

Workshop on New Opportunities in Ultrafast Science using X-rays

April 14-17, 2002, in Napa, CA

Abstracts of Poster Presentations

(Posters will be posted by number in Part III of the Chardonnay Room.)

SYNCHROTRON RADIATION EXPERIMENTS

1. **Bernhard W. Adams** - *APS/ANL* - (1) Picosecond laser pump, x-ray probe experiment on GaAs
2. **Bernhard W. Adams** - *APS/ANL* - (2) Coherent Control of Femtosecond X-Rays
3. **Yves Acremann** - *ETH Zurich / SSRL* - Ultrafast generation of magnetic fields in a Schottky diode
4. **Michael Becker** - *Brookhaven Nat'l Lab*
Towards Structure Determination of Membrane Proteins in 2-D Crystals Using Next-Generation Hard X-Ray Sources
5. **Christian Bressler** - *University of Lausanne* - Structural Dynamics With Current and Future X-Ray Sources
6. **Lin X. Chen** - *Argonne National Laboratory* - Photoexcited State Molecular Structures in Solution Studied by Pump-Probe XAFS
7. **Eric Collet** - *University of Rennes* - Ultra-fast light-induced structural order evidenced by time-resolved crystallography
8. **Steven L. Johnson** - *Lawrence Berkeley National Laboratory*
Properties of Liquid Silicon and Carbon Studied by Ultrafast X-Ray Absorption Spectroscopy
9. **Jorgen Larsson** - *Lund Institute of Technology* - Single-shot, time-resolved rocking curves using tilted optical wavefronts
10. **Aaron Lindenberg** - *Lawrence Berkeley National Laboratory*
Structural Dynamics in Semiconductor Nanocrystals probed by Time-resolved X-ray Diffraction
11. **J. Lüning** - *SSRL/SLAC* - Coherent Resonant Soft X-Ray Scattering from Magnetic Domain Patterns
12. **Andreas Scholl** - *Lawrence Berkeley National Laboratory*
A New Time Resolved Experiment for the Microscopic Study of Magnetization Dynamics Using X-PEEM
13. **Wolf Widdra and Bernd Winter** - *Max-Born-Institut*
Time-resolved core level photoemission: Surface photovoltage dynamics at the SiO₂/Si(100) interface

LASER PLASMA AND LASER HARMONIC EXPERIMENTS

14. **Davide Boschetto** - *LOA, Ensta-CNRS-Ecole Polytechnique* - Femtosecond X-ray diffraction applications to biology and solid-state physics
15. **Sylvain Fourmaux** - *LOA, Ensta-CNRS-Ecole Polytechnique* - Application of ultrafast x-ray diffraction to the study of condensed matter
16. **Yan Jiang** - *Brown University* - Ultrafast laboratory-based x-ray source and its application to molecular structure of solvated molecules
17. **J.C. Kieffer** - *INRS, University of Quebec* - The INRS ultrafast x-ray science program
18. **Yanwei Liu** - *UCBerkeley / Lawrence Berkeley National Lab* - Spatially Coherent Ultrafast Soft X-ray Generated by HHG in a Hollow Fiber
19. **Antoine Rousse** - *LOA* - Lab-top femtosecond x-ray sources and their applications in the study of femtosecond atomic processes
20. **Klaus Sokolowski-Tinten** - *Institut fuer Laser- und Plasmaphysik, Universitaet Essen*
Structural dynamics in laser-excited solids investigated by femtosecond time-resolved x-ray diffraction

TECHNIQUES

21. **Rafael Abela** - *Swiss Light Source* - Sub-picosecond Hard X-Ray Pulses from the SLS Storage Ring
22. **Roger Carr** - *Stanford Synchrotron Radiation Laboratory* - Measuring Pump-Probe Intervals with < 100 fsec Resolution for the SPPS
23. **Shaukat Khan** - *BESSY* - Thoughts on Femtosecond-Pulse Generation at BESSY II (2 KB)
24. **Andrew MacPhee** - *University of California, Berkeley* - Ultra-fast streak camera development
25. **Robert Schoenlein** - *Lawrence Berkeley National Laboratory* - Ultrafast X-ray Science at the Advanced Light Source

X-RAY SOURCES

26. **John Arthur** - *Stanford University, SSRL* - (1) The Linac Coherent Lights Source, an X-Ray FEL at SLAC
27. **James R. Boyce** - *Jefferson Lab* - FEL High Flux, Sub-picosecond, X-ray Source
28. **Joel D. Brock** - *Cornell University* - Cornell's Proposal for an Energy Recovery Synchrotron Source: applications to ultra-fast science using x-rays
29. **John Corlett** - *Lawrence Berkeley Nat'l Lab* - A Recirculating Linac Based Synchrotron Light Source for Ultrafast X-ray Science
30. **Jerry Hastings** - *Stanford University, SSRL* - The Sub-Picosecond Photon Source: An ultrafast x-ray experiment at SLAC
31. **V.N. Litvinenko** - *FEL Laboratory, Duke University* - Project for Generation of Femtosecond X-ray Beams from the Duke Storage Ring
32. **Elaine A. Seddon** - *CLRC Daresbury Laboratory* - 4GLS: The UK's fourth generation light source project at Daresbury Laboratory
33. **Paul Springer** - *LLNL* - Ultrafast Materials Probing at PLEIADES, a Subpicosecond Thomson X-ray Source at the LLNL Electron Linac
34. **Thomas Tschentscher** - *HASYLAB at DESY* - (1) VUV-FEL radiation at 100 nm: Radiation properties and first experimental results
35. **Thomas Tschentscher** - *HASYLAB at DESY* - (2) The route towards 0.1 nm FEL radiation at DESY

