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University of Michigan

Ultrafast X-Ray Coherent Control

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5 Executive Summary

This main purpose of this grant was to develop the nascent field of ultrafast x-ray science using accelerator-based sources, and originally developed from an idea that a laser could modulate the diffracting properties of a x-ray diffracting crystal on a fast enough time scale to switch out in time a shorter slice from the already short x-ray pulses from a synchrotron. The research was carried out primarily at the Advanced Photon Source (APS) sector 7 at Argonne National Laboratory and the Sub-Picosecond Pulse Source (SPPS) at SLAC; in anticipation of the Linac Coherent Light Source (LCLS) x-ray free electron laser that became operational in 2009 at SLAC (all National User Facilities operated by BES). The research centered on the generation, control and measurement of atomic-scale dynamics in atomic, molecular optical and condensed matter systems with temporal and spatial resolution . It helped develop the ultrafast physics, techniques and scientific case for using the unprecedented characteristics of the LCLS. The project has been very successful with results have been disseminated widely and in top journals, have been well cited in the field, and have laid the foundation for many experiments being performed on the LCLS, the world's first hard x-ray free electron laser.

6 Goals and Objectives.

The main goals and objectives of the grant as articulated in the original proposal and its three subsequent renewals were broadly geared towards advancing the nascent field of ultrafast x-ray science on accelerator based sources, looking forward toward the eventual construction and operation of the LCLS. In particular, our proposed objectives were to perform fundamental investigations in the following areas:

- Ultrafast X-ray Switches
- High Amplitude Coherent Optical Phonons
- Electro-optic detection of ultrafast electrons at SPPS
- Measurement and Control of the Interatomic Potential
- Strong field alignment of molecules
- X-ray scattering from aligned molecules

As described below in the summary of project activities, the PIs have made important advancements in all these areas. It is also documented in yearly progress reports of areas is also documented in Principal Investigators' meetings of the Atomic Molecular and Optical Sciences program of the Fundamental Interactions team within the Chemical Sciences, Geosciences, & Biosciences Division of BES. The reports of which can be found at:

http://science.energy.gov/bes/csgb/principal-investigators-meetings/.

7 Summary of Project Activities

The project activities were carried out at the Advanced Photon Source sector 7, with the MHATT-XOR collaboration (previously MHATT-CAT); and at the Stanford Linear Accelerator Center (now SLAC National Accelerator Laboratory), with the SPPS collaboration and the Stanford PULSE Center. The research concentrated on ultrafast and coherent dynamical processes in solids and molecular systems. Along with new science, we developed new ultrafast methodology that is currently being utilized one next generation ultrafast x-ray sources such as LCLS.

Ultrafast science probes dynamics on the atomic and molecular scale of bond vibration in a molecule or optical phonons in a solid. This is typically on the order of 100 fsec, but can in some cases be as long a few picoseconds. X rays are ideal probes of motion on these scales because of their short wavelength, and there has been much progress towards producing ultrafast x-ray pulses for transient dynamical studies. This played a large part in the motivation for the LCLS free electron laser, which became operational shortly after the end of this project and provides coherent hard x-ray pulses of a few femtosecond duration with peak brightness more than a billion times of its predecessors.

Early efforts in the field concentrated on laser-generated plasmas, which can produce x-ray bursts as short as a few hundred femtoseconds. These sources are quite weak, however, and this has limited their use to a few very high contrast problems, such as crystalline melting. Electron beam-based x-ray sources can be much brighter. The challenge therefore has been to make these accelerator-based x-ray pulses short, so that they can record more than the timeaveraged motion of transiently excited materials.

Third generation synchrotron pulses, which are 30-100 psec in duration , are tantalizingly close to the required duration, and so we and others have been working with them to learn about dynamics of transient molecular and solid state processes. Techniques that we and others have developed include picosecond x-ray streak cameras; x-ray switches based on x-ray transmission through or reflection from transiently excited crystals; and methods to shorten the electron bunches in the synchrotron.

