potential in performing images of isolated nanoscale objects with unprecedented space and time resolution. Active research is actually pursued to push the capability of this technique using coherent X-ray sources recently available. In this context, we present a generalization of Fourier transform holography. A major advance shown here is the use of extended holographic reference to perform soft X-ray nanoscale imaging. The direct reconstruction process of the object is simple and robust. Moreover, the design of the holographic reference is easy to implement. We demonstrate here single shot imaging with table top soft X-ray source based on the high harmonics generation process. A spatial resolution of 110 nm is obtain with an integration time resolution of 20 fs. Using harder X-rays available at femtosecond X-ray free electron lasers, extended holographic references can be used to capture dynamical processes at a sub-nanometer scale and in real time.

Ultra-fast, Ängström Scale Structure Determination of Molecules via Photoelectron Holography

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Abstract

We examine a new scheme that enables us to realize a molecular movie with femtosecond time and Ångström spatial resolution for small and medium sized molecules based on the (i) upcoming brilliant X-ray Free Electron Laser (FEL) sources, (ii) novel energy and angular dispersive, large-area electron imagers and (iii) the photoelectron holography. Here, photoelectrons produced via core-level excitation and launched at specific and well-defined atomic sites will scatter on “their way out”on the multi-atomic potential of the parent molecule generating a hologram on the detector that encodes the molecular structure at the instant of photoionization. Due to the large photo-absorption and electron elastic scattering cross sections the method extends X-ray diffraction based, time-dependent structure investigations envisioned at FELs to new classes of samples that are not accessible by any other method. Among them are dilute samples in the gas phase such as aligned, oriented or conformer selected molecules, ultra- cold ensembles and/or molecular or cluster objects containing mainly light atoms that do not scatter X-rays efficiently.

Explosions of Xe-Clusters in Intense Soft-X-Ray and X-Ray Pulses

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Abstract

Intense femtosecond x-ray pulses from free electron lasers open the door for novel experiments in a wide spectrum of sciences ranging from atomic, molecular and plasma physics over chemical and surface dynamics to diffraction imaging of non-periodic objects and biological samples. The interaction of intense x-ray pulses with matter is so far only scarcely investigated, even though its understanding is a prerequisite for virtually all future experiments in this field. Clusters, bridging the gap between the atom and bulk solid, are ideal to investigate the light matter interaction. They exhibit the density of bulk solids but due to their finite size hidden energy dissipation into the surrounding media is virtually absent.

The presentation will show results of the interaction of Xe-Clusters consisting of up to <N> ~10,000 atoms with the FLASH radiation at a photon energy of 90 eV at a pulse length of 10 fs resulting in a maximum intensity of 8x10^{14} W/cm^2 in the focus. At this photon energy one photon can ionize the 4d-innershell electrons of xenon. The absorption of 90 eV-photons is rather complex for xenon including multi-photon processes and auger effects.

Simulating the ion kinetic energies in an electrostatic model suggests that highly charged ions explode off the surface due to Coulomb repulsion while the inner core expands in a hydro- dynamic expansion [1]. The current results yield evidence for efficient ionization of the clusters in addition to direct multistep photoemission [2,3]. Further a model for the induced multi- electron dynamics can be shown which reveals that fast electrons originate from an equilibrated electron plasma of supra-atomic density [3]. The plasma has sufficiently high temperature to support fast electrons without traditional laser plasma heating, which is not operative at 90 eV. This results will be compared to results of the very recent experiments at LCLS on xe-clusters at a photon energy up to 2 keV and similar pulse lengths. In the experiments at FLASH and LCLS ion- and electon-spectra were recorded using the time-of-flight-technique.

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Structural Dynamics in Molecules

Watching proteins function in real time via 150-ps time-resolved X-ray diffraction and solution scattering

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Abstract

To generate a deeper understanding into the relations between protein structure, dynamics, and function, we have developed X-ray methods capable of probing changes in protein structure on time scales as short as ∼100 ps. This infrastructure was first developed on the ID09B time- resolved X-ray beamline at the European Synchrotron and Radiation Facility, and more recently on the ID14B BioCARS beamline at the Advanced Photon Source. In these studies, a picosecond laser pulse first photoexcites a protein, then a suitably delayed picosecond X-ray pulse passes through the laser-illuminated volume of the sample and the scattered X-rays are imaged on a 2D detector. When the sample is a protein crystal, this pump-probe approach recovers time- resolved diffraction snapshots whose corresponding electron density maps can be stitched together into movies that unveil correlated protein motions at near atomic resolution. When the sample is a protein solution, we recover time-resolved small- and wide-angle X-ray scattering patterns that are sensitive to changes in the size, shape, and structure of the protein. Scattering studies of proteins in solution unveil structural dynamics without the constraints imposed by crystal contacts; thus, these scattering "fingerprints"at low spatial resolution complement results obtained from high-resolution diffraction studies. Studies of structural dynamics in wild-type and mutant scapharca hemoglobin, a homodimer that exhibits cooperative ligand binding, unveil non-exponential tertiary relaxation followed by a quaternary R to T structural change that alters the binding affinity of its two ligand binding sites. The structural dynamics characterized by X- ray scattering are highly correlated with spectral changes observed via time-resolved optical spectroscopy, thereby allowing us to make a structural assignment for the spectroscopic states. These studies are leading to a comprehensive characterization of the structural dynamics that contribute to the cooperative binding of ligands in this allosteric protein. *This research was supported in part by the Intramural Research Program of the NIH, NIDDK.*

But my crystals aren't light-sensitive....help!

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Abstract

Studying the structure-based, ultrafast dynamics of biological systems by e.g. Laue crystallography requires a means of initiating the reaction in the crystal rapidly, smoothly and with high efficiency. In practice, this has meant using a brief laser pulse in the fs to ns range, and restricting the crystals under study to those of naturally light-sensitive systems such as photoreceptors or the CO-complexes of heme proteins. This substantially restricts the applicability of ultrafast time-resolved crystallography, a fact not lost on peer reviewers.

There are two possible rejoinders: find other means of ultrafast rapid initiation e.g. temperature jump or dielectric relaxation, particularly challenging if time scales less than s are to be probed; or, confer light sensitivity on otherwise light-inert systems. In tackling the latter, we base our approach on key features of natural signaling photoreceptors: they are modular in architecture, containing several compactly-folded domains; and different functions are located in different domains. For example, they contain one or more sensor or input domains that respond to a physical signal e.g. absorption of light, or a chemical signal e.g. binding of a small molecule, and an effector or output domain whose activity e.g. catalytic, DNA binding is influenced by the signal. Thus information is transferred from the sensor domain to the effector domain. Further, the sensor domain(s) is usually located near the N-terminus of the effector domain, and is covalently joined to it by a linker that may be α-helical or a coiled coil. One class of sensor domain e.g. a blue-light-sensing LOV domain is found joined to many different types of effector domains. The last argues against structure-specific interaction between the sensor and effector domains.

