Ultrafast time-resolved X-ray diffraction from laser-irradiated crystals

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Abstract.

We show results from laser pump/X-ray probe experiments. We utilize a laser, which provide pulses with duration of 100 fs, in order to melt or heat a semiconductor crystal. The structural order is subsequently probed by time-resolved x-ray diffraction. We use techniques providing a temporal resolution, which is not limited by the duration of the X-ray pulse. The experiments have been carried out at the Advanced Light Source (ALS) and European Synchrotron Radiation Facility (ESRF) synchrotrons.

1 Introduction

The use of time-resolved x-ray diffraction for studies of disordering of crystals has recently obtained a great deal of attention [1-5]. In this paper, we apply time-resolved x-ray diffraction in order to study the properties of semiconductors following irradiation with ultrashort pulse lasers.

Theoretical studies have suggested that excitation of about 10% of the valence electrons in crystals with a diamond structure would give rise to melting even though the lattice temperature was below melting. A more speculative result of these studies is that this bond softening could result in structural instability which give rise to a transition into a new ordered phase[7]. By probing laser-irradiated material using ultrafast time-resolved x-ray diffraction, it should be possible to directly determine the mechanism for ultrafast disordering. However, the lack of

both ultrashort-pulse x-ray sources and ultrafast x-ray detectors has so far prevented such studies.

2 Experimental set-up

We have used two similar synchrotron radiation beamlines at the Advanced Light Source (ALS) in Berkeley and at the European Synchrotron Radiation Facility (ESRF) [25 ,26]. The Berkeley beamline is described in some detail and shown in Figure 1. A Ti:Al_2O_3-based 100fs, 1 kHz laser is synchronized to a electron storage ring with jitter less than 10 ps. A bending magnet beamline source emits a broad spectrum of x-ray radiation, extending to a useful photon energy of about 12 keV. The delay between the laser and the x-ray pulse can be varied using an optical delay line in the laser beam path. X-rays are monochromatized and focused onto the sample. Diffracted x-rays are detected by a x-ray sensitive avalanche photodiode (APD) which has a yield of about 10^5 electrons per incident x-ray photon. The response time of this detector is 10 ns. The signal is recorded using a digital oscilloscope or a gated-integrator connected to a computer. In all the pump probe experiments discussed below, the gated-integrator was used, since that allowed us to record data at 1 kHz, as determined by the laser repetition rate.

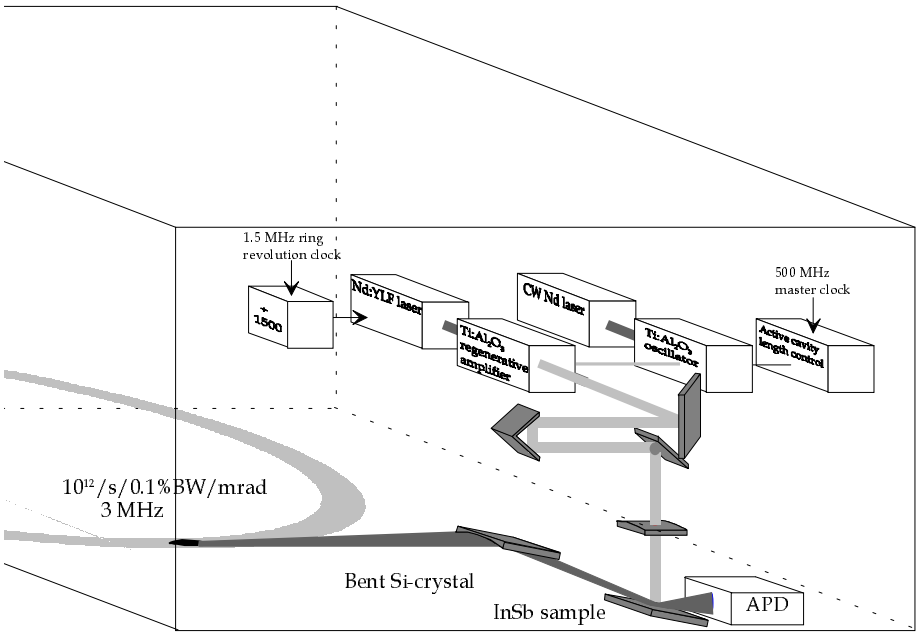

Figure 1. Experimental set-up

3 Melting and recrystallization of InSb

In order to obtain a sufficiently good S/N ratio in our experiments, data must be acquired over multiple shots. We thus rely on the fact that the sample recover between laser pulses (< 1 ms). It is therefore relevant to ask if phase-transitions such as melting are followed by regrowth. The fluence incident on target is typically 30 mJ/cm^2 . We do not observe irreversible damage of the InSb; even after exposure times totaling more than 1 hour we do not observe a noticeable decrease in the x-ray signal. However at a 20% higher fluence we observe irreversible damage within less than a second; it is both visible to the eye and results in a lower time-averaged x-ray diffraction efficiency of about 30%.

In order to understand the origin of the observed recovery, we constructed a simple model of the recrystallization of a melted surface layer. The simulation solves a one-dimensional diffusion equation in two regions, a liquid surface region on a heated solid region with a moving boundary condition, the liquid-solid interface. The propagation velocity is determined by balancing the latent heat carried by the melt-front with the heat conducted into the solid and liquid regions. The initial temperature profile is derived from the linear absorption properties of InSb given an initial laser fluence of 40 mJ/cm^2 The temperature dependence of the thermal conductivity of InSb is included. Using a Crank-Nicholson method, the computed regrowth velocity is 5 m/s. We note that the speed of the regrowth is so slow that it can not explain the partial recovery manifesting itself as a minimum in the delay scan when the laser is incident 150 ps before the x-ray pulse.