7.1 Characterization of Ultrafast Strain Generation

Our work on ultrafast x-ray science began in 2001 with studies of methods for an ultrafast laser-induced Bragg switch. These first experiments developed and used the first femtosecond laser synchronized to the x rays at APS, on the sector 7 insertion device (ID) beamline. The Bragg scattering was modified by a coherent acoustic wavepacket (coherent strain) generated in impulsively laser-excited InSb. Uniaxial longitudinal strains on order of a percent, corresponding to pressure of many Mbars, and response times corresponding to modulation frequencies up to tens of GHz were not uncommon. False color images of

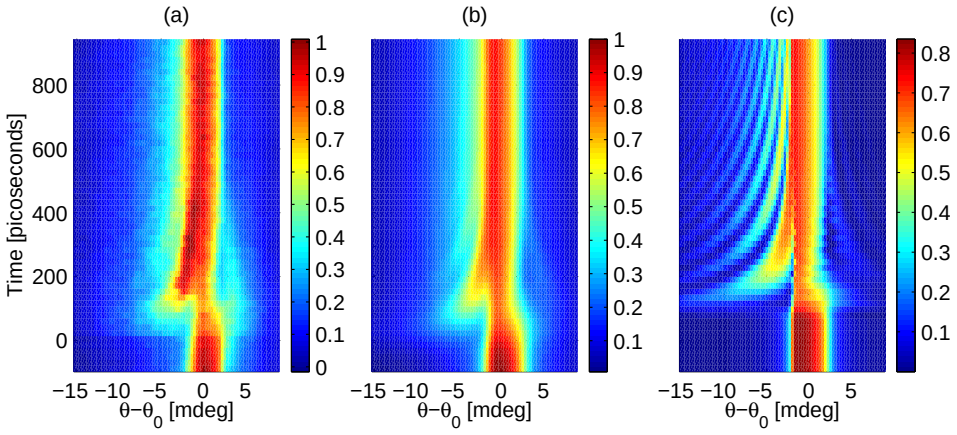

Figure 1: Time-resolved rocking curves: 10 keV x rays, impulsively strained InSb, 111 reflection. (a) Data and (b) dynamical diffraction simulation for a 0.51% peak strain with a 100 nm laser absorption depth and a 100 ps, 1.25 mdeg FWHM Gaussian resolution functions; and (c) dynamical diffraction simulation for the reflectivity of a monochromatic x-ray pulse with ideal temporal resolution.

the time-resolved diffraction pattern for laser excited InSb as a function of angle and time-delay are given in Fig 1 [30]. The relatively large flux at the APS made it possible to study the initial excitation and subsequent motion of the lattice in much greater detail than ever before, and we made quantitative comparisons to the existing thermoelastic model which separated the transient strain into surface heating (thermal expansion) and coherent acoustic components. We found that the model calculations significantly underestimate the coherent acoustic phonon contribution to the overall strain [30].

7.2 Coherent Control of Pulsed X rays

A number of dramatic experiments on x-ray modulation and x-ray switching were conducted in 2002-2004, in transmission (i.e. Laue) geometries in crystalline Ge. Figure 2 demonstrates the use of impulsively generated coherent acoustic phonons to control x-ray diffraction in a thick crystal by modulating the anomalous transmission (Borrmann Effect)of x rays propagating many x-ray incoherent absorption depths through the material. We proposed this mechanism to modulate x-ray pulses on an ultrafast time-scale. This switches x-ray energy between two separated x-ray beams (the forward-diffracted and deflected-diffracted beams) faster than the synchrotron pulse duration.

This effect is coherent and low-loss. It is the temporal analog of the Pendellösung effect: The ultrafast acoustic pulse acts like a thin interface layer dividing the crystal into two regions, whose relative sizes change at the speed of sound. By simply adjusting the delay between the x rays and the acoustic pulse one can coherently control the exit beams

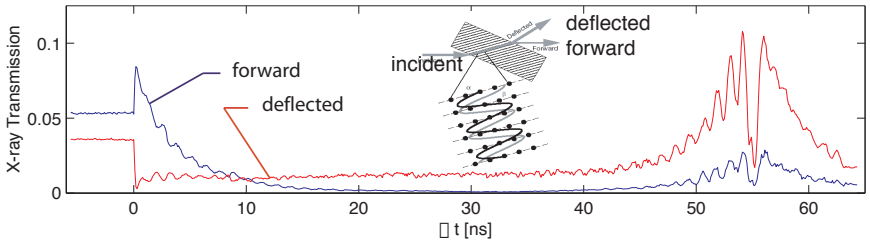

Figure 2: X-ray control through diffraction from the asymmetric $20\bar{2}$ planes of a thick Ge [001] crystal following laser impulsive excitation from [28].

to preferentially switch energy from one beam to another. Different arrangements were studied, including a switch made from two counter-propagating acoustic pulses that collide deep inside the crystal. This doubles the acoustic modulation frequency, and also provides one means to observe a localized transient strain inside an otherwise opaque material [28].