We exploit these natural principles to confer sensitivity to light on the DNA-binding trp repressor (Strickland et al., PNAS 105, 10709-14 (2008)), a histidine kinase (Moeglich et al., J.Mol.Biol. 385, 1433-44 (2009)) and kinases with more than one sensor domain (unpublished). The last raises the additional complexity of interaction between signals: such molecules can act as logic elements whose output depends on more than one input.

This new area has been labelled "optogenetics"(see Miesenbock, Science 326, 395-9 (2009)), the genetic encoding of natural e.g. channelrhodopsin and artificial, designed light-sensitive systems.

Molecular Structural Dynamics Visualized by Pump-Probe X-ray Liquidography and Crystallography

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Abstract

The principle, experimental technique, data analysis, and applications of time-resolved X-ray diffraction and scattering to study spatiotemporal reaction dynamics of proteins in single crystals and solutions will be presented. X-ray crystallography, the major structural tool to determine 3D structures of proteins, can be extended to time-resolved X-ray crystallography with a laser-excitation and X-ray-probe scheme, and all the atomic positions in a protein can be tracked during their biological function. However time-resolved Crystallography has been limited to a few model systems with reversible photocycles due to the stringent prerequisites such as highly- ordered and radiation-resistant single crystals and crystal packing constraints might hinder biologically relevant motions. These problems can be overcome by applying time-resolved X-ray diffraction directly to protein solutions rather than protein single crystals. To emphasize that structural information can be obtained from the liquid phase, this time-resolved X-ray solution scattering technique is named time-resolved X-ray liquidography (TRXL) in analogy to time- resolved X-ray crystallography where the structural information of reaction intermediates is obtained from the crystalline phase. Using ultrashort optical pulses to trigger a reaction in solution and detecting time-resolved X-ray diffraction signals to interrogate the molecular structural changes, TRXL 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. Application examples on spatiotemporal kinetics and structural dynamics of a halomethane, a triatomic molecule, haloethanes, and an organometallic catalyst are presented. In addition, we demonstrate tracking of proteins structural changes in solution using TRXL. TRXL permitted us to investigate the tertiary/quaternary conformational change of human hemoglobin in nearly physiological conditions triggered by laser induced ligand photolysis. Data on optically induced tertiary relaxations of myoglobin and refolding of cytochrome c are also reported to illustrate the wide applicability of the technique. By providing insights into the structural dynamics of proteins functioning in their natural environment, TRXL complements and extends results obtained with time-resolved spectroscopy and X-ray crystallography.

Molecular Snapshot in Solar Energy Conversion Processes Taken by Ultrafast X-rays

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Abstract

A decade of studies on excited state structures of transition metal complexes for solar energy conversion using laser and x-ray transient absorption spectroscopy will be briefly reviewed including the details for the excited state dynamics, structural diversity in solution and hot vibrational states. We will discuss three examples on 1) metalloporphyrins excited state structure and photoinduced ligation/deligation, 2) interplays of structure and dynamics of MLCT excited state transition metal complexes for photoinduced charge separation and electron transfer, and 3) excited state transition metal complexes at interfaces of hybrid material for solar electricity generation/catalysis. The current advances and limitations in resolving excited state structures during photochemical reactions will be presented. New needs in theoretical computation and modeling will be addressed for these studies to exert the full potentials of resolving otherwise elusive excited state structures. The potential and prospective in excited state structural dynamics studies using new light sources, such as XFEL will be discussed.

Towards Femtosecond X-Ray Spectroscopies

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Abstract

Femtosecond X-Ray Science is an emerging field aiming to deliver a detailed understanding of the ultrafast elementary steps in complex processes involving changes in nuclear, electronic and spin states. Such processes are vital ingredients in chemistry and biology, but also in technological applications, including efficient charge transport in solar energy converters and ultrafast switchable molecular magnets.

This talk will present results obtained on a prototype spin transition phenomenon in aqueous Fe(bpy)$_3$. Optical techniques explore the ultrafast changes in the valence states, but ultrafast x-ray spectroscopies reveal the underlying nuclear and electronic changes during this spin transition process. While picosecond resolved XANES and EXAFS are exploited to understand the altered geometrical structure of the molecule after the spin transition is complete, Femtosecond XANES is able to monitor the evolution of this process in real-time. Finally, a recent experiment exploiting time-resolved XES of the Kα emission with picosecond resolution established a direct measurement of the short-lived (0.7 ns) high-spin state. Combing these spectroscopic tools with the intense intensity and femtosecond time resolution at x-ray free electron lasers will allow us to deliver a motion picture of the interplay between the nuclear, electronic and spin degrees of freedom during complex chemical reactions, and an outlook towards exploiting XFEL machines currently in operation or under construction will be given.

Photo-Induced Spin-State Conversion in Transition Metal Complexes Probed via Ultrafast Soft X-ray Spectroscopy

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Abstract

A precise understanding of the transient valence charge distribution in solvated tran- sition metal complexes is of great scientic interest due to their important role in chemical reaction and biological processes. By exploiting the capability of time- resolved L-edge spectroscopy to deliver unique information about transient valence electronic states in transition metal compounds, we investigated the photo-induced spin crossover reaction in a solvated iron (II) model complex via femtosecond soft x- ray spectroscopy. Our recent experimental results in combination with charge trans- fer multiplet calculations relate to important aspects of general chemistry and reveal a wealth of information on the changes of the electronic valence charge distributions and the role of ligand -back-bonding in different molecular structures. Upon photo-excitation to the singlet metal-to-ligand charge transfer state, the in- tricate coupling of nuclear and electronic degrees of freedom results in an ultra- fast singlet-to-quintet spin state conversion within 200fs mediated by large structural and electronic changes. The transient valence electronic structure of the metastable high-spin state features strongly altered orbital hybridization and delocalization, de- creased ligand-eld splitting, and strongly suppressed π-back-bonding, increasing the ionic character of the central transition metal atom in the dilated ligand cage.

