ELSEVIER
Nuclear Instruments and Methods in Physics Research A 467–468 (2001) 986–989
NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A
www.elsevier.com/locate/nima

Ultrafast X-ray diffraction of laser-irradiated crystals

P.A. Heimann[a,*], A.M. Lindenberg[b], I. Kang[b], S. Johnson[b], T. Missalla[c], Z. Chang[d], R.W. Falcone[b], R.W. Schoenlein[e], T.E. Glover[a], H.A. Padmore[a]

[a] *Advanced Light Source, Lawrence Berkeley National Laboratory, 1 Cyclotron Road, MS-2-400, Berkeley, CA 94720, USA*
[b] *Physics Department, University of California, Berkeley, CA 94720, USA*
[c] *Lawrence Livermore National Laboratory, Livermore, CA 94551, USA*
[d] *Center for Ultrafast Optical Science, University of Michigan, Ann Arbor, MI 48109, USA*
[e] *Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA*

Abstract

Coherent acoustic phonons have been observed in the X-ray diffraction of a laser-excited InSb crystal. Modeling based on time-dependent dynamical diffraction theory has allowed the extraction of fundamental constants, such as the electron-acoustic phonon coupling time. A dedicated beamline for time-resolved studies has been developed at the Advanced Light Source with special considerations toward high transmission, low scattering and a wide photon energy range. The facility combines a bend magnet beamline, time-resolved detectors and a femtosecond laser system. © 2001 Elsevier Science B.V. All rights reserved.

PACS: 61.10.−e; 07.85.Qe

Keywords: Time-resolved X-ray diffraction; Synchrotron radiation beamline

1. Time-resolved X-ray diffraction of InSb

Laser sources currently produce ultrashort pulses of infrared, visible and ultraviolet wavelengths, which can be used to study the dynamics of valence electrons in atoms, molecules and solids. However, to directly probe structural properties, the methods of choice are X-ray diffraction and X-ray absorption fine structure (XAFS). Important structural dynamics occur on time scales from nanoseconds to femtoseconds with one fundamental limit set by a vibrational period, ~100 fs. Recent experiments, using both synchrotron and plasma X-ray sources, have observed chemical reactions and phase transitions. In biology, real-time studies of photo-initiated reactions in complex molecules such as photoactive yellow protein (PYP) have been performed [1].

In the femtosecond optical excitation of solids, the ultrafast excitation of carriers can lead to several interesting phenomena such as ultrafast melting and generation of coherent phonons. The time scale of a phase transition is normally limited by the electron-phonon coupling times, thermal diffusion inside the material and the rate of heating. The melting of InSb in 300 fs has been directly observed by X-ray diffraction using a laser plasma source [2]. At lower fluence, the ultrafast heating of a thin layer at the crystal surface results in the generation of coherent acoustic phonons.

*Corresponding author. Tel.: +1510-486-7628; fax: +1510-486-7696.
E-mail address: heimann@lbl.gov (P.A. Heimann).

PII: S0168-9002(01)00547-2

The spectrum of acoustic phonons is peaked at a wavevector of order one inverse laser penetration depth.

This study focused on the coherent acoustic phonons of laser-irradiated InSb [3]. The (1 1 1) reflection was studied at an X-ray wavelength of 2.4 A. 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°. The laser fluence was 20% below the damage threshold of 15 mJ/cm^2 allowing recovery of the crystal between laser shots. By varying the crystal angle different acoustic phonon modes are probed according to wavevector matching considerations.

Fig. 1 shows the time-dependent diffracted intensity measured at 0, +20, and +40 arcseconds from the Bragg peak. The figure also includes calculated (normalized) diffracted intensities based on dynamical diffraction theory coupled to an analytic solution for the laser-induced strain profiles [4]. There are three adjustable parameters in the model: the electron–phonon coupling time and the amplitudes of two kinds of strain. A thermal strain develops over the electron-acoustic phonon coupling time, while the strain from the deformation potential is effectively instantaneous. The best fits correspond to a coupling time of 12 ps, a thermal strain of 0.17% and a non-thermal contribution a factor of two smaller. At higher laser fluence, 10% below the damage threshold, no temporal oscillations occur. This experiment studying coherent acoustic phonons demonstrates requirements for high time resolution and high flux.