7.3 Supersonic Strain Front Propagation

The initial coherent transient x-ray switch in this Borrmann geometry is considerably faster than half of a Pendellösung cycle, and measurements with an x-ray streak camera revealed that the strain travels about 8 times faster than sound when it first begins (see figure 3). More detailed experimental results were compared with dynamical diffraction simulations, showing that the strain generation process is dominated by rapid ambipolar diffusion of the dense electron-hole plasma and the subsequent coupling to the lattice through the deformation potential. As the transport of carriers is diffusive, the region of stress can initially outrun the strain wave, leading to a strain *front* that propagates faster than the speed of sound[25].

7.4 Optical phonon switches

Faster switching times can be achieved if we utilized high amplitude optical phonon distortions in x-ray scattering. We carried out optical studies of high-energy ultrafast laser-induced large amplitude coherent motion of crystalline bismuth through impulsive stimulated Raman scattering. These large lattice distortions could then modulate x rays on an ultrafast time scale. As early as 2001 we measured optical reflectivity changes larger than 1%, and showed that this corresponds to lattice deformations of $0.1\mathring{A}$. As the amplitude of the optical phonon increases, a frequency shift was also observed [29]. This led directly to later studies of ultrafast x-ray modulation using the new ultrafast x-ray source, SPPS at SLAC, and later experiments on LCLS.

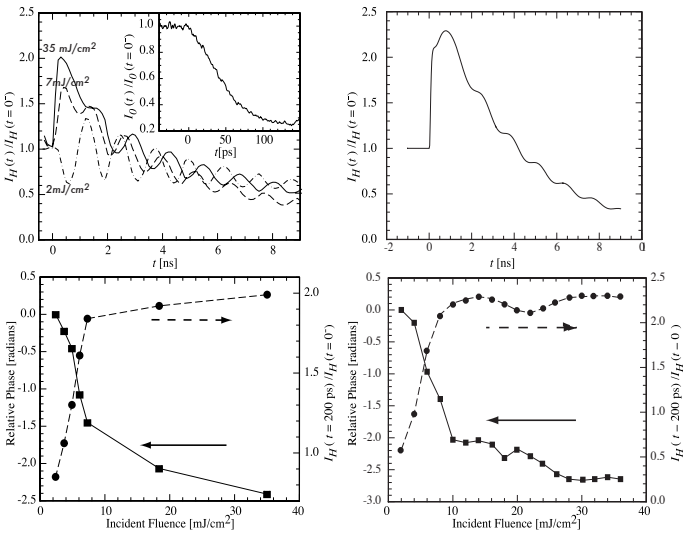

Figure 3: Clockwise from upper left: (a) Pendellösung oscillations for the asymmetric $\bar{2}02$ planes (001) surface in the forward-diffracted beam for laser excited Ge at different fluences of 35, 7 and 2 mJ/cm^2. Inset streak camera data of deflected diffracted beam at a fluence of 35 mJ/cm^2. (b) Calculated Pendellösung for 35 mJ/cm^2. (c) Amplitude of transient and phase of Pendellösung as a function of incident fluence and (d) Calculated transient and phase.

7.5 Single Shot Timing Measurements at SPPS

SPPS (the sub-picosecond pulse source) was an experiment at SLAC in 2003-2006 to produce and use sub-100 fs hard x rays. The SLAC linac produced ultra-short high charge (3 nC) electron bunches that passed through a 3 m undulator to produce 10^7 1.5 Angstrom x rays at 10 Hz. The electron pulse widths, and therefore the x-ray pulse widths, were on the order of 80 fs FWHM.

