Time-resolved x-ray photoabsorption and diffraction on timescales from ns to fs

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Time-resolved x-ray diffraction with picosecond time resolution is used to observe scattering from coherent acoustic phonons in laser-excited InSb crystals. The observed oscillations in the crystal reflectivity are in agreement with a model based on dynamical diffraction theory. Synchrotron radiation pulses of ~ 300 fs in duration have been generated by femtosecond laser pulses modulating the electron beam in the Advanced Light Source.

1. INTRODUCTION

Time-resolved x-ray photoabsorption and diffraction enable one to probe the electronic and structural changes associated with phase transitions, solid state dynamics and chemical reactions. Important structural dynamics occur on the timescales from nanoseconds to femtoseconds with one fundamental limit set by vibrational periods, ~100 fs. Recent experiments, using both synchrotron and laser-plasma based sources, have observed phase transitions and chemical reactions on picosecond time-scales. In biology, real-time studies of photo-initiated reactions in complex molecules such as photoactive yellow protein (PYP) have been performed (1). Diffraction experiments utilizing laser-plasma x-ray sources have observed laser-induced disorder in Langmuir-Blodgett films (2) and a coherent acoustic pulse in GaAs (3).

Two approaches are being pursued at the Advanced Light Source (ALS): the first utilizing ultrafast detectors and the second developing a femtosecond x-ray source. The long range order of the semiconductor InSb has been probed by the diffraction of

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x-rays into a streak camera. Oscillations in the InSb crystal reflectivity are observed because of scattering from coherent acoustic phonons. A femtosecond x-ray source has been produced by co-propagating a femtosecond laser pulse with an electron bunch in a wiggler. In recent proof-of-principle experiments, we have successfully generated ~ 300 fs synchrotron pulses for the first time.

2. TIME-RESOLVED X-RAY DIFFRACTION OF InSb

An ALS bending magnet beamline (7.3.3) and a Si (111) monochromator crystal provide x-rays at a wavelength of 2.4 Å (4). The diffracted beam is then directed onto an InSb crystal oriented near the Bragg angle for the (111) reflection. To better match the penetration depths of the laser and x-rays, the crystal is asymmetrically-cut so that the diffracted beam leaves the crystal at a grazing angle of about 3 degrees. We use a Ti:Al_2O_3-based 150 fs, 1 kHz, 800 nm laser, synchronized to the synchrotron radiation time structure with jitter less than 5 ps. The time-resolved x-ray diffracted intensity following laser excitation is then measured using a streak camera triggered by a GaAs photoconductive switch (5). The time resolution of the camera is 3 ps.

We now discuss the observation of laser-induced coherent phonons at a fluence 20 % below the damage threshold of 15 mJ/cm^2. Figure 1 shows the time-dependent diffracted intensity measured at 0, +20, and +40 arcseconds from the Bragg peak.

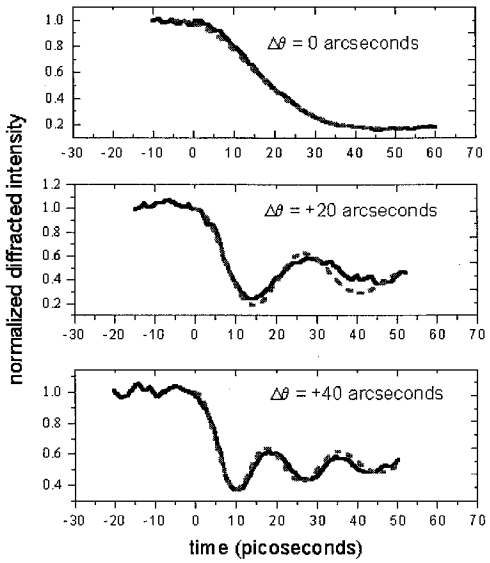

FIGURE 1. Experimentally measured (solid line) and simulated (dashed line) time-resolved diffracted intensity at crystal angles of 0, +20, and +40 arcseconds from the Bragg peak.

Impulsive excitation of a solid on a timescale shorter than the material's acoustoelastic response time generates coherent acoustic phonons across a range of wavevectors peaked at a wave-number of order one inverse laser penetration depth, 100 nm. As the crystal angle is varied, different phonon modes are probed out of the spectrum of excited modes. By wavevector matching considerations, the phonon frequency ω observed at a deviation $\Delta\theta$ from the Bragg angle θ of a symmetric crystal reflection is given by

$$\omega = v \mid \mathbf{G} \mid \Delta\theta \cot\theta, \qquad (1)$$

where v is the speed of sound within the crystal and **G** the reciprocal lattice vector.

Figure 1 includes calculated (normalized) diffracted intensities based on dynamical diffraction theory coupled to analytic solutions for the laser-induced strain profile. For an instantaneously heated crystal with assumed exponential temperature profile near the surface, the time-dependent strain profiles have been derived by Thomsen et al (6). There are three adjustable parameters in the model: the time for thermal transfer of energy to the lattice and the amplitudes of the thermal and deformation potential generated strain. The best fits correspond to a coupling time of 12 ps, a thermal strain of 0.17 % (just below that of InSb at its melting temperature) and a non- thermal contribution a factor of two smaller. We extract a sound velocity of ~ 4000 m/s, in agreement with the known value for InSb 3900 m/s (7).