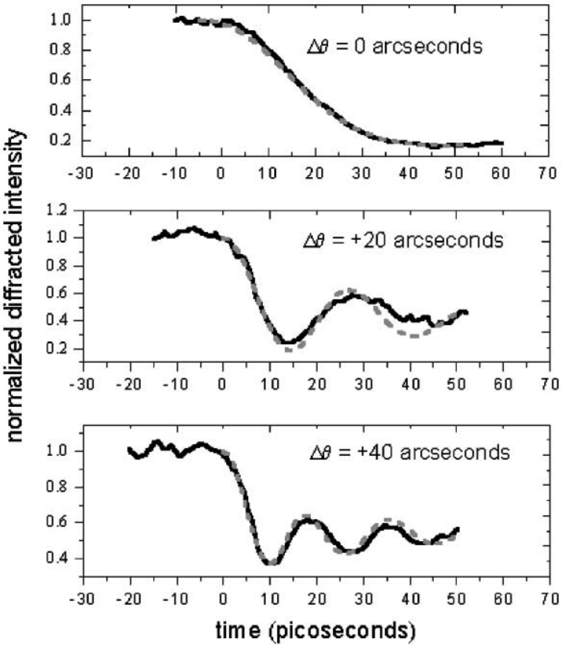

Fig. 1. Experimentally measured (solid line) and simulated (dashed line) time-resolved diffracted intensity at crystal angles of 0, +20, and +40 arcseconds from the Bragg peak.

2. ALS beamline 5.3.1

The Advanced Light Source beamline 5.3.1, which has recently been completed, is dedicated to time-resolved laser pump/X-ray probe experiments. This bend magnet beamline is designed for high transmission, low scattering, and a wide photon energy range. Special requirements are set by the laser-electron beam modulation technique (described in this conference by Schoenlein et al.), which generates femtosecond X-ray pulses by propagating ultrashort laser pulses through the sector 5 wiggler and exchanging energy with the electron beam.

The beamline 5.3.1 is located at the center bend magnet in the arc sector directly following the wiggler straight section. The center bend magnet provides the maximum dispersion, which determines the horizontal displacement of the modulated electrons. The location of the bend magnet also limits the time-of-flight broadening of the femtosecond pulse to <100 fs.

High X-ray flux is required because of the mismatch of the repetition rates of the synchrotron radiation and the femtosecond laser system. The Advanced Light Source produces X-ray pulses at 500 MHz while the laser amplifier operates at 5 kHz. A large aperture 3×0.3 mrad^2 is collected from the bend magnet source. Secondly, a minimum number of optical elements, one toroidal mirror and two crystals, are used.

In the laser-electron beam modulation technique, a fraction of the electrons, $\sim 10^{-3}$, is displaced horizontally by $\sim 5\sigma$ or 0.5 mm.

Producing a nearly Gaussian horizontal focus is important for separating the femtosecond X-ray pulse from the background of X-rays originating from unperturbed electrons. X-ray scattering in the horizontal plane is minimized by having the mirror reflect in the vertical plane. For a mirror with a grazing incidence angle, in-plane scattering is greater than out-of-plane scattering because of the different length scales of roughness sampled [5]. Measurements of the X-ray beam profile from a similar mirror at ALS beamline 7.3.3 demonstrated that the scattering in the horizontal plane can be suppressed by 6×10^{-4} compared to the peak specular intensity.

The beamline is designed to take advantage of the whole spectrum emitted by the bend magnet source. By employing differential pumping instead of a beryllium window both hard and soft X-rays may be provided by the same beamline. The visible light from a separate mirror in the front end will be used as a timing diagnostic.

Fig. 2 shows a schematic diagram of the beamline. In the front end a mirror with 10° incidence angle reflects visible light. A toroidal mirror provides 1 : 1 image of the source. After the mirror there is a pair of slits, which may be used to reduce the angular aperture. A differential ion pump and 1000 Å thick C foil provide a vacuum barrier between the UHV part of the beamline upstream and the chambers inside the hutch at 10^{-6} Torr pressure. Just inside the X-ray hutch is a chopper, which reduces the average power by an order of magnitude. The next optical component is a two crystal monochromator.

Visible light is reflected from the front end mainly for a diagnostic of the laser-electron beam modulation. A side-cooled plane mirror reflects $6 \times 2\,\mathrm{mrad}^2$ of visible synchrotron radiation toward the roof of the accelerator tunnel. In the hutch the visible synchrotron radiation will be crossed with optical pulses from the laser in a nonlinear crystal. Such cross correlation measurements provide a determination of the X-ray pulse duration.