1. Picosecond laser pump, x-ray probe experiment on GaAs

B.W. Adams, D.A. Reis, M.F. DeCamp, E.M. Dufresne

Following a proposal for a femtosecond-resolving x-ray detector^[1], we measured the effect of femtosecond laser excitation of GaAs on the x-ray absorption cross section at the Ga K edge, using the Ga K alpha fluorescence yield as a measure of the x-ray absorption. An increase in the normalized Ga fluorescence of about 4 to 5 percent was observed with a persistence of about 200ps, which is consistent with the lifetime of laser-excited holes in the GaAs valence band. Further work could lead to an ultrafast x-ray detector^[1], and to a spectroscopic technique with an absolute reference energy level for electron dynamics in photoexcited semiconductors.

^[1]B. Adams, Proposal for a Femtosecond X-Ray Detector, Nucl. Instrum. and Meth. A 459, 339--346 (2001).

2. Coherent Control of Femtosecond X-Rays

B.W. Adams

The propagation of x-rays under the conditions of dynamical diffraction is very sensitive to changes in the crystal structure factor. This opens the possibility of coherent control of the propagating x-rays by inducing a rapid change of the structure factor with a laser, for example by massive excitation of coherent optical phonons. Just as a streak camera can reach a time resolution beyond typical RF frequencies, one may reach time constants which are less than the optical phonon frequencies. I have extended the Takagi-Taupin theory to explicitly include time-dependence and proposed several x-ray optical elements^[1,2] which are based on the theory. Examples are an all-optical 'streak camera' with few-femtoseconds time resolution and a femtosecond-switchable x-ray phase retarder.

^[1] B.W.A., conference proceedings no. 4500 of the SPIE; ^[2] B.W.A., Rev. Sci. Instrum, 3/02 in print.

3. Ultrafast generation of magnetic fields in a Schottky diode

Yves Acremann, *ETH Zurich / SSRL*

We introduce a novel scheme for the ultrafast generation of local magnetic fields in ferromagnet-semiconductor contact. The basis of our approach is to optically pump a Schottky diode with a focused, 150-fs laser pulse. The laser pulse generates a current across the semiconductor-metal junction, which in turn gives rise to an in-plane magnetic field. Specific advantages of this technique include the ability to rapidly create local fields along any in-plane direction on the sample without the need of electrical waveguides and microcoils.

4. Towards Structure Determination of Membrane Proteins in 2-D Crystals Using Next-Generation Hard X-Ray Sources

Michael Becker, *Biology Dept., Brookhaven National Laboratory, P.O. Box 5000, Upton, NY 11973*

It has been proposed that hard X-Ray Free Electron Lasers, such as the Linac Coherent Light Source (LCLS) planned for development at Stanford, may be useful for structure determination of membrane proteins in 2-D crystals^[1,2]. More recently, several proposals to develop Energy Recovery Linacs have been put forth. Both types of future X-Ray sources are expected to provide hard X-Rays pulses that are ultrashort and ultrabright; these characteristics are essential for high-resolution structure determination of 2-D samples. Such experiments may someday yield high-resolution structures, and, in conjunction with electron-crystallography experiments, detailed information on electrostatics. Appropriate sample preparation will likely be the main practical difficulty in realizing these aims. Application of pulsed X-Ray techniques to water-soluble proteins in 2-D crystals may also be fruitful.

^[1] Becker, M. (1999) *Biophysical Journal* 76, A121.

^[2] Becker, M. (1999) "Transparencies from the EMBO Workshop: Potential Future Applications in Structural Biology of an X-Ray Free Electron Laser at DESY", EMBL, Hamburg, pp. 184-198.

5. Structural Dynamics With Current and Future X-Ray Sources

Christian Bressler¹, Melanie Saes^{1,2}, Majed Chergui¹, Rafael Abela², Philip Pattison³

¹ Institut de Physique de la Matière Condensée, Université de Lausanne, CH-1015 Lausanne-Dorigny, Switzerland (E-mail: christian.bressler@ipmc.unil.ch)

² Swiss Light Source, Paul Scherrer Institut, CH-5232 Villigen-PSI, Switzerland

³ Institut de Cristallographie, Université de Lausanne, CH-1015 Lausanne-Dorigny, Switzerland

Following structural dynamics on the picosecond and femtosecond time scales is one of the frontier areas in Femtochemistry on condensed matter systems. With established structural tools like x-ray absorption spectroscopy (XAS) or diffraction methods it is possible to observe the structural changes during a photochemical reaction. Extending such studies in the long term to, e.g., biomolecular systems may provide unique insights in the nature of biological functions.

We present our current efforts to visualize the formation of photoexcited aqueous product species (e.g., short-lived radicals) exploiting hard x-radiation from a dedicated time-resolved x-ray beamline at the Advanced Light Source. Hereby we probe the temporal evolution via x-ray absorption spectroscopy methods like EXAFS. The results quantify the general feasibility, and model calculations are presented, which permit us to link the current results with more challenging experiments as well as to evaluate the utility of alternative sources of ultrashort x-rays.