We were able to study the regrowth process by recording rocking curves for different temporal delays between the laser pump and X-ray probe as shown in figure 2. We find little or no broadening of the rocking curves and an attenuation which we attribute to the photo-absorption in the disordered layer. Assuming that the drop in intensity is due to the photo-absorption we determine the thickness of the disordered layer to be 110 nm.

When recording rocking curves for longer time-delays we find that the intensity increases with time. We attribute this slower recovery to recrystallization By looking at the intensity of the rocking curves obtained at a delays of 2 ns and 36 ns we can estimate the recrystallization speed of the material to be approximately 2 m/s. We find this in agreement with the simple 1-D model for regrowth.

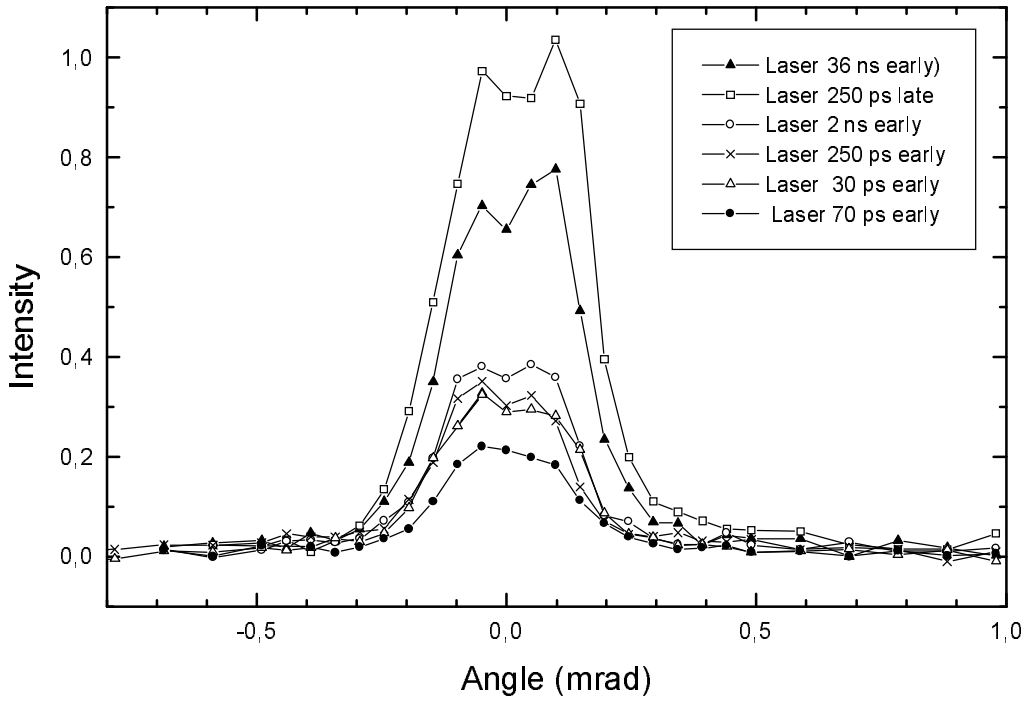

Figure 2. Rocking curves for different time-delays show the depth of the molten layer.

4 Melting and strainwaves in GaAs

We have recently observed laser-induced melting of GaAs on a time scale of 10 ps or faster using the ID9 beamline and the ESRF ultrafast x-ray streak camera, as shown in figure 3. We furthermore directly observe propagation of a strain wave through the wafer (Figure 4). This strain wave can also be interpreted as an excitation of a range of acoustic phonon modes.
The current results will be important in designing experiments which will elucidate the role of optical and acoustic phonons in the disordering of semi-conductors following laser illumination.

The results shown here represent the fastest event recorded at the ESRF and can be used as a straight-forward way of producing rapid structures in the time-profile of the x-ray radiation. Theoretical modelling is ongoing in order to interpret the results and to show the complementary nature on models based on phonon theory and on dynamical diffraction theory.

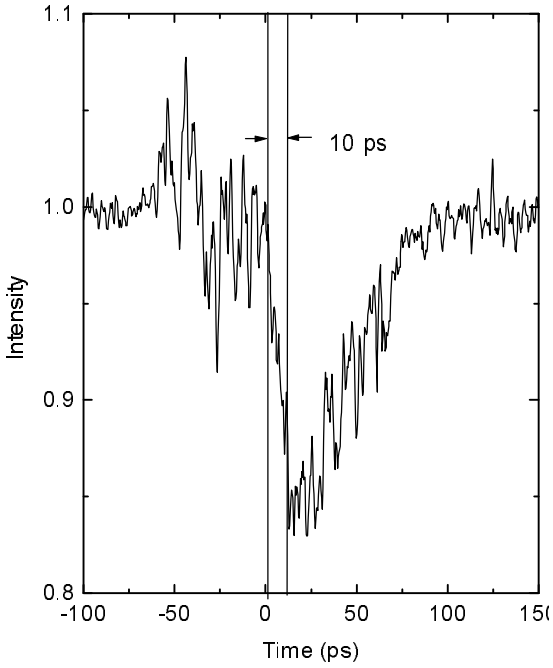

Figure 3. The degree of order curve correspond to the intensity of a bragg spot.