Observation of multiphoton processes in the x-ray regime: First experiments at LCLS

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Abstract

The worlds first x-ray free electron laser, the LCLS at SLAC National Accelerator Laboratory, provides access to ultraintense x-ray radiation for the first time. Understanding the atomic response to such radiation is of fundamental importance for planning any future work where matter will be exposed to ultraintense x-ray beams. Therefore, we investigated the most basic aspects of intense x-ray/matter interactions by observing photoionization of the prototypical neon atom, an atom that exhibits rich physics over the initial energy range of 800-2000 eV. At high photon energy, one expects sequential single photon absorption to dominate the XFEL-atom interaction with, e.g., six-photon absorption leading to fully-stripped neon. Such processes depend only on the fluence of the radiation. However, with ultraintense x-ray radiation (focused intensities of $\sim 10^18$ W/cm^2) one can photoinduce sequential K-shell absorption prior to the intraatomic Auger decay (2.4 fs) to create exotic hollow atom states with high probability. By contrast, at low x-ray intensity hollow atoms are only formed indirectly via rare one-photon, two- electron processes that require electron correlation. The versatility of the LCLS allowed us to investigate the nature of photoelectric x-ray absorption processes over a wide range of photon energy, pulse energy and pulse duration. We tracked the evolution of the neon atom using one ion and five electron spectrometers to view the interaction region. The electron energy and angular distributions reveal details of the photoabsorption mechanism. We observed an intensity-induced transparency, photoproduction of hollow neon, and considerable valence ionization. The observations are qualitatively explained by sequential multiphoton processes. A comparison with a simple rate equation model demonstrates the need to include shake and double Auger processes for quantitative agreement. The simplicity of the neon target provides useful diagnostics of the XFEL beam.

Results of the CAMP Instrument Commissioning at LCLS

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Abstract

The CFEL-ASG MultiPurpose (CAMP) instrument designed and constructed by the Max Planck Advanced Study Group at CFEL has recently been commissioned during the first user run at LCLS in November/December 2009. The general layout and capabilities of the CAMP instrument will be reviewed and first results of the successful instrument commissioning will be reported.

Structural Dynamics with Bound Electrons: Isomeric and Conformeric Motions of Hot Molecules

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Abstract

The binding energy of a Rydberg electron that orbits a positively charged ion core is a uniquely sensitive probe of the structure of the underlying molecular ion core. The structure sensitivity can be traced to the very same phase shifts that give rise to electron diffraction patterns. When the electron binding energy is measured in an ionization transition, the resulting spectrum is free of vibrational progressions: the spectrum is purely electronic in character. Showing only the usual orbital and magnetic angular momentum states of the Rydberg electrons, the complexity of the spectra does not scale with the size of the molecular system. Moreover, since the Rydberg orbits are large compared to the dimensions of usual molecules, the structure sensitivity extents to the entire molecule. The global structure sensitivity coupled with the insensitivity towards vibrations makes Rydberg electron binding energy spectra ideally suited to observe structural dynamics, including transformations between isomeric and conformeric forms of highly excited molecules. The drawback of the technique is that unlike a diffraction pattern, the data cannot easily be inverted to obtain molecular structures. This talk outlines the essential features of the technique and illustrates it with examples from a series of investigations on tertiary amines.

All tertiary amines exhibit a very rapid structural change that can be traced to the initial planarization of the amine bond upon electronic excitation. In tripropylamine and trimethylamine, little further signature of structural dynamics is found. Triethylamine, however, shows a rich time-dependent spectrum. The ethyl groups of triethylamine, rotating about the C-N single bond, create a complex energy landscape that serves as a model system for conformational dynamics with highly coupled degrees of freedom. Electronic excitation to a 3p or 3s Rydberg level leads to a high-energy Rydberg state conformer that rapidly relaxes to other, more stable conformeric forms with a 232 fs time constant. A new equilibrium is established on a sub- picosecond time scale. Even so, the molecules retain a large dispersion of molecular structures about the equilibrium position. For the close-lying minima in the energy landscape, the variation of the Rydberg electron binding energy is the determining parameter of the landscape.

N,N-dimethylphenethylamine (PENNA), a molecule with two functional groups, is able to form an intramolecular cation-pi bond between a positive ion core at the amine site and the phenyl ring. Excitation of the initially stretched molecule to a 3p Rydberg state triggers the formation of the cation-pi interaction, which is seen in the binding energy spectrum as a sizable time- dependent shift. Structural dispersion in this system is again large, leading to a broad line width.

The Rydberg electron binding energy also depends strongly on the presence of neighboring molecules, opening an experimental avenue to study the kinetics of transitions between isomeric forms of molecular clusters. In tetramethylethyldiamine and dimethylpropylamine clusters, we observe that the binding energies of small molecular clusters ($n<10$) are shifted by about 0.5 eV from their monomer energies. The time dependence of the spectrum reveals the reorganization of the solvent surrounding the newly formed molecular ion core.

Poster Presentations

Ultrafast Electron Diffraction from Selectively Aligned Molecules

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Abstract

Electron diffraction has been very successful for determining the structure of molecules in the gas phase, and also for investigating ultrafast conformational changes. However, due to the random orientation of the molecules in the gas phase only 1D information (the interatomic distances) can be extracted from the diffraction patterns, which limits the size of molecular structures that can be studied. Having a sample of aligned molecules would greatly increase the information encoded in the diffraction pattern and potentially allow for reconstructing the full 3D molecular structure.

Here we show electron diffraction patterns recorded from a sample of transiently aligned molecules. In our experiments molecules are aligned selectively using photodissociation of $C_2F_4I_2$ (1,2- diiodotetrafluoroethane). The diffraction pattern is captured by probing the sample with picosecond electron pulses shortly after dissociationbefore molecular rotation causes the alignment to vanish. The transition dipole moment of $C_2F_4I_2$ is parallel to the C-I bond, along which the dissociation takes place. Therefore, the C_2F_4I radicals emerge preferentially with the dissociated C-I direction aligned along the laser polarization vector. Our results clearly show that the angular distribution of the molecules becomes anisotropic after dissociation. The alignment was found to decay with a time constant of 2.6±1.2 ps.