The M1 toroidal mirror produces a 1 : 1 image of the bend magnet source in the hutch. The 900 mm long silicon mirror was polished as a cylinder by Frank Cooke, Inc. The mirror is then bent into a toroidal shape by a S-spring bender, which also provides side cooling. To preserve the vertical bend magnet source size with a 12 m focal distance is difficult. From the long trace profiler data, a spot size of 240 μm horizontal × 110 μm vertical is calculated with a 490 mm length of the mirror illuminated.

The double crystal monochromator covers the photon energy range from 1.8 to 12 keV. Si(1 1 1), Ge(1 1 1), Ge(3 1 1) and InSb(1 1 1) crystals will alternatively be used. The first crystal is back cooled, and the second crystal translates to maintain constant exit beam height. The beamline flux has been calculated including the mirror reflectivity, foil and Ge(1 1 1) crystal transmission as well as the repetition rate of the laser. The flux available for picosecond pump-probe experiments is $\sim 2 \times 10^8$ 1/s. The flux in the femtosecond X-ray pulses is $\sim 2 \times 10^5$ 1/s because of the duration of the modulating laser pulse compared with the

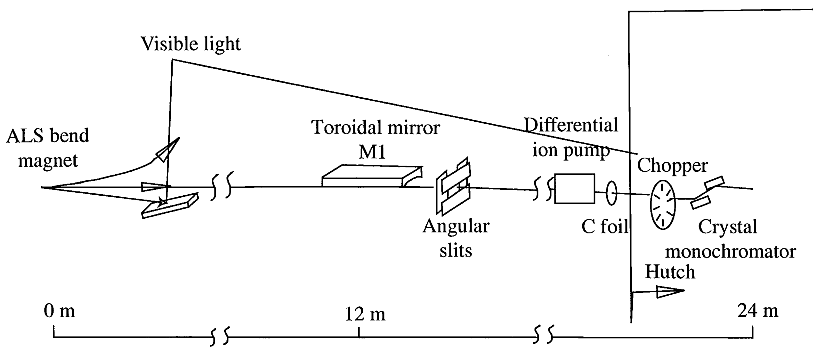

Fig. 2. Schematic diagram of ALS beamline 5.3.1.

electron bunch and because of a modulating efficiency. In general, these calculated intensities will meet the requirements of X-ray diffraction and near-edge X-ray absorption experiments.

The laser system consists of a $Ti:Al_2O_3$ oscillator and regenerative amplifier operating at a repetition rate of 5 kHz. The pulse duration is 100 fs, and the pulse energy is 1 mJ. The laser pulses are synchronized to the X-ray pulses by locking the laser oscillator frequency to the master RF clock of the ALS.

The X-rays are detected by an avalanche photodiode, microchannel plates or streak camera depending on the requirements of the experiment. Avalanche photodiodes and microchannel plates have sufficient time resolution to detect individual ALS X-ray pulses. Avalanche photodiodes have a quantum efficiency of ~ 1. Microchannel plates are suited to be used with phosphor screen and CCD camera as an imaging detector. The time resolution of the streak camera is 2 ps in averaging mode [6,7]. The synchronization of the streak camera to the laser is maintained by a GaAs photoconductive switch, which generates the high-voltage pulse for the sweep.

Acknowledgements

This work was supported by the US Department of Energy through Lawrence Berkeley and Lawrence Livermore National Laboratories. The ALS was supported by the Director, Office of Science, Office of Basic Energy Sciences, Materials Sciences Division of the US Department of Energy under Contract No. DE-AC03-76SF00098.

References

[1] B. Perman et al., Science 279 (1998) 1946.
[2] A. Rousse et al., Conference on Lasers and Electrooptics 2000, San Francisco, CA, 2000.
[3] A.M. Lindenberg et al., Phys. Rev. Lett. 84 (2000) 111.
[4] C. Thomsen, H.T. Grahn, H.J. Maris, J. Tauc, Phys. Rev. B 34 (1986) 4129.
[5] E.L. Church, P.Z. Takacs, Surface scattering, Handbook of Optics, 7.1, Vol. 1, McGraw-Hill, Inc., New York, 1975.
[6] Z. Chang et al., Appl. Phys. Lett. 69 (1996) 133.
[7] J. Larsson et al., Optics Lett. 22 (1997) 1012.