6. Photoexcited State Molecular Structures In Solution Studied By Pump-Probe XAFS

Lin X. Chen, Guy Jennings, Tao Liu, *Chemistry Division, Argonne National Laboratory, Argonne, Illinois 60439*

Donald V. Scaltrito, Gerald J. Meyer, *Chemistry Department, Johns Hopkins University, Baltimore, Maryland 21218*

Knowing transient molecular structures during photochemical reactions is important for understanding fundamental aspects of solar energy conversion and storage. Fast x-ray techniques provide direct probes for these transient structures. Using x-ray pulses from the Advanced Photon Source at Argonne, the laser pulse pump, x-ray pulse probe XAFS (LPXP-XAFS) technique has been developed to capture transient molecular structures in disordered media with nanosecond time resolution. We have carried out pump-probe XAFS measurements on the MLCT state structure of Bis(2,9-dimethyl-1,10-phenanthroline) Copper(I) $[\text{Cu}(\text{I})(\text{dmp})_2]^+$ in toluene. The transient MLCT structure determined by x-ray absorption confirmed a whole charge transfer from the copper center to the ligand and the Cu coordination geometry change from tetrahedral to trigonal bipyramidal. The transient structural information combined with the kinetics data from transient optical absorption and emission spectroscopy established a new four-level energy diagram for the photophysics of $[\text{Cu}(\text{I})(\text{dmp})_2]^+$.

This work is supported by the Division of Chemical Sciences, Office of Basic Energy Sciences, U. S. Department of Energy, under contract W-31-109-Eng-38.

7. Ultra-fast light-induced structural order evidenced by time-resolved crystallography

E. Collet^a, M.H. Lemée-Cailleau^a, M. Buron^a, H. Cailleau^a, S. Techert^b, M. Wulff^c

^a GMCM CNRS-University of Rennes 1, 35042 Rennes, France, ^b Max-Planck Institute for Biophysical Chemistry, D-37070

Goettingen, Germany, ^c ESRF, 38043 Grenoble, France

An important and fascinating feature of some photo-active materials is the possibility of tuning their optical, magnetic, conduction or other physical properties by light stimuli. This arises by virtue of photo-induced co-operative phenomena, where the structural relaxation of the electronic excited state following the absorption of a photon is not localized on one atom or molecule but entails a drastic structural distortion involving several electrons and atoms or molecules, yielding a transformed nano-domain. When a large enough number of nano-domains is created at the same moment, the material switches between two different macroscopic states, and a photo-induced phase transformation towards a new lattice structure and electronic order. This opens the way to a new type of manipulation of matter by light, where for instance light can induce order, i.e. this differs from well-known light-induced disordering phenomena, such as surface melting or demagnetisation. This new type of photo-induced effects is exemplified by molecular charge-transfer materials that are readily tuned between competing neutral (N) and ionic (I) ground states. The photo-induced phase transformation in these materials, taking place on a ps time-scale, are highly co-operative and highly non-linear. The present birth of time-resolved photo-crystallography at synchrotron source gives a golden opportunity to directly observe the

photo-induced structural change and the symmetry breaking by recording “complete” diffraction pattern with a ps time resolution. Here we present this direct evidence of the N-to-I photo-induced structural phase transformation, i.e. after photo-irradiation of the high-symmetry N phase, stable at high temperature. The monochromatic X-ray diffraction data establish unambiguously the ps time-evolution of the intensity of some Bragg peaks and the appearance of a new Bragg peak characterising the ferroelectric order in the photo-induced I state.

8. Properties of Liquid Silicon and Carbon Studied by Ultrafast X-Ray Absorption Spectroscopy

Steven Johnson, *University of California, Berkeley*

Time-resolved absorption spectroscopy can be used to measure the electronic and structural properties of highly volatile states of matter created by the rapid heating of thin foils with ultrafast laser pulses. Experiments at ALS beamline 5.3.1 have used this technique to study the L-edges of liquid silicon and the K-edge of liquid carbon. Models of liquid silicon, using a combination of molecular dynamics simulations and the x-ray absorption code FEFF, agree reasonably with much of the experimental data. The data on liquid carbon suggest an sp bonding geometry, consistent with earlier work to simulate the "low-density" phase of liquid carbon.

9. Single-shot, time-resolved rocking curves using tilted optical wavefronts

O Synnergren (*Malmo University*), M. Harbst (*Lund Institute of Technology*), G. Katona (*Chalmers Institute of Technology*), R. Neutze (*Chalmers Institute of Technology*), R. Wouts (*Uppsala University*), **J. Larsson** (*Lund Institute of Technology*)

The aim of this investigation was to develop a method for time-resolved x-ray diffraction experiments taking advantage of the divergence of the laser-produced plasma source. This was implemented in an experiment where the temporal dependence of the x-ray reflectivity was recorded by hitting the sample with a tilted wave front from the laser. As a test case the well-known phenomenon of strain propagation in semi-conductors was used.

10 Structural Dynamics in Semiconductor Nanocrystals probed by Time-resolved X-ray Diffraction

Aaron Lindenberg, *UC Berkeley*

We present initial time-resolved x-ray diffraction measurements probing optically-induced structural transitions in semiconductor nanocrystals and nanorods in a liquid jet. Rapid changes in the (110) diffraction ring are observed following excitation.