Radio-frequency compression of electron bunches applied to Ultrafast Electron Diffraction at kV energies

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Abstract

Ultrafast Electron Diffraction (UED) has evolved into a versatile tool for studies of structural dynamics in molecules and materials at sub-Angstrom spatial resolution. The time resolution obtainable with this approach has steadily improved since the “picosecond barrier”was broken in 2003. In fact, electron pulse durations of several hundred femtoseconds are available from state-of-the-art kV electron sources as long as the bunch charge is kept below approximately 2 fC. These are impressive advances, however it is important to note that time resolution below 100 fs is required for many experiments, and that an electron beam dose in the range of 1 - 1000 pC is needed for diffraction patterns of sufficient quality for most studies. This is a combination of requirements that cannot be currently realized due to the space-charge temporal broadening inherent to high charge density electron bunches. Thus, improvements in electron source performance are desirable for the further development of UED. In this work we will show how the introduction of a specially designed Radio- Frequency (RF) cavity into the UED beamline removes many of the technical limitations on the current generation of electron sources. For example, state-of-the-art particle tracking simulations show that it is possible to produce electron pulses below 100 fs that contain less than 1 pC of charge at the kV energies preferred for electron crystallography experiments. In addition, this approach allows for much greater control over the electron beam illumination conditions (at the specimen) than is possible with the current generation of sources. Finally, the fundamental limit to the performance of a UED diffractometer will be discussed. It will be shown that the space-charge temporal broadening of electron bunches is but a hurdle to overcome; that is, the true limit to performance results from the required transverse coherence length of the electron beam for a given experiment, and the initial brightness of the photoemission itself.

Ultra Fast Electron Sources A New Conclusion

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Abstract

According to our research most ultra fast electron sources waste much of the current they so painstakingly create, obtaining a brightness that does not match that of a continuous source.The reduced/normalised brightness (which scales as current over normalised emittance) B_r is a key source parameter, because apart from statistical interactions it is a conserved quantity. Also B_r defines the current I in an illuminated area A,

$$I = A\alpha 2\pi V B_r \tag{1}$$

where α is the half opening angle and V the potential.We examined existing and proposed sources, making a table of B_r , pulse length and energy spread (where pos- sible at source and sample). We concluded: (1) Accurate information about source design and performance is limited;(2) Surprisingly, despite modern mode-locked lasers, pulsed, experimentally proven, Br is much below continuous field emitters and Schottky(thermal field) emitters. We find photofield emission very promising, both [1] and [2] have claimed $B_r > 10^{10} A/(m^2 srV)$ but no proper, experimental evidence is given. For a Schottky emitter Van Veen showed that statistical coulomb forces decrease B_r as early as $10^8 A/(m^2 srV)$ [3]. The photofield emitter may do even worse [4].

We suggest chopping a high Br continuous source as an alternative for stroboscopic imaging. This could also be used for ultra fast ion microscopy, unleashing a whole new area of research.

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Building a Modular Compact/Radio-Frequency Ultrafast Electron Diffractometer: First Experiments in Compact Geometry

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Abstract

We will present our progress towards the development and implementation of a flexible new ultrafast electron diffractometer at 100-150kV energies. This diffractometer can be configured in both a compact geometry and expanded into a geometry that allows for the temporal compression of electron pulses using a RF cavity. In the compact geometry the electrons are allowed to freely expand via space-charge interactions so it is important to have the ability to place the electron source as close as possible to the sample. This configuration provides temporal resolution of approximately 800fs with 10^4 electrons per pulse. The RF configuration uses a synchronized RF cavity to temporally compress the electron pulses to below 100fs while allowing up to 6x10^6 electrons per pulse; this is an improvement of several orders of magnitude compared to the current state of the art.

We will report on initial experiments to characterize the diffractometer in compact geometry. These experiments include studies of the electron relaxation dynamics and lattice heating in thin film gold. The films are excited using approximately 50 femtosecond 400nm optical pump pulses below the damage threshold. The relatively slow heating dynamics of the gold thin film leads it to be an excellent initial experiment to characterize our system by comparing it to previously published results. Progress towards implementing RF pulse compression in this instrument will also be described.

A picosecond time-resolved X-ray scattering facility at BioCARS

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Abstract

BioCARS, a national user facility for time-resolved X-ray scattering studies at the Advanced Photon Source (APS), has recently completed commissioning of a focused pink-beam beamline for single-shot laser-pump/X-ray-probe measurements with a time resolution of $\sim$100 ps. Each x-ray pulse can contain up to $\sim$3 x 10^{10} photons, giving a time- averaged flux similar to that of fourth-generation free electron laser sources. A broadly tunable laser system provides a pulse width of $\sim$1 to 150 ps depending on its configuration and has an energy density of $\sim$5 mJ/mm^2 at the sample. Two in-line undulators with periods of 23 and 27 mm give continuous 6.8-20 keV first-harmonic coverage and can be combined for maximum flux at 12 keV. In combination with a high-heat-load shutter that reduces the average power load, a Kirkpatrick-Baez mirror system focuses the x-ray beam to a spot size of 90 μm (horizontal) by 20 μm (vertical). A high-speed Jülich shutter isolates radiation from individual 100-ps storage-ring bunches at a 1-kHz rate and is compatible with the most common storage ring fill patterns. This strategy allows almost full utilization of the entire run period at the APS. The facility will be described, along with some recent scientific results that highlight the unique features of the beamline. Additionally, a proposed experiment to use energy-chirped X-ray pulses at the Linac Coherent Light Source will be discussed.

To apply for beamtime or for more information about the BioCARS facility, visit http://biocars.org.

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Evaluation of lattice motion with vacuum-free compact designed time- resolved X-ray diffraction

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Abstract

Hard X-ray from femtosecond laser-produced plasma has gained much interest, as unique time resolved X-ray diffraction (TRXRD) experiments demonstrated and reveal ultrafast atomic dynamics of chemical reactions, phase transitions and coherent phonon vibrations.[1-3] Elucidating such ultrafast phenomena will lead to the fundamental understanding of energetic beam science and also further understanding of physical phenomena in the uncharted nanoscale extreme conditions. Recently, compact tabletop millijoule femtosecond lasers have been reported to be available for generating hard X-ray in vacuum with an intensity of about 1081010 cps/sr with the Kα X-ray conversion efficiency of 105106.[4-5] The experimental scale of a femtosecond laser could be reduced with a tabletop laser; however difficulties remain when using a huge and complex vacuum chamber system. We have constructed a compact designed and high intensity ultrafast pulsed Cu X-ray source in helium atmospheric pressure. A vacuum- free TRXRD system has also been constructed with this X-ray source. It is possible to reduce the overall size of X-ray source system without the complexity of a vacuum system.[6,7] It is also feasible to place the samples which are measured with TRXRD close to the X-ray source without vacuum system, enabling the use of the generated X-ray more efficiently.