We participated in both experiments and in source characterization at SPPS. Our first task was to find and demonstrate a solution to the problem of synchronization. X rays produced by electrons in a linear accelerator are not naturally synchronized to a laser, so sophisticated methods are required to deliver timing signals with sub-picosecond precision. We implemented electro-optic sampling of the coherent THz radiation produced by the relativistic electrons to establish the relative arrival time of the x rays and the laser. We designed and implemented a vacuum chamber just upstream from the undulator, containing a thin ZnTe crystal placed only 1cm from the electron beam. Part of the light from the laser system is split off *before amplification*, shaped to pre-compensate for dispersion during propagation, and sent to the EO apparatus through 150m of single-mode, polarization-preserving fiber [20]. Current x-ray free electron lasers in the U.S. and Japan make use

of similar methods for single-shot timing and synchronization based on x-ray and optical cross-correlations.

Also as part of the SPPS collaboration, we led the effort that showed that synchronization can also be obtained by taking advantage of the large scale changes in structure that occur during laser melting of crystalline materials. The change in x-ray scattering is so dramatic that merely a single x-ray pulse is sufficient to record the dynamics. We studied the transient disorder in laser-heated crystalline InSb excited by a short pulse laser. The laser and the x rays impinged the sample at different angles, so that the relative arrive time varied across the irradiated spot, as shown in Fig. 4. Data recorded on each laser shot showed how the arrival time varied, sometimes by more than a picosecond. Using this as an independent measurement of the x-ray arrival time, we were able to show that we could correlate the EOS to the arrival time of the x rays to within 60fs, rms [19] (Fig. 5).

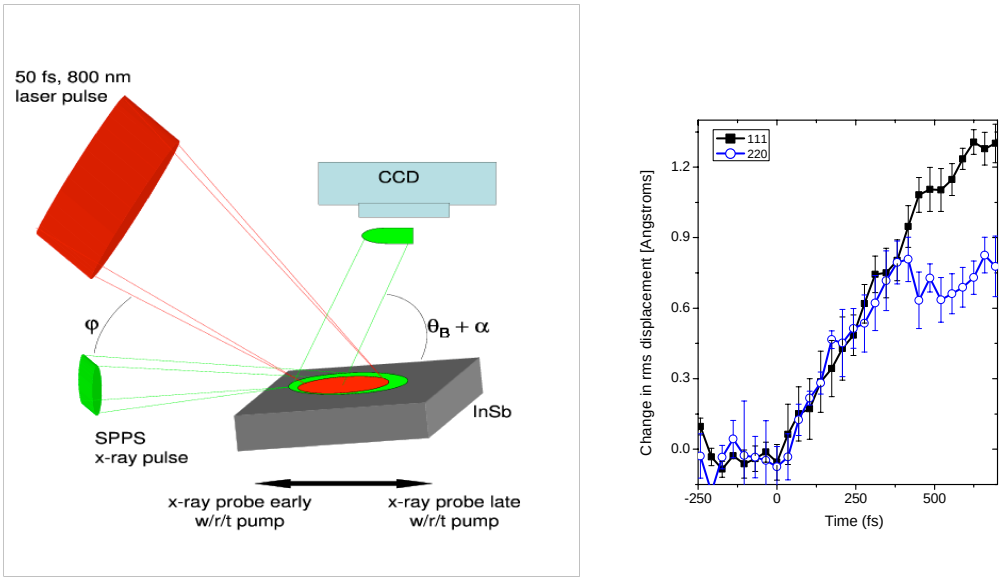

Figure 4: Change in the rms displacementof InSb, extracted from the time-dependent x-ray diffracted intensity. The slope at short times corresponds to the room-temperature rms atomic velocity, [17]

7.6 Structural phase transitions in InSb

The InSb melting experiments were the highest time resolution structural study of melting to date, and it revealed new physics concerning the nature of laser-induced phase transitions in this material. In the first hundreds of femtoseconds following excitation, the x-ray diffraction efficiency decreases at a rate that appears to be limited only by the ballistic