11. Coherent Resonant Soft X-Ray Scattering from Magnetic Domain Patterns

J. Lüning, J. Stöhr, *SSRL, Stanford Linear Accelerator Center, Menlo Park CA 94025, USA*
S. Eisebitt, M. Lörger, W. Eberhardt, *BESSY GmbH, Albert-Einstein-Str. 15, 12489 Berlin, Germany*
A. Rahmim, T. Tiedje, *AMPEL, University of British Columbia, Vancouver B.C., V6T 1Z4 Canada*

In coherent x-ray scattering, a sample volume smaller than the coherence volume of the incident x-ray beam is illuminated. As a consequence, the scattering pattern contains a fine structure called "speckle", which is due to the fact that radiation scattered at different points in the sample can interfere. The speckle fine structure contains information about the individual spatial arrangement in the sample, which is beyond the average, statistical properties obtained in incoherent scattering experiments. Oversampling of the diffraction pattern has been mathematically shown to be a possible approach to overcome the phase problem, which has to be solved in order to extract the spatial information from the experimental data [1]. Recent experiments exploiting soft x-ray charge scattering from gold dots have demonstrated the viability of this approach in reconstructing the real space structure of submicron objects [2].

In this work, we are trying to lay the foundations in order to extend this concept to the investigation of magnetic domain patterns. Such patterns can be probed by resonant magnetic scattering. Circular and linear x-ray dichroism give rise to magnetization dependent contributions to the scattering cross-section and can be used as a contrast mechanism for coherent small angle scattering. We present first reconstruction results on worm domain structures in Co/Pt multilayers with perpendicular magnetic anisotropy.

The speckle imaging approach is of particular interest for future time resolved studies involving femto second x-ray pulses from a free electron laser. As a photon-in photon-out technique it has several advantages over electron based approaches, in particular, space charge restrictions are of no concern. An advantage over other photon imaging techniques is that no sophisticated optical components like zone plates are necessary to obtain nanometer spatial resolution.

[1] R. Barakat, G. Newsam, J. Math. Phys. 25, 3190 (1984)

[2] J. Miao, P. Chararlambous, J. Kirz, D. Sayre, Nature 400, 342 (1999)

12. A New Time Resolved Experiment for the Microscopic Study of Magnetization Dynamics Using X-PEEM

A. Scholl, S.-B. Choe, Y. Acremann, A. Lindenberg, C. Stamm, S. Andrews, H.-C. Siegmann, J. Stöhr, H.A. Padmore

Obtaining a microscopic understanding of the dynamics of magnetization reversal process is of principal importance for high-density magnetic data storage, e.g. on hard disk media and in novel magnetic random access memory devices (MRAM). Phenomena that appear on a ps to ns time scale are magnetization precession, magnetization damping, thermal fluctuations, spin waves and magneto-static modes. New schemes of ultra-fast manipulation of the magnetization can be devised based on the availability of fast pulses, e.g. utilizing the precession of the magnetization in an applied field pulse or pulsed injection of a spin polarized current into a ferromagnet. Study of these phenomena requires in addition to ps time resolution a very high spatial resolution and sensitivity because of the small size of the switching volume. X-ray PEEM offers high spatial resolution in addition to good chemical and magnetic sensitivity and is therefore well adapted to the investigation of magnetic phenomena on a nano-scale. We are currently designing a synchronized f-sec laser – synchrotron PEEM experiment, which will in its final version allow magnetization dynamics studies with 50 ps time and <5 nm spatial resolution. The test system that is currently set up at the Advanced Light Source has a more moderate spatial resolution of <100 nm. The setup includes a 125 MHz Ti:sapphire laser which is synchronized to the repetition rate of the Advanced Light Source. The laser is used to either stroboscopically excite the sample or to trigger a photoconductive switch driving a current through a coil or directly through the magnetic device. The evolution of the magnetization can then be followed as function of the delay between the laser pump and x-ray probe pulse which has a length of approximately 50 ps. Future f-sec x-ray sources could extend the time resolution of such an experiment into the sub-ps regime.

13. Time-resolved core level photoemission:

Surface photovoltage dynamics at the SiO₂/Si(100) interface

W. Widdra, D. Bröcker, T. Gießel, I.V. Hertel, D. Pop, R. Weber, and **B. Winter**
Max-Born-Institut, Max-Born-Str. 2A, D-12489 Berlin, Germany

We present a time-resolved surface photovoltage study with combined Laser synchrotron radiation (SR) in a pump-probe scheme which has been performed at the MBI undulator beamline at BESSY with synchronized Laser pulses from a specially designed Nd:YVO₄ laser system (both operating at 1.25 MHz). By sum frequency generation the width of the laser SR cross correlation could be determined to about 60 ps, the SR pulse width in the single bunch mode. Time-resolved Si 2p photoemission upon laser excitation for different SiO₂/Si(100) interfaces shows SPV shifts which decay on different time-scales. Future two-color two-photon photoemission (2C-2PPE) experiments will focus on the excited state dynamics in well-characterized OLED (organic light emitting diode materials) thin films, e.g. in adsorbed para-sexiphenyl, and the coupling to metal surfaces. The essentials of the experiment will be outlined.