We performed TRXRD on the 3 mJ/cm2 infrared femtosecond laser irradiated bulk sample of a CdTe single crystal with this vacuum-free compact designed TRXRD system. The CdTe is one of the suitable samples for TRXRD because the penetration depths of infrared light and Cu Kα X-ray into CdTe are almost the same degrees about 0.5 mum.[3] The integrated intensities of Kα X-ray diffraction lines from CdTe (111) were decrease by 5.6% in the time scale of $\sim$100 ps. The irradiation of infrared light at the intensity of 3 mJ/cm2 raises the temperature of CdTe by $\sim$50 K, and the thermal lattice vibration and expansion could occur. They would reduce the intensity of X-ray diffraction line by 56% due to the change of Debye-Waller factor. It takes $\sim$100 ps for the thermalized lattice in CdTe with acoustic velocity to expand $\sim$0.5 μm depth. Thus, the changes of the integrated intensity of X-ray diffraction line would be induced by thermal vibration and expansion of CdTe lattice. This vacuum-free compact designed TRXRD system would be a desirable tool for time-resolved atomic dynamics measurements.

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Ultrafast Time Resolved Electron Diffraction of Dynamics of Adsorbates on Silicon Surfaces

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Abstract

Dynamic processes of surfaces like electron excitation and relaxation, electron-phonon coupling, phase transition and phonon-phonon coupling take place on the femto- and picosecond timescale. Ultrafast time resolved electron diffraction is an excellent technique to study such processes on surfaces after excitation by a fs laser pulse. The laser energy will excite the electron system and heat the topmost atomic layers by electron-phonon coupling. In our experiment surface sensitivity is obtained by a RHEED (reflection high energy electron diffraction)-geometry [1]. In order to study the energy dissipation of an adsorbate systems after vibrational excitation we have performed time resolved measurements on the ($\sqrt{3}$x$\sqrt{3}$) Pb reconstruction on a Si(111) surface. ($\sqrt{3}$x$\sqrt{3}$) Pb reconstruction has a coverage of 4/3 monolayer and was prepared by deposition of Pb on Si(111) - (7x7) at 300 K followed by an annealing step to 500 K. After excitation of the Pb layer the heat transport into the silicon substrate is determined by studying the cooling process using the Debye Waller effect on the diffraction patterns taken at different delays between pumping laser pulse and probing electron pulse. The measured time constant of 150 ps can be explained by the huge difference in mass of Si and Pb atoms which prevents effective coupling of the Pb vibrational modes to the phonon bath in Si substrate. In order to study the dynamics of strongly driven phase transitions at surfaces far away from thermal equilibrium we performed time resolved experiments on the Peierls like phase transition from a (8x"2") to a (4x1) reconstruction of a Indium terminated Si(111) surface upon laser excitation at a sample temperature of 40 K [2]. The In-chains form 1-dimensional system currently being discussed whether the formation of a charge density wave (CDW) or the rearrangement of atoms in the In-chains is responsible for the formation of reconstruction. After excitation the (8x"2")-diffraction spots instantaneously disappears, while the intensity of the (4x1)-spots increases. This increase of the (4x1) spot intensity excludes an explanation by the Debye-Waller-Effect and is evidence for a true structural phase transition at a surface.

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Miniaturized RF Technology Towards a Novel Technique for Sub-Picosecond Electron Bunch Generation

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Abstract

Due to the extremely fast nature of interactions that occur on the microscopic scale, appropriate spatial and temporal resolution of these processes is desired. One way this is being pursued is through the use of electron bunches with a temporal length below picoseconds ($<10^{-12}$ s). To date, sub-picosecond electron bunches have been realized through the use of femtosecond lasers interacting with photocathodes.

We present a novel technique utilizing RF technology towards the creation of sub-picosecond electron bunches without the compulsory use of femtosecond laser systems. Utilizing RF technology and tricks developed as far back as the 1930s, we show through simulations and calculations that sub-picosecond electron bunches can be created with extremely low emittance growth to the electrons.

The design implements a 30 keV electron source from an SEM and highly underdamped electromagnetic standing wave cavities designed for high field strengths with low power consumption. The experimental setup is currently in the construction phase. Initial measurements are planned to progress shortly hereafter.

Coherent acoustic phonons in ultrathin monocrystalline Bismuth

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Abstract

Femtosecond electron diffraction (FED) is a very important technique to study structural dynamics of matter in ultrathin films. When a femtosecond laser pulse photoexcites a thin film it generates electronic and thermoelastic stresses which are finally released as acoustic waves. The propagation of those waves, being constrained by the film thickness, is responsible for the launching of coherent acoustic modes. The corresponding vibrational periods of those modes are in good agreement with that predicted by standing waves established by the boundary conditions[1]. FED is very sensitive, due to their very small de Broglie wavelength, to lattice displacements in transverse direction. When an ultrashort electron pulse probe the sample, reveals information about elastic properties of those films, including shear modes which are usually within the noise in all-optical studies[2]. Taking into account the speed of sound in solids ($\sim$ 5 km/s) and the thickness of the films (tens of nanometers), the period of those oscillations is typically in the order of few picoseconds. In this work we report on the generation and detection of coherent acoustic phonons in free-standing single crystalline Bismuth films.

A Compact Ultrafast Electron Diffractometer with MeV Electron Pulses Generated by RF photocathode

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Abstract

For determining transient structures in dynamical phenomena further and for developing electron microscope with fs-temporal resolution, we have constructed an ultrafast electron diffraction system of a transmission mode for a pulse including 10^7 electrons. A compact gigahertz rf (S band) photocathode, with an extremely small energy spread (ΔE/E $< 10^{-4}$) and emittance (<0.1mm mrad) was specially designed to make the entire diffraction system a laboratory-sized equipment [1]. Photoelectrons from Cu target were generated by the third harmonics of Ti:Sapphire laser, and accelerated by rf with a repetition rate of 10Hz. For a pulse with 10^6 electrons generated by 70-fs laser pulse and accelerated to 2MeV, the temporal width is estimated to be as short as 80 fs.

The system is designed to be especially rich in the electron beam configuration equipped with a condenser lens, an objective lens, and a projector lens, similar to a conventional transmission electron microscope. Therefore, both electron diffraction and imaging are possible. The illuminations with parallel/focused electron beam are easily switched, and the camera length is also adjustable. The sample chamber is at an ultra-high-vacuum ($\sim 10^{-9}$Pa) with several manipulation capabilities. Diffraction patterns can be recorded in two ways: one is real-time imaging with a sensitive CCD camera combined with an efficient scintillator for pump-probe experiments of reversible phenomena, and the other is for single shot experiments of non- reversible phenomena with extremely sensitive emulsion films used for high-energy physics experiments.