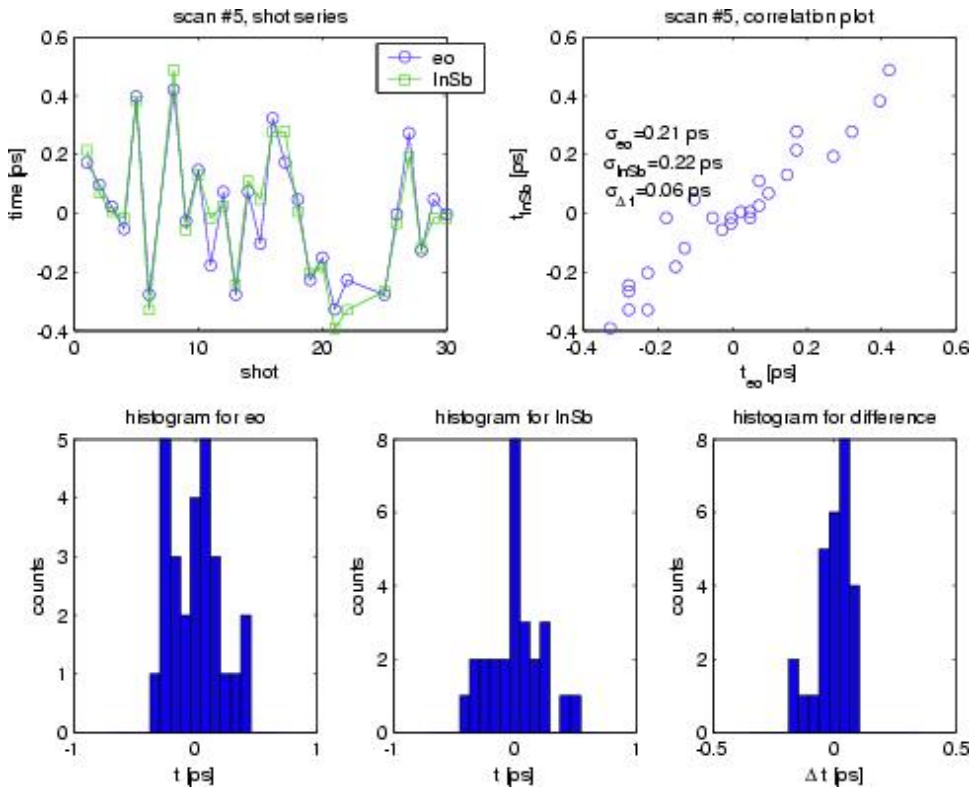

Figure 5: Correlation between the relative timing derived by EO sampling, and by diffraction during laser-induced melting of InSb at SPPS

motion of atoms away from their lattice sites. This was shown in a series of experiments in an SPPS collaboration with A. Lindenberg and K. Gaffney for each of the two x-ray diffraction planes studied. These results are consistent with an isotropic flattening of the interatomic potentials, such that the atoms are "free" to move about essentially with the same momentum set by their thermal motion before the laser excitation [17]. On a longer time scale (ps), the atoms begin to collide and the motion transitions from ballistic to diffusive behavior. In this case, however, the disordering occurs faster along the initial tetrahedral bonding directions [15]. On this time scale, the material is still solid in the sense that long range order has not been destroyed, and the liquid has not reached equilibrium.

7.7 Coherent optical phonons observed in Bi at SPPS

The EO determination of synchronization of each x-ray pulse was used to study the excitation and decay of coherent phonons in crystalline Bi. The resulting time resolution