14. Femtosecond X-ray diffraction applications to biology and solid-state physics

D. Boschetto^(a), C. Rischel^(b), I. Uschmann^(c), J. Etchepare^(a) and A. Rousse^(a)

^(a) Laboratoire d'Optique Appliquée, ENSTA/Ecole Polytechnique, UMR 7639 CNRS, INSERM U451, Chemin de la Hunière, 91761 Palaiseau, France

^(b) Niels Bohr Institute, Blegdamsvej 17, DK-2100 Copenhagen Ø, Denmark and Department of Mathematics and Physics, Royal Veterinary and Agricultural University, Thorvaldsensvej 40, DK-1871 Frederiksberg C, Denmark

^(c) X-ray Optics Group, Institute of Optics and Quantum Electronics, Friedrich Schiller University Jena, Max-Wien-Platz 1, 07743 Jena, Germany

X-ray diffraction temporally resolved on the femtosecond time scale is a strong instrument for studying ultrafast processes in all the fields of scientific interest, from biology to physics and chemistry. The development of the laser-plasma source for producing femtosecond X-ray flash has given rise to the first applications of this technique to follow dynamical processes^[1]. Here we report our last results on two parallel topics of our field of interest. First, we present an experimental result in which we realize for the first time a Laue diffraction pattern from a protein crystal (lysozyme) by using the femtosecond X-ray source. This represents a key step toward ultrafast dynamics study on biological samples. In fact, to resolve the dynamics of complex systems such as protein, many diffraction spots need to be detected at the same time. Standard Laue method consists in using a polychromatic and collimated beam, and the diffraction pattern is detected behind the sample. In our case, we use a different geometry, which takes into account the features of laser-plasma source: monochromatic and divergent. Because of the divergence, the reciprocal space is investigated by the different wave vectors available and therefore many different reflections are allowed for diffraction. Note that the interest on this geometry is amplified by the fact that new generation source of ultrafast X-ray pulse like the X-FEL is strongly monochromatic, and therefore divergence is needed to detect many diffraction peaks at the same time. The goal is of course to follow the full dynamics by detecting a Laue pattern for each time delay between the excitation pulse and the X-ray pulse. The second topic concerns solid-state physics application of femtosecond X-ray diffraction. Our goal is the detection of high frequency vibrational modes (optical phonons) in condensed materials. We show theoretical results of dynamical simulation of the rocking curve perturbed by optical phonon for some particular interesting materials. These simulations show some specific features that are a necessary guide to perform the real experiment.

^[1] C. Rischel et al., *Nature* **390**, 490 (1997).

15. Application of ultrafast X-ray diffraction to the study of condensed matter

S. Fourmaux⁺, I. Uschmann, C. Rischel and A. Rousse⁺

⁺Laboratoire d'Optique Appliquée-ENSTA, CNRS URA1406, Chemin de la Hunière, 91761, France.

Ultrafast melting is believed to arise from a strong modification of the inter-atomic forces due to laser induced promotion of a large fraction of the valence electrons to the conduction band. Following excitation of the electrons, the atoms find themselves far from the new equilibrium positions and immediately begin to move, gaining enough kinetic energy to produce sub-picosecond melting¹.

We used the technique of time-resolve X-ray diffraction with a plasma x-ray source produced from the interaction of a 10Hz, 120fs, 16mJ Ti:Sa laser beam focused on a silicium target². The emitted 7.12 keV X-ray radiation is expected to last 100fs, which can give a clear mapping of the atomic motions during phase transition. The X-ray radiation is collected by a toroidal quartz crystal and focussed onto Indium Antimony (InSb) semiconductor samples. We used a second laser pulse from this laser system to excite the sample at fluences ranging from 200mJ/cm² to 5mJ/cm², which is well below the damage threshold of the semiconductor. The depth and the time scale of non-thermal process have been fully characterized³. We also conducted experiments on thin film of Cadmium Telluride (CdTe) deposited on GaAs. The time scale of the transition is found to be similar to the behaviour of bulk InSb for the high exciting fluences, even within the first layers of the crystal.

¹ Shumay, I. L. and Hofer, U., "Phase transformations of an InSb surface induced by strong femtosecond laser pulses," *Phys. Rev. B*, **53**, 1996.

² Rischel, C. et al., "Femtosecond time-resolved X-ray diffraction from laser heated organic films," *Nature* **390**, 1997.

³ Rousse, A. et al., "Non-thermal melting in semiconductors measured at femtosecond resolution," *Nature* **410**, 2001.

16. Ultrafast laboratory-based x-ray source and its application to molecular structure of solvated molecules

Yan Jiang, *Brown University*

An x-ray system suitable for ultrafast x-ray absorption fine structure (XAFS) and x-ray diffraction measurements of solutions and crystals has been constructed in our lab. It consists of a Ti:sapphire laser system specifically designed for ultrafast x-ray pulse generation at 2 kHz. With this system, ultrafast x-ray pulses are produced from various solid and liquid metal targets. XAFS-spectra of transition metal complexes solvated in various solvents are measured and analyzed. We demonstrate that the simultaneous application of x-ray and optical spectroscopy provide correlated structural information.

17. The INRS ultrafast x-ray science program

P. Forget, L. Chen, J.C. Kieffer - INRS, *University of Quebec*

The new INRS laser system (10TW, 60fs, 10Hz) has been commissioned on solid targets at ultra-high intensities. We will present the most recent results obtained on the characterization of the ultrafast x-ray source obtained with the new laser system, and will discuss our application program.

