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Projecting picosecond lattice dynamics through x-ray topography

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A method for time-resolved x-ray diffraction studies has been demonstrated. As a test case, coherent acoustic phonon propagation into crystalline InSb is observed using a laser plasma x-ray source. An extended x-ray topogram of the semiconductor's surface was projected onto a high spatial resolution x-ray detector and acoustic phonons were excited by rapidly heating the crystal's surface with a femtosecond laser pulse. A correlation between the spatial position on the x-ray detector and the time of arrival of the laser pulse was encoded into the experimental geometry by tilting the incident laser pulse with an optical grating. This approach enabled a temporal window of 200 ps to be sampled in a single topogram, thereby negating the disadvantages of pulse-to-pulse fluctuations in the intensity and spectrum of the laser-plasma source. © 2002 American Institute of Physics. [DOI: 10.1063/1.1476957]

Both laser plasma x-ray sources and synchrotron radiation facilitate time resolved x-ray diffraction studies of light induced structural changes on a time scale of picoseconds. Notable successes at synchrotron sources have followed light induced structural rearrangements in crystals of macromolecules,^{1,2} have enabled the observation of coherent acoustic phonons,^{3,4} and have quantified large amplitude motions within a photoexcited chemical system.⁵ Laser plasma x-ray sources have facilitated gas phase⁶ and solution phase⁷ picosecond x-ray absorption experiments, and have enabled the growth of disorder in an organic sample,⁸ the nonthermal melting of a semiconductor surface,⁹ and the consequent propagation of a strainwave through a semiconductor lattice,¹⁰ to be visualized at near (or even sub) picosecond resolution.

When visualizing rapid structural rearrangements, the very high peak x-ray brilliance of synchrotron radiation provides a tremendous advantage. The choice either a polychromatic or a tunable monochromatic x-ray source with low angular divergence (as well as low pulse-to-pulse fluctuations) enables considerable freedom when designing an experiment. The primary disadvantage of synchrotron radiation is the relatively long duration of the x-ray pulses, which cannot be pushed significantly below 50 ps without severe loss of x-ray intensity.¹¹ The use of cross-correlation techniques¹² or x-ray streak cameras,⁴ however, have enabled improvements in the temporal resolution beyond the pulse length of synchrotron radiation when studying the laser induced melting and reordering of semiconductor surfaces. Schemes for generating synchrotron based x-ray pulses with a duration of a few hundred femtoseconds have also been

pursued,^{13,14} and used for time-resolved x-ray diffraction studies.¹⁵

Laser plasma x-ray sources provide pulses approximately two¹⁰ to three^{8,9} orders of magnitude shorter in duration than those available at synchrotrons. Although the peak number of x-ray photons generated is comparable with that generated at a synchrotron source, experiments using a laser-plasma source are hampered by a high beam divergence and large pulse-to-pulse fluctuations. In particular, laser plasma generated x-ray photons are emitted over the full sphere (10^8 photons/pulse in this study), and the integrated intensity can fluctuate by one or two orders of magnitude. Nevertheless, by accumulating a large body of experimental data, Rose-Petruck *et al.*¹⁰ succeeded in piecing together a movie following strain-pulse propagation through a semiconductor lattice after the surface was rapidly heated by a femtosecond (fs) laser pulse. The data were of sufficient quality that a quantitative description of the lattice dynamics could be given over a 250 ps window.