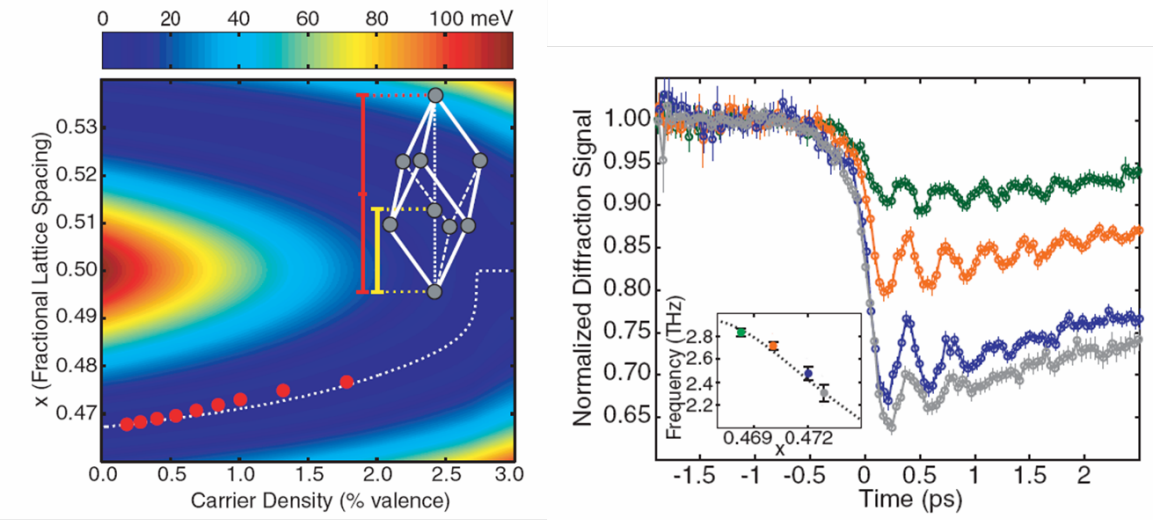

Figure 6: Left. Calculated excited state potential of bismuth with the experimentally determined equilibrium positions overlayed. Right. x-ray diffraction efficiency as a function of time delay between the optical excitation pulse and x-ray probe for excitation fluences of 0.7 (green), 1.2 (red), 1.7 (blue), and 2.3 mJ/cm2 (gray). The inset displays the optical phonon frequency vs. atomic equilibrium position along the body diagonal of the unit cell as measured by x-ray diffraction [8]. Dotted line is the prediction of [16]

was sufficient to reveal new details about the inter-atomic potential energy surface of this material [8]. In the Bi experiment, an ultrafast laser pulse was used to promote a large fraction of the valence electrons into the conduction band of a thin bismuth film, during a few tens of femtoseconds. The presence of such a dense electron-hole plasma significantly alters the interatomic potential before the atoms have a chance to move. Following this excitation, the atoms move in very large amplitude coherent motion which was measured via x-ray diffraction. From this, we were able to follow the equilibrium position and curvature of the double-well potenital as function of carrier density and time over the first few picoseconds of excitation (see Fig 6). These are the two key parameters that determine the Peierls distorted structure. The experiments were in excellent agreement with our previous optical experiments and density functional theory [16], which indicates that we are very close to a solid-solid phase transition.

7.8 Diffuse scattering and short pulse experiments at the APS

One of the main goals of our experimental progress at APS was to control x-ray diffraction in order to produce a femtosecond switch for x rays. On the picosecond time-scale we used

coherent control of vibrational excitations as a means to exchange energy between two distinct diffracted beams in the anomalous transmission of x rays. But, we also used this new phenomenon to study the dynamics of the dense electron hole plasma generated in semiconductors following ultrafast laser-excitation. These experiments centered on the zone center coherent acoustic phonons generated through the deformation potential. However, the path to equilibration is much more complicated, involving processes that occur from tens of femtoseconds to microseconds. We are currently looking at the initial cooling of the electrons through the emission of phonons with wavevectors spanning the entire Brillouin zone, through time-resolved x-ray diffuse scattering. This resulted in the first truly microscopic of measurement of hot phonon emission, where we found a long-lived non thermal state in the important optoelectronic semiconductor InP [1].

7.9 Molecular alignment towards LCLS

In collaboration with the PULSE Institute at SLAC, we investigated methods to coherently align molecules for x-ray targets for LCLS high field and time-resolved x-ray scattering work. Two of our early results included the investigation of impulsive laser alignment of molecular iodine at a density of $\sim 10^{18}$ molecules/cm^3, i.e. high enough column densities for elastic scattering experiments at LCLS. Such coherent rotational ensembles maintain alignment for on the order of 0.5ps, which is enough for studies of photo-induced vibration or dissociation. We have also performed x-ray studies at SSRL of the iodine cation trimer I_3^- in solution or in a polymer matrix. In the latter case we can align the trimer by stretching the polymer, and this could be a very interesting method for producing aligned molecules at LCLS.