The photocathode was stable over hours, and the current density could be tuned precisely for various types of experiments. The current is in the range of 0.1$\sim$2pA, corresponding to $10^6 \sim 10^7$ electron/pulse that is sufficient for single shot experiments. Using the CCD based detection, high quality diffraction patterns were recorded from a thin film (70nm) of polycrystalline aluminium. Diffraction rings were clearly resolved up to 1.4Å^{-1} that is sufficient for further processing to obtain, for example, the radial distribution function. It seems that a sample with the thickness close to the penetration depth of the laser can be investigated. Diffraction patterns were recorded also from single crystal mica without obvious degradation in the pattern due to possible charge buildup. The capability of single-shot imaging is reported, and the challenges to the goal of fs-time resolved electron microscope are discussed.

In situ observations of amorphous Silicon and Germanium nanocrystallisation by Ultrafast Transmission Electron Microscopy

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Abstract

High quality structural information on the equilibrium states of most materials can be routinely obtained through several standard approaches. Detailed structural characterization of short-lived nonequilibrium states of materials, however, has proved very challenging since revealing the dynamics of structural transformation requires direct observations on the nanosecond to femtosecond timescale with spatial resolution of few nanometers.

The transmission electron microscope (TEM) is a powerful and versatile tool for the characterisation of materials, offering high spatial resolution (as low as 0.5Å); however, due to the poor temporal resolution of conventional TEMs it is rarely used for in situ direct imaging of structural transitions. In this work we will discuss recent developments in enhancing the temporal resolution of TEMs to produce a new class of Dynamic Transmission electron microscope (DTEM) at Lawrence Livermore National Lab. By improving TEM temporal resolution to the nanosecond timescale while preserving high spatial resolution studies of even irreversible structural transformations can be made.

We have used this new capability to study the crystallization dynamics of Amorphous Silicon (a-Si) and Germanium (a-Ge) specimens at a temporal resolution of 20 ns. Crystallization of these amorphous films has been induced by 532nm nanosecond laser pulses of variable fluence. Time-resolved TEM images have shown that the crystallisation process for a-Si begins at approximately 20ns and its duration is strongly influenced by the incident fluence of the laser beam. At low fluences the a-Si undergoes solid-state nanocrystallisation. At intermediate fluences a melt pool is generated and large radially oriented crystals eventually form. At high fluences the film was entirely melted and dewetting of the surface occurs with eventual crystallisation in large droplets onto the supporting SiO_2 membrane. Numerical modeling of heat conduction in the laser excited film was also performed and is in good agreement with the observed TEM images.

This work performed in part under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344 and supported in part by the US Department of Energy, Office of Basic Energy Sciences.

Four-Dimensional Visualization of Electron Dynamics by Attosecond Diffraction

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Abstract

We report here on the extension of ultrafast electron diffraction to the attosecond regime of charge densities in motion. Four-dimensional imaging of electronic structures and their changes by diffraction requires electron pulses with attosecond duration, in free space and at keV-range energies. We present two of our concepts, using synchronized microwave cavities or counter- propagating optical fields for electron pulse compression towards durations approaching ∼15 attoseconds [1-2]. Results on the roles of space charge and phase matching are presented. In contrast to attosecond photon pulses at around 100 eV [3], these attosecond electron pulses have by factors of 1000 shorter wavelengths and allow for diffraction with atomic-scale resolution [4]. Two potential applications are discussed for the example of molecular iodine: One involves measuring changes in bond order and the associated reshaping of the molecular charge density; the other regards attosecond charge oscillations in dielectrics and the buildup of the refractive index at optical frequencies [4]. We also present the results of quantum model simulations of the electron scattering process on an attosecond time scale and investigate the magnitude of radiation damage, the role of electron exchange interaction, and the influence of the molecular orbitals to diffraction [5]. These calculations support the possibility of using electron diffraction for imaging the structural motion of charge density in four dimensions, and also point out ways for exciting attosecond electron dynamics with keV-range electron pulses.

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Dynamic Transmission Electron Microscopic Investigation of Telluride Phase Change Materials

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Abstract

Using the technologically important phase change material $Ge_2Sb_2Te_5$ as an example, we show how a combination of single-shot real-space nanosecond transmission electron microscope imaging, time-resolved electron diffraction, and computation can reveal details of the interactions among geometry, optical absorption, and nucleation and growth kinetics in amorphous-crystalline transformations. We find the crystal nucleation density in this material to be exceedingly high (with many nuclei appearing per cubic m even after nanosecond-scale incubation times), such that large-scale molecular dynamics simulations are directly relevant for interpretation of the results. Grain growth and ensuing morphological changes happen much more slowly, on the scale of microseconds. We also show how principal component analysis of time-resolved diffraction data can provide a multi-dimensional picture of the evolution of various aspects of the transformation while suppressing noise and irrelevant information. Finally, we explore the interaction between geometry and laser absorption through the in situ study of nanostructured phase change materials coupled with multiphysics finite element simulations.

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

How to Extract Time-resolved Signal from Laue Diffraction by an Energy-Chirped Hard X-ray Pulse: a Proposal

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Abstract

A method to generate an energy-chirped hard X-ray pulse using a scheme based on overcompression of electron bunches is currently being developed at the LCLS. These energy- chirped pulses are expected to reach 1-1.5% bandwidth at 8 keV and a subpicosecond temporal width. We propose to use these chirped pulses to study light-initiated reactions in biological macromolecules like myoglobin and photoactive yellow protein at ultra-fast time-resolution in the time domain from 100 ps to 10 fs. One of the research and development areas required by this study is an effective numerical algorithm to extract time-resolved signal from Laue diffraction images produced by these chirped pulses. We will present a proposal of such algorithm and some preliminary data.

Single crystal Laue diffraction by a polychromatic X-ray beam is recorded as a pattern of spots on an area detector when the sample is stationary during exposure. Most spots arise from satisfying the Bragg condition at specific wavelengths within the bandwidth of the polychromatic source. A small fraction of the spots, known as multiples, are caused by satisfying the Bragg conditions simultaneously at two or more harmonic wavelengths, all represented within the source bandwidth. If the bandwidth is small enough, as it would be for the proposed energy chirp, there would be virtually no multiple spots. In an oversimplified statement, single crystal diffraction by such a narrow bandwidth, polychromatic source produces a pattern of spots, each of which can be traced back to a specific wavelength present in the source. If the source features an energy-chirped pulse, i.e. the arrival time at the crystal of each X-ray photon is highly correlated with its energy, each spot in a Laue pattern can be further mapped to its time of diffraction. This spot-to-time mapping suggests that Laue patterns produced by chirped pulses, although they do not appear to differ from those produced by conventional, unchirped pulses, are capable of recording time-dependent information with an intrinsic time-resolution substantially less than their pulse duration. A sufficient number of these Laue patterns may yield time- resolved data that is complete in diffraction space and span the entire desired time range.