In this work an experimental configuration is presented which simultaneously takes advantage of natural beam divergence and negates problems associated with pulse-to-pulse fluctuations of a laser plasma x-ray source. The experimental setup is illustrated in Fig. 1 and is motivated by a proposal to use topography as a methodology for recovering subpicosecond temporal resolution in x-ray diffraction experiments.¹⁶ A crystalline sample of InSb was first aligned relative to the iron target laser plasma x-ray source so as to satisfy Bragg's condition for diffraction from a surface. Due to the natural divergence of the x-ray beam, an x-ray topogram could be recorded on a high-spatial resolution charge coupled device (CCD) camera. A 100 fs laser pulse ($\lambda = 800$ nm, 2.5 mJ/pulse) was used to rapidly heat the crystal's surface (experimental area ≈ 25 mm²) and its arrival relative to the x-ray

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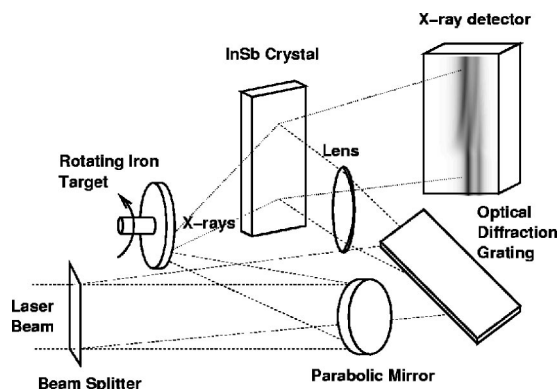


FIG. 1. Schematic of the experimental geometry. An optical diffraction grating tilts the fs laser pulse relative to its direction of propagation, thereby creating an optical path difference of up to 6 cm for the different rays incident upon the sample's surface. This enables a 200 ps window to be imaged in a single x-ray topogram using an iron target laser plasma x-ray source.

pulse was set by an optical delay line (not illustrated in the schematic). Although a single laser pulse heated the entire surface exposed to x rays, an additional time delay at different spatial locations along the crystal's surface was introduced by reflecting the laser pulse from a diffraction grating prior to it being imaged onto the sample (Fig. 1). As such the reflected wave front became tilted relative to its direction of propagation, with the result that different rays traversed different optical path lengths before arriving at the semiconductor's surface. Since the maximum difference in the optical path lengths of different rays was 6 cm, a temporal window of 200 ps was sampled in a single x-ray topogram (Fig. 1), reflecting the finite time required for the tilted wave front to sweep across the crystal's surface exposed to x rays.¹⁶ Through this approach the disadvantages of pulse-to-pulse fluctuations in the intensity and spectrum of a laser-plasma source were negated, since all sampled time points were exposed to an identical (accumulated) x-ray probe and, hence, were directly comparable.

Figure 2(a) illustrates a typical x-ray topogram from a crystal of InSb without any laser induced heating, as recorded on a CCD detector with high spatial resolution (25 μm per pixel). Because the x-ray source is polychromatic, different wavelengths satisfy the Bragg diffraction condition at different angles of incidence, hence, the two Fe $K\alpha$ lines of the laser plasma's target are projected as two parallel lines on the x-ray topogram [Fig. 2(a)]. When the sample is rapidly heated by the fs laser pulse, a structure in the x-ray topogram becomes visible [Fig. 2(b)], with one Fe $K\alpha$ line first being split, then remerging, as the position along the detector's x axis is increased. This change in the x-ray topogram reflects the fact that laser induced heating first perturbs the crystalline lattice and later, as energy disperses throughout the crystal, the original lattice spacing is recovered. The dependence of the measured scattering intensity on the spatial position of the detector is due to the correlation between the spatial position and the time of arrival of the laser pulse, which was encoded into the experimental geometry (Fig. 1). This results in an image which is a time-resolved rocking curve.