8 Products Developed

The primary products of the research was new knowledge that was disseminated in the form of high profile publications. In addition, as part of this research, several important collaborations were fostered, including with scientists at Argonne National Laboratory, the Sub-Picosecond Pulse Source team, the PULSE Institute at SLAC (where the two PIs are now Director and Deputy Director), and many of the scientists at LCLS. Reis was also one of the team leaders on the design of the x-ray pump-probe end station of LCLS, Bucksbaum was an author of the LCLS first experiments document, and both Reis and Bucksbaum participated in commissioning and early experiments on LCLS.

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B. SCIENTIFIC/TECHNICAL REPORTS

Final Scientific/Technical Report

Content. The final scientific/technical report must include the following information and any other information identified under Special Instructions on the Federal Assistance Reporting Checklist:

1. Identify the DOE award number; name of recipient; project title; name of project director/principal investigator; and consortium/teaming members.

2. Display prominently on the cover of the report any authorized distribution limitation notices, such as patentable material or protected data. Reports delivered without such notices may be deemed to have been furnished with unlimited rights, and the Government assumes no liability for the disclosure, use or reproduction of such reports.

3. Provide an executive summary, which includes a discussion of 1) how the research adds to the understanding of the area investigated; 2) the technical effectiveness and economic feasibility of the methods or techniques investigated or demonstrated; or 3) how the project is otherwise of benefit to the public. The discussion should be a minimum of one paragraph and written in terms understandable by an educated layman.

4. Provide a comparison of the actual accomplishments with the goals and objectives of the project.

5. Summarize project activities for the entire period of funding, including original hypotheses, approaches used, problems encountered and departure from planned methodology, and an assessment of their impact on the project results. Include, if applicable, facts, figures, analyses, and assumptions used during the life of the project to support the conclusions.

6. Identify products developed under the award and technology transfer activities, such as:

   a. Publications (list journal name, volume, issue), conference papers, or other public releases of results. If not provided previously, attach or send copies of any public releases to the DOE Project Officer identified in Block 11 of the Notice of Financial Assistance Award;

   b. Web site or other Internet sites that reflect the results of this project;

   c. Networks or collaborations fostered;

   d. Technologies/Techniques;

   e. Inventions/Patent Applications, licensing agreements; and

   f. Other products, such as data or databases, physical collections, audio or video, software or netware, models, educational aid or curricula, instruments or equipment.

7. For projects involving computer modeling, provide the following information with the final report:

   a. Model description, key assumptions, version, source and intended use;

   b. Performance criteria for the model related to the intended use;

   c. Test results to demonstrate the model performance criteria were met (e.g., code verification/validation, sensitivity analyses, history matching with lab or field data, as appropriate);

   d. Theory behind the model, expressed in non-mathematical terms;

   e. Mathematics to be used, including formulas and calculation methods;

   f. Whether or not the theory and mathematical algorithms were peer reviewed, and, if so, include a summary of theoretical strengths and weaknesses;

   g. Hardware requirements; and

   h. Documentation (e.g., users guide, model code).

Electronic Submission. The final scientific/technical report must be submitted electronically-via the DOE Energy Link System (E-Link) accessed at http://www.osti.gov/elink-2413.

Electronic Format. Reports must be submitted in the ADOBE PORTABLE DOCUMENT FORMAT (PDF) and be one integrated PDF file that contains all text, tables, diagrams, photographs, schematic, graphs, and charts. Materials, such as prints, videos, and books, that are essential to the report but cannot be submitted electronically, should be sent to the Contracting Officer at the address listed in Block 12 of the Notice of Financial Assistance Award.

Submittal Form. The report must be accompanied by a completed electronic version of DOE Form 241.3, "U.S. Department of Energy (DOE), Announcement of Scientific and Technical Information (STI)." You can complete, upload, and submit the DOE F.241.3 online via E-Link. You are encouraged not to submit patentable material or protected data in these reports, but if there is such material or data in the report, you must: (1) clearly identify patentable or protected data on each page of the report; (2) identify such material on the cover of the report; and (3) mark the appropriate block in Section K of the DOE F 241.3. Reports must not contain any limited rights data (proprietary data), classified information, information subject to export control classification, or other information not subject to release. Protected data is specific technical data, first produced in the performance of the award that is protected from public release for a period of time by the terms of the award agreement.