The energy-angle correlation inherent in Braggs Law suggests an even more detailed mapping between each detector pixel and time. Each pixel associated with a Bragg spot has a known mean energy and spans a small energy range proportional to its linear dimension. In all previous analyses of Laue diffraction images, each spot spanned many (often $\sim$25) pixels, integration of diffraction intensities was carried out across all pixels and each spot was associated with a single X-ray energy. Here, our basic strategy is to analyze all Laue spots pixel by pixel without spot integration in order to take full advantage of the pixel-to-time mapping. This strategy requires joint modeling of the crystal mosaic structure and the spectral distribution of photon energy in each chirped pulse. The spectral distribution is anticipated to vary markedly from pulse to pulse, but both it and the mosaic structure are constant across the few hundred spots on each image. When these functions are jointly modeled, the remaining variation in pixel intensity across a spot arises from a combination of time-resolved signal, that is, the desired quantity synchronized in time from spot to spot, and experimental noise. This gives us the opportunity to apply singular value decomposition to extract the signal synchronized in the time domain.

Progress of mega-electron volt ultrafast electron diffraction at Tsinghua University

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Abstract

Time-resolved ultrafast electron diffraction (UED) is a promising tool to probe structural changes on the fundamental temporal and spatial scales of atomic motions. There have been recent efforts to employ mega-electron volt (MeV) electron beam from photocathode radio-frequency (RF) gun for UED application, mainly to achieve a better temporal resolution and eventually single-shot patterns with good signal-to- noise ratio. While, when using RF technology and MeV electron beam, several issues are worth careful consideration before applied for scientific experiments, e.g. the RF amplitude jitter, the RF-to-laser synchronization jitter, and how to detect MeV electrons with high enough efficiency. We optimized the configuration and parameters of a MeV UED system by start-to-end numerical simulation, and built and optimized such a prototype system at the Tsinghua Thomson scaterring X-ray source (TTX) facility. We obtained high-quality single-shot diffraction patterns of a 200 nm polycrystalline aluminum foil in which the first few rings are clearly distinguishable. We will also present considerations on improving several key components and discuss the futural plan.

Ultracold plasma electron source for imaging biological molecules

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Abstract

The molecular structure of biological molecules such as bacteriorhodopsin can be determined by electron diffraction, but general application of the technique has been limited by the brightness of conventional electron sources. Brightness is proportional to current and inversely proportional to temperature. Recent advances in atomic physics have made the prospect of high brightness electron beams from cold atomic clouds a promising alternative to conventional high temperature (104 K) sources [1,2]. Cold atoms in a magneto-optic trap (MOT) can be photoionized with a laser tuned just above threshold, releasing electron bunches with temperatures as low as 10 K. Although the number of electrons that can be extracted from a MOT is relatively small, the dramatic reduction in temperature may enable brightness that is competitive with conventional alternatives.

We created a MOT of 10^8 ^{85}Rb atoms, which were then ionized by two-step photoexcitation using the 780 nm MOT trapping beams and a 5 ns pulsed dye laser tuned near the ionization threshold (480 nm). The electrons were accelerated by an electrostatic field up to 200 V/cm between parallel accelerator plates, and electrostatically focussed using a third electrode. The electron bunches were detected using a microchannel plate, phosphor screen, and standard scientific CCD camera.

We are investigating the coherence and brightness of the extracted electron bunches, and in particular the effect of controlling the initial spatial distribution of the atoms to generate a uniform density elliptical charge distribution. Such elliptical bunches intrinsically preserve their brightness, and can for instance be refocused with conventional accelerator techniques [3].

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Ultra-fast dynamics of dimeric rhodium in a rigid ligand framework

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Abstract

[Rh_2 dimen$_4$](PF_6)$_2$ is a system with unusual bond shortening upon photo-excitation. In order to understand the switching behavior of this compound in more detail ultra- fast X-ray diffraction and transient absorption spectroscopy have been performed. The transient optical spectroscopy in the NIR regime suggests a coherent behavior on the femtosecond time scale, where as time resolved X-ray diffraction reveals mod- ulations of the integrated intensities of the observed Bragg reections on slower time scales (about 10 ps). However, no dynamics of the crystal lattice was induced with the excitation power used. The experimental data will be compared to theoretical calculations of the cation at TD-DFT level of theory.

The time resolved XANES and X-ray fluorescence high average power beam- lines at the Advanced Laser Light Source (ALLS) facility

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Abstract

We are investigating performances of ultrafast laser-based x-ray sources for dynamic imaging of various materials using time resolved X-ray spectroscopy [1,2]. We will present our effort in developing time resolved XANES and X-ray fluorescence beam lines at the ALLS facility at INRS with femtosecond and picosecond resolutions. A prototype beam line has been developed and coupled to the 100Hz laser system at ALLS [3]. This Ti:Sapphire CPA system is delivering 100mJ at 800nm with 100Hz repetition rate (giving 10W of average power) and 25 fs pulses (giving 4TW of peak power). Our most recent improvements include the control of the thermal loading of the beam line components at the 10W average power level in order to achieve high brightness and high stability x-ray source, and very high signal to noise ratio data collection. The source performances will be discussed and our preliminary experiments to follow the dynamics of photoexcited myoglobin will be presented. The ongoing effort to achieve sub-hundred femtosecond x-ray pulses with the 200TW/50W ALLS system (5J, 10Hz, 25fs) will be briefly sketched.

The ALLS facility has been funded by the Canadian Foundation for Innovation (CFI). This work is supported by NSERC, the Canada Research Chair Program and by Ministre de lducation du Qubec.

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Extreme phonon softening in laser-excited Bismuth towards an inverse Peierls-transition

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Abstract

Irradiation of a solid material with intense ultrashort laser pulses can lead to significant changes of the interatomic forces. Upon photoexcitation electrons are usually promoted from bonding states to less bonding or even anti-bonding states, thereby setting off atomic motion in the system. A prominent example is the so-called displacive excitation of coherent phonons (DECP) [1]. It has been found that DECP occurs only in materials with phonon modes of A_1-symmetry which do not lower the symmetry of the material, and that only A_1-modes are excited. The equilibrium structure of these materials can be derived by a Peierls-type transition from a state of higher symmetry.