18. Spatially Coherent Ultrafast Soft X-ray Generated by HHG in a Hollow Fiber

Yanwei Liu, David Attwood; *Center for X-ray Optics, Lawrence Berkeley National Laboratory, Berkeley, CA 94720*

Randy Bartels, Ariel Paul, Henry Kapteyn, Margaret Murnane, Sterling Backus; *JILA, University of Colorado, Boulder, CO 80309*

The spatial coherence of the radiation from a light source is critical for many of its potential applications. High harmonic generation (HHG) is a promising method for generating ultrafast and coherent radiation throughout the ultraviolet and extreme ultraviolet (EUV) to soft x-ray regions, due to its nature of coherent excitation by femtosecond laser. However, in previous experiments on HHG sources, the full coherence of the fundamental driving beam was not well retained. In this paper, we show that EUV radiation generated in a hollow core fiber has nearly perfect spatial coherence. This result took advantage of the selective phase-matching associated with the guided pump field inside the fiber waveguide, thus reduced the inherent limitation on spatial coherence of HHG due to the intensity- and space-dependent phase factor.

19. Lab-top femtosecond x-ray sources and their applications in the study of femtosecond atomic processes

Antoine Rousse, *Laboratoire d'Optique Appliquee, LOA-ENSTA, Chemin de la Huniere, 91761 Palaiseau cedex FRANCE, email: rousse@ensta.ensta.fr*

The femtosecond x-ray tube developed and used at LOA has allowed the first direct observation of ultrafast structural changes in the matter. This laser-produced plasma x-ray radiation is becoming widely used to probe the atomic dynamics at such ultimate timescale. Based on the experiments done at LOA, the capabilities of this existing source as well as the new generation of plasma x-ray source will be discussed.

20. Structural dynamics in laser-excited solids investigated by femtosecond time-resolved x-ray diffraction

Klaus Sokolowski-Tinten, *Institut fuer Laser- und Plasmaphysik, Universitaet Essen*

Ultrashort x-ray pulses offer a unique combination of atomic-scale spatial and temporal resolution, which permits direct measurements of structural transients on an ultrafast time scale. Using time-resolved X-ray diffraction with femtosecond, multi-keV X-ray pulses we have studied transient lattice dynamics in optically excited Germanium and and Bismuth. In Germanium non-thermal melting occurs within a few hundred femtoseconds followed by strong acoustic perturbations (shock waves) on a picosecond time-scale. In Bismuth excitation of large amplitude optical phonons is observed prior to the disordering of the material.

21. Sub-picosecond Hard X-Ray Pulses from the SLS Storage Ring

G. Ingold, A. Streun, B. Singh, **Rafael Abela**, P. Beaud, D. Grolimund, G. Knopp, L. Rivkin, V. Schlott, T. Schmidt, H. Sigg, J.F. van der Veen, A. Wrulich, (SLS) S. Khan (BESSY)

We propose to develop a facility for sub-picosecond hard X-ray pulses based on the electron-beam slicing method demonstrated at the ALS, Berkeley^[1]. The modulator and the radiator should be placed in one long straight section of the SLS storage ring, providing the smallest temporal stretching of the electron bunch slice and thus the shortest x-ray pulses (<100 fs). As a radiator, an in-vacuum undulator is foreseen with a period length of 17 mm, covering the photon range from 5 keV to 17 keV.

^[1] R. W. Schoenlein, S. Chattopadhyay, H.H Chong, T. E. Glover, P. A. Heimann, C. V. Shank, A. A. Zhotolents, M. S. Zolotarev, SCIENCE 287, (200), 2237

22. Measuring Pump-Probe Intervals with < 100 fsec Resolution for the SPPS

Roger Carr, *Stanford Synchrotron Radiation Laboratory*

Because of RF jitter, the interval between the Sub-Picosecond Photon Source x-ray probe and a visible laser pump pulse cannot be determined by a priori synchronization. It will be determined by a posteriori measurement, and data will be binned accordingly. We present here an accelerator-based technique to measure this interval using Compton backscattering of the probe laser off the electron beam, and conversion of time interval to position on a position sensitive detector. Depending on detector performance, single shot intervals of less than 100 fsec can be measured.

23. Thoughts on Femtosecond-Pulse Generation at BESSY II

Shaukat Khan, *BESSY (Berlin)*

User demand has triggered a new study of generating ultrashort synchrotron radiation pulses at the BESSY-II storage ring. The poster presents the status of this project.

24. Ultra-fast streak camera development

Andrew G. MacPhee, S. Johnson, A. Lindenberg, R.W. Falcone (*UC Berkeley*), P.A. Heimann (*LBNL*), R.W. Lee (*LLNL*), Z. Chang (*Kansas State University*)

Progress with the development of a ~0.1ps X-ray streak camera is described. A simulation of the image converter tube suggesting satisfactory imaging properties is presented, along with calculations of the field required to overcome resolution loss due to temporal dispersion.