When the time of arrival of the x-ray pulse relative to the

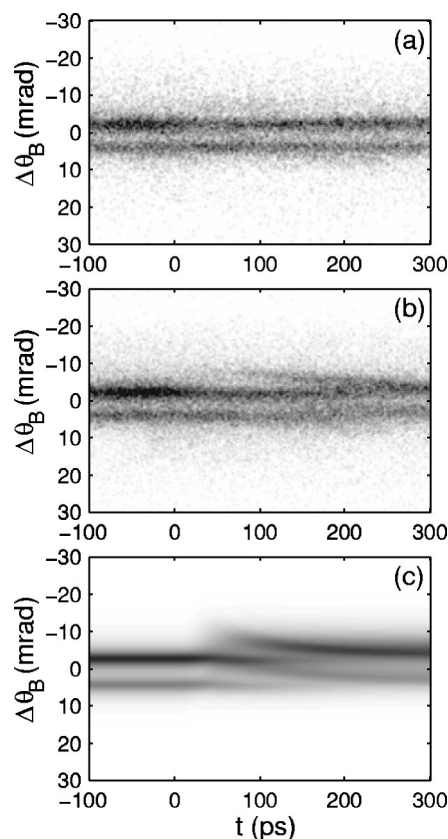


FIG. 2. Time-resolved rocking curves recorded from crystalline InSb using a high spatial resolution CCD camera. (a) Topogram recovered without laser excitation. (b) Topogram recovered following rapid heating by a fs laser pulse. (c) Theoretical simulation of the x-ray topogram when modeling the propagation of coherent acoustic phonon into crystalline InSb.

laser pulse was altered (by changing the optical delay line), all of the laser induced features of the x-ray topogram [Fig. 2(b)] were reproduced, but were translated along the x axis of the detector. This observation is quantified in Fig. 3, which plots the translation of the x-ray topogram versus the temporal change in the optical delay line. From this result the effective temporal resolution of the x-ray detector could be calibrated as 1.2 ps per pixel, which is in agreement with that determined from the optical configuration.

Although the acquisition time required to sample this 200 ps window was relatively short [data shown in Fig. 2(b) derived from a single 60 min run] the diffraction data were of sufficiently high quality that a good theoretical fit to the experimental results could be obtained. In Fig. 2(c) we show a simulated x-ray topogram for comparison. A detailed description of the theoretical model is given in Refs. 17 and 18,

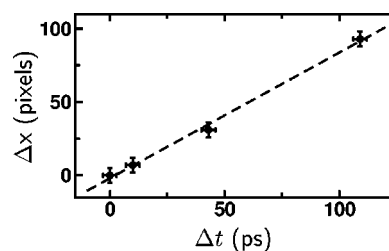


FIG. 3. Temporal calibration of the x-ray topogram. When the time of arrival of the laser pulse relative to the x-ray pulse was altered, the features seen in the x-ray topogram [Fig. 2(a)] became translated.

and it has proven to be applicable to a broad spectrum of experimental results.^{3,10} Intuitively, energy from the fs laser pump couples into the semiconductor by exciting electrons from the valence to the conduction band. After absorption, energy is initially dispersed as optical phonons, which are characterized by specific motions of atoms within the unit cell. On a slower time scale coherently excited acoustic phonons are generated,^{3,10} which are characterized by specific motions of the crystalline lattice and their propagation is determined by the optical, electronic and acoustic properties of the material.¹⁷ The propagation of these acoustic waves through the semiconductor perturbs the lattice spacing on a time scale of picoseconds, and results in the splitting (and subsequent relaxation) of the Fe $K\alpha$ line modeled in Fig. 2(c).

Several extensions to the experimental protocol presented here are immediately foreseeable. The use of a large area x-ray camera with a high spatial resolution would enable several diffraction spots to be recorded simultaneously, thereby recovering more structural information than that accessible from surface diffraction alone. Furthermore, the potential temporal resolution can be manipulated by the choice of experimental geometry. Should the fs laser pulse sweep directly across the sample's surface¹⁶ then the effective temporal resolution of each pixel of the x-ray camera would be a few tens of femtoseconds. Such a change in the experimental geometry would not result in any loss in signal-to-noise, although the temporal resolution achievable in practice, even using a deconvolution algorithm, cannot be pushed significantly beyond the duration of the x-ray probe.¹⁹ Nevertheless, when used in conjunction with sufficiently short x-ray pulses, the experimental approach presented here promises a direct route to observing subpicosecond phase transitions during laser induced melting of well-diffracting samples.

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