Bismuth is a prominent example in which this type of coherent vibrational excitation has been studied in great detail. The majority of published results are based on time-resolved all-optical studies which cannot provide direct structural information. More recently time-resolved X-ray diffraction has also been used to directly follow the atomic motion associated with the laser- excited coherent phonon [2-4]. In particular the work performed at the Sub-Picosecond Pulse Source [3] has allowed, for the first time, to quantitatively measure the transient changes of the potential energy surface which underlie DECP and the softening of the phonon modes. In the present work we have used time-resolved X-ray diffraction to extend our studies of coherent optical phonons in laser-excited Bismuth to a higher fluence range that has not been studied previously. Femtosecond X-ray pulses at 8 keV (Cu Kα) from a laser-produced plasma served as probe pulses in an optical pump X-ray probe experiment. The transient changes of the (111)- and the (222)-diffraction peaks of a crystalline, 50 nm thick Bismuth film have been measured in a symmetric Bragg-configuration. For absorbed laser fluences above 2 mJ/cm^2 our experimental data reveal an extreme softening of the A_{1g}-mode down to frequencies of about 1 THz, only 1/3 of the unperturbed A_{1g}-frequency. The observed softening follows qualitatively the predictions of density functional calculations [5]. For even higher fluences (above 3 mJ/cm^2) the measured diffraction signals no longer exhibit an oscillatory behaviour. Our experimental observations present strong indication that upon intense laser-excitation the Peierls-transition which determines the equilibrium structure of Bismuth can be reversed and that the material is transformed into a transient ordered state of higher symmetry.

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Time-resolved crystallographic studies of heme proteins

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Abstract

The ultimate goal of time-resolved crystallographic studies of biological macromolecules is to visualize intermediate states along a reaction pathway at atomic resolution and at physiological temperatures, without trapping of the intermediates by chemical or physical methods. This is accomplished by taking X-ray snapshots of the molecule in the crystal as a reaction proceeds following the reaction initiation. The technique has reached a mature phase with demonstrated ability to detect small structural changes on ns and sub-ns time scale (1-6) and with important advances in the analysis of time-resolved crystallographic data, such as the use of Singular Value Decomposition (SVD) method to determine the structures of intermediates and elucidate the reaction mechanism (3-5). We present results of time-resolved crystallographic studies of heme proteins: structural relaxation processes in myoglobin and allosteric action in real time in a more complex, cooperative dimeric hemoglobin, as well as ligand migration pathways in both molecules. Myoglobin studies reveal sub-ns protein relaxation following ligand photo- dissociation and provide first direct experimental evidence of the main ligand exit pathway (7). Dimeric hemoglobin studies capture an early photoproduct intermediate and identify a possible trigger for a transition from the initial R-state to a tertiary T-like state that occurs on a s time- scale (8,9). These time-resolved experiments were conducted at the BioCARS beamline 14-ID at the Advanced Photon Source (USA).

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Direct Observation of Domain Behavior in a Multiferroic Structure Under Applied DC Bias

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Abstract

Select multiferroic materials exhibit a coupling between ferroelectric and magnetic order parameters, mediated by a quantum-mechanical exchange interaction. One of the most widely studied magneto-electric multiferroics is the perovskite $BiFeO_3$ (BFO). The magneto-electric coupling in BFO allows for control of the ferroelectric domain structures via applied electric fields. Recent advances in synthesis techniques have enabled the growth of high quality, epitaxial thin films. Because of these unique properties, BFO and other magneto-electric multiferroics constitute a promising class of materials for incorporation into devices such as high density ferroelectric and magnetoresistive memories, spin valves, and magnetic field sensors. Before BFO can be integrated into devices, an understanding of its ferroelectric and antiferromagnetic domain behavior across a range of time and length scales needs to be developed. Improved control of ferroelectric domain structures is critical for increasing the performance of ferroelectric and magnetoresistive memories, because memory switching speed and capacity are limited by domain wall mobility and domain size, respectively. We investigated the ferroelectric domains in BFO using transmission electron microscopy (TEM). Diffraction contrast was used to distinguish adjacent domains with different polarization directions, and high resolution images were analyzed to determine the atomic structure of domain walls. We present in situ TEM experiments designed to probe the response of BFO thin films to an applied DC bias, thereby enabling control of ferroelectric switching in the BFO thin film. Domain wall movement will be captured using digital streaming video at 30Hz, at both low and high magnifications. In our experiments, domain motion was studied at millisecond timescales; however, as industry aims to reduce device sizes, we look to ultrafast TEM to examine domain kinetics at timescales otherwise unattainable.

Ultrafast coherent imaging using UV-X harmonic beamline

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Abstract

X-ray lensless imaging extends standard X-ray diffraction towards imaging of individual nanosystems with unrivalled space and time resolutions. Up to now, this ability was limited to intense femtosecond coherent pulses from a free electron laser. High harmonics generation (HHG) sources would represent an excellent alternative since they are widely available and show the required properties. Moreover, HHG pulses are synchronized on sub-femtosecond time scale with the driving infrared femtosecond laser, allowing a vast flexibility in time resolved experiments. However, their brightness has so far restricted their application to static phenomena. In Saclay we developed a new harmonic source, based on a significant improvement of UV-X yield from HHG in gases, driven in enhanced phase-matching conditions. Using a long gas cell and a long focal length lens (5.5 m), together with a high quality UV-X optical line, allows reaching up to 2x10^9 photons per shot for the 25^{th} harmonic (=32nm) on the sample.

This high energy level allowed performing coherent imaging under several different configurations. We first realized a coherent diffracting imaging (CDI) experiment. The UV-X light is diffracted by a micrometer size sample. The diffraction pattern of the object exit wave is collected on a CCD camera in the far field regime. Since only the intensity of the diffracted coherent wave is measured, the phase information is missing and must be recovered. Image reconstruction with 60nm resolution was carried out using iterative phase retrieval techniques. To demonstrate the potential of our CDI beamline we then decreased the exposure times down to 20 femtoseconds. In the single shot acquisition regime, we achieved a 120nm resolution (Ravasio et al., PRL 103 028104 (2009)), which we recently lowered down to 80nm after optimization of the harmonic wavefront.

We then implemented a recently proposed holographic technique using extended references. This technique, easy to implement, allows a direct non iterative image reconstruction. In the single shot regime, we demonstrated a spatial resolution of 110nm.

This opens fascinating perspectives in imaging dynamical phenomena to be spread over a large scientific community. Investigation of ultrafast phase transitions in mesoscopic systems, ultrafast spin-reversals of magnetic nano-domains or large molecule rearrangements in biological environments are some examples.