25. Ultrafast X-ray Science at the Advanced Light Source

R.W. Schoenlein^a, A. Cavalleri^a, H.H.W. Chong^b, T.E. Glover^c, P.A. Heimann^c, H.A. Padmore^c, C.V. Shank^a, A.A. Zholents^d, and M.S. Zolotarev^d

^a Materials Sciences Division, Ernest Orlando Lawrence Berkeley National Laboratory

1 Cyclotron Rd. MS: 2-300, Berkeley CA 94720; phone: (510) 486-6557, email: rwschoenlein@lbl.gov

^b Applied Science and Technology Graduate Group, University of California Berkeley, Berkeley CA 94720

^c Advanced Light Source Division, Lawrence Berkeley National Laboratory

^d Accelerator and Fusion Research Division, Lawrence Berkeley National Laboratory

An important research frontier is the application of x-ray techniques such as diffraction and EXAFS to investigate structural dynamics (atomic motion and the making and breaking of chemical bonds) which drive phase transitions in solids, chemical reactions, and rapid biological processes. The fundamental time scale for such processes is an atomic vibrational period, ~100 fs, which is nearly three orders of magnitude beyond the present capabilities of synchrotrons. We have recently generated <150 fs synchrotron pulses from the Advanced Light Source using ultrashort laser pulses to manipulate the stored electron beam. We are presently developing a bend-magnet beamline with 100 fs time resolution for ultrafast x-ray science, and have proposed a femtosecond undulator beamline for the ALS. This poster will describe the techniques used in generating femtosecond x-rays from a synchrotron and will provide an overview of the research program in ultrafast x-ray science that is being developed at the ALS.

26. The Linac Coherent Light Source, an X-Ray FEL at SLAC

John Arthur and Jerry Hastings, Stanford Linear Accelerator Center, Stanford, CA 94309

The Linac Coherent Light Source (LCLS) program involves a collaboration of six US National Laboratories and universities with the goal of designing and building the first 4th-generation hard x-ray source, an x-ray free-electron laser (FEL). This FEL will utilize extremely short, intense, low-emittance electron pulses created by the high-energy linear accelerator at the Stanford Linear Accelerator Center. The FEL radiation produced will feature unprecedented peak brightness, short pulse length, and spatial coherence, tunable over an energy range of 0.8 - 8 keV. With favorable funding, major construction will begin in 2005 and the LCLS will begin operating in 2008. The LCLS facility will include two experiment halls, with room for several experimental stations, but only one experiment will be active at a time. Proposed experiments range from atomic physics through chemistry and biology to materials science.

27. FEL High Flux, Sub-picosecond, X-ray Source

James R. Boyce, Jefferson Lab (Abstract not available at time of printing.)

28. Cornell's Proposal for an Energy Recovery Synchrotron Source:

Applications to ultra-fast science using x-rays

Joel D. Brock - Cornell University (Abstract not available at time of printing.)

29. A Recirculating Linac Based Synchrotron Light Source for Ultrafast X-ray Science

J.N. Corlett, W. Barry, J. M. Byrd, S. DeSantis, P. Heimann, S. Lidia, D. Li, R. Rimmer, K. Robinson, R. Schoenlein, J.

Tanabe, S. Wang, W. Wan, R. Wells, and A. Zholents (LBNL, 1 Cyclotron Road, Berkeley, California 94720, USA)

We describe the design of a proposed source of ultra-fast synchrotron radiation x-ray pulses based on a recirculating superconducting linac. The source produces x-ray pulses with duration of <100 fs at a 10 kHz repetition rate, optimized for the study of ultra-fast dynamics. A high-brightness rf photocathode provides electron bunches which are manipulated to provide a large x/y emittance and small vertical emittance. An injector linac accelerates the beam to the 100 MeV range, and is

followed by four passes through a 600 MeV recirculating linac. Short x-ray pulses are obtained by a combination of electron pulse compression, transverse temporal correlation of the electrons, and x-ray pulse compression. We describe developments in key areas of accelerator technology including high rep-rate rf photocathode design, flat-beam production, collective effects, lattice design, magnets, deflecting cavities, x-ray beamline optics, and synchronization between experimental pump lasers and the x-ray pulse.

** This work was supported by the U.S. Department of Energy under Contract No. DE-AC03-76SF00098*

30. The Sub-Piconsecond Photon Source: An ultrafast x-ray experiment at SLAC

Jerry Hastings and John Arthur, *Stanford Linear Accelerator Center, Stanford, CA 94309*

The Sub-Piconsecond Photon Source (SPPS) experiment is a collaboration of laboratories and universities from around the world. The with the goal of the collaboration is to exploit subpicosecond x-ray pulses that can be generated utilizing extremely short, intense, low-emittance electron pulses from the high-energy linear accelerator at the Stanford Linear Accelerator Center. The x-rays produced will feature unprecedented peak fluxes and short pulse lengths tunable over an energy range of 8- 16 keV. Construction will begin in June 2002 and the SPPS will be operational until the start of LCLS construction stops operations of the FFTB. The SPPS experiment will provide the opportunity to address important scientific questions ranging from atomic physics through chemistry and biology to materials science. The design study for the SPPS is available, SLAC-Pub-8950.

31. Project for Generation of Femtosecond X-ray Beams from the Duke Storage Ring

V.N.Litvinenko, O.A.Shevchenko, S.F.Mikhailov, Y. Wu - *FEL Laboratory, Duke University, P.O. Box 90319, Durham, NC 27708, U.S.A*

In this paper we describe the project for developing an intense X-ray source at Duke storage ring. The project is based on a mm-wave FEL, which will be used for creating and maintaining electron bunches with sub-picosecond (femtosecond) time structure and for generating hard X-ray beams with circular polarization via Compton intracavity backscattering. A strong coherent synchrotron radiation (CSR) is the main limiting factor for proposed mode of operation. We present the results of theoretical and numerical studies of instabilities caused by CSR and the attainable parameters for femtosecond electron bunches in the Duke storage ring. Based on these studies, we present the design and predicted performance of the 1.5-75 keV X-ray pulsed source with of 10-fsec RMS duration and 105 photons per pulse. The rep-rate of the source can be tuned from few Hz to tens of MHz. (Work is supported by the Dean of Natural Sciences, Duke University.)