At a slightly higher laser fluence, 10 % below the damage threshold, no

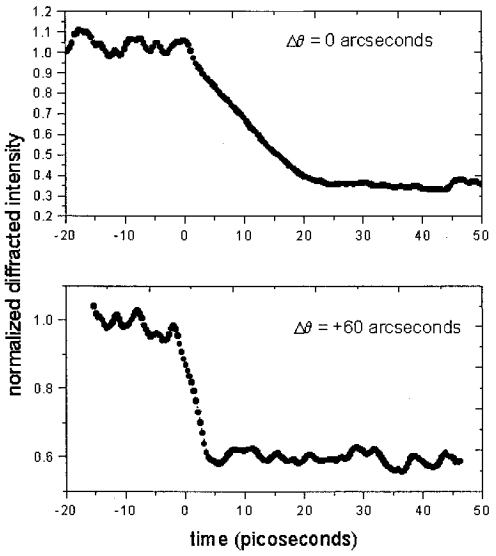

FIGURE 2. Time-resolved diffracted intensity at 0 and +60 arcseconds from the Bragg peak near the laser damage threshold. At 60 arcseconds, the diffracted intensity falls within 3 ps.

temporal oscillations occur, as shown in Fig. 2. At +60 arcseconds from the Bragg maximum, a 40 % reduction in intensity occurs on a time-scale limited by the streak camera time resolution (3 ps). In effect, only the first half-period of the oscillations induced at lower fluences are observed. We interpret these results as follows. At a critical fluence, the lattice is driven into a disordered state on a timescale set by one-half a phonon period. Since the observed 3 ps drop in the diffracted intensity occurs faster than the time for thermal transfer of energy to the lattice, we conclude that the first step in the observed disordering transition is essentially non-thermal in nature.

3. A FEMTOSECOND X-RAY SOURCE

The technique for generating femtosecond x-rays from the ALS is based on extracting a ~100 fs slice of the long (30 ps) electron bunch using a femtosecond laser pulse. (8) Co-propagation of a femtosecond laser pulse with the electron bunch through an appropriately tuned wiggler results in acceleration (and deceleration) of an ultrashort slice of electrons. These electrons will separate spatially from the main electron bunch in a dispersive section of the storage ring. Radiation from the modulated electrons at a bend magnet source can be imaged onto a slit isolating the ultrafast x-ray pulse.

The laser and wiggler emission in the near and far fields are overlapped using diagnostics at the wiggler front end. The spectrum of the laser is also matched to the fundamental of the wiggler. The efficiency of the interaction between the laser and electron beam is tested by measuring the gain in the laser pulse energy.

Femtosecond duration synchrotron pulses are directly measured by cross-correlating the visible light from bend-magnet beamline (6.3.2) at the ALS with the synchronized laser pulses in a non-linear crystal. Figure 3(a) shows a laser synchrotron cross-correlation measurement on a long time scale. The measured pulse duration, 39 ps FWHM, corresponds to the overall electron bunch duration. Measurement with higher time resolution (Fig. 3b) shows the femtosecond "dark"

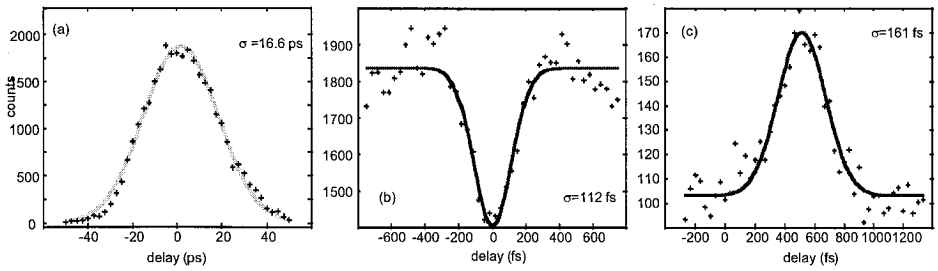

FIGURE 3. Cross correlation of the visible synchrotron pulse with a femtosecond laser pulse: (a) overall synchrotron pulse, (b) femtosecond dark pulse from on-axis radiation, (c) femtosecond pulse from off-axis radiation.

pulse (260 fs FWHM) originating from the central core of the electron bunch. Figure 3(c) shows a measurement with a knife edge at the $3\sigma_x$ position, where the femtosecond "bright" pulse is observed (380 fs FWHM). The femtosecond time structure will be invariant over the entire spectral range of bend-magnet emission from the near infrared to x-ray wavelengths. The observed pulse duration is limited by the position of the bend magnet beamline used in these characterization measurements. For a bend magnet immediately following the wiggler, an x-ray pulse duration of ~ 100 fs is calculated.

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