32. 4GLS: The UK's fourth generation light source project at Daresbury Laboratory

Elaine A. Seddon, *CLRC Daresbury Laboratory*

4GLS is a suite of accelerator-based light sources planned to provide the UK with state-of-the-art radiation in the low energy photon regime. Superconducting energy recovery linac (ERL) technology will be utilised in combination with a variety of free electron lasers (IR to XUV), undulators and bending magnets. The 4GLS undulators will be optimised to generate spontaneous high flux, high brightness radiation, of variable polarisation, from 3-100 eV. However, they will also generate usable radiation (in the higher harmonics) up to around 500 eV. The ERL technology of 4GLS will allow shorter bunches and higher peak photon fluxes than possible on storage ring sources. It will also give users the added bonuses of pulse structure flexibility and effectively an infinite beam lifetime; benefits unavailable to storage ring users. The UV free electron lasers will be used to generate short pulses (in the fs regime) of extreme ultraviolet light that is broadly tuneable and more than a million times more intense than the equivalent spontaneous undulator radiation. A strong feature of the scientific programme planned for 4GLS is dynamics experiments.

33. Ultrafast Materials Probing at PLEIADES, a Subpicosecond Thomson X-ray Source at the LLNL Electron Linac

Paul T. Springer¹, Greg P. Le Sage¹, C.P.J. Barty¹, James Rosenzweig², Fred H. Streitz¹, Winthrop Brown¹, John Crane¹, Rick Cross¹, Todd Ditmire⁴, David Fittinghoff¹, David Gibson³, Fred Hartemann¹, Dennis Slaughter¹, Aaron Tremaine¹, Scott Anderson², Jaroslav Kuba¹, John Moriarty¹, Andy McMahan¹, Hyunhae Cynn¹, Choong-shik Yoo¹

¹ Lawrence Livermore National Laboratory, 7000 East Ave., Livermore, CA. 94550

² UCLA Department of Physics and Astronomy, 405 Hilgard Ave, Los Angeles, CA. 90095

³ UCD Department of Applied Science, 661 Hertz Hall, Livermore, CA, 94550

⁴ UT Department of Physics, Robert Lee Moore Hall, Dean Keeton and Speedway, Austin, Texas 78712

The use of ultrafast laser pulses to generate very high brightness, ultrashort (10^{-14} to 10^{-12} s) pulses of x-rays is a topic of great interest to the x-ray user community. In principle, femtosecond-scale pump-probe experiments can be used to temporally resolve structural dynamics of materials on the time scale of atomic motion. However, further development of this field is severely hindered by the absence of a suitably intense x-ray source that would drive the development of improved experimental techniques and establish a broader range of applicability. We report on a project at the Lawrence Livermore National Laboratory to produce a novel x-ray source and essential experimental techniques that will enable unprecedented dynamic measurements in matter. Based on scattering of a sub-50-fs, multiterawatt, multibeam laser from a co-synchronous and highly focused relativistic electron bunch, PLEIADES (Picosecond Laser Electron Interaction for Dynamic Evaluation of Structures) will produce tunable, ultrafast, hard x-ray (10- 200 keV) probes that greatly exceed existing 3rd generation synchrotron sources in speed (100 fs - 1 ps), peak brightness (10^{20} ph/mm²s mrad² 0.1% BW, and $>10^9$ ph/pulse), and simplicity (100-fold smaller). Such bright, ultrafast high energy x-rays will enable pump-probe experiments using radiography, dynamic diffraction, and spectroscopy to address the Equation of State and dynamics of phase transitions and structure in laser heated and compressed heavy dense metals of interest for materials science.

This work was performed under the auspices of the U.S. Department of Energy by the University of California, Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

34. VUV-FEL radiation at 100 nm: Radiation properties and first experimental results

Thomas Tschentscher and The TESLA test facility collaboration

The vacuum-ultraviolet free-electron laser at the TESLA test facility (TTF) has reached saturation in a wavelength range around 100 nm. Several methods of characterisation have been employed to analyse the radiation properties of the self-amplified spontaneous emission. A pulse duration of 30 to 100 fs and peak brilliance of 2×10^{28} phts/(s•mm²•mrad²•0.1%) were determined. First experiments have employed the FEL radiation in investigations of damage of optical material and in investigation of interaction of FEL radiation with cluster beams.

35. The route towards 0.1 nm FEL radiation at DESY

Thomas Tschentscher, HASYLAB/DESY

DESY at present extends the existing TESLA test facility in order to provide short-wavelength FEL radiation for first user-experiments in the wavelength range 40 to 6 nm. Using a single undulator a beam distribution system will serve five to six experiments. User-operation of TTF is scheduled to resume in 2004. The proposal for an x-ray free-electron laser laboratory for 0.1 to 6 nm wavelength (200 eV - 14 keV) is evaluated at present and could start operation around 2010. The FEL radiation will be transverse coherent, have a duration of ~ 100 fs and reach a peak brilliance of 1033 phts/(s•mm²•mrad²•0.1%) with 1012-1013 photons per pulse. Five SASE undulators and five undulators for spontaneous radiation will serve up to 30 experimental stations. Flexible modes of operation and dedicated instrumentation will allow to operate several user-experiments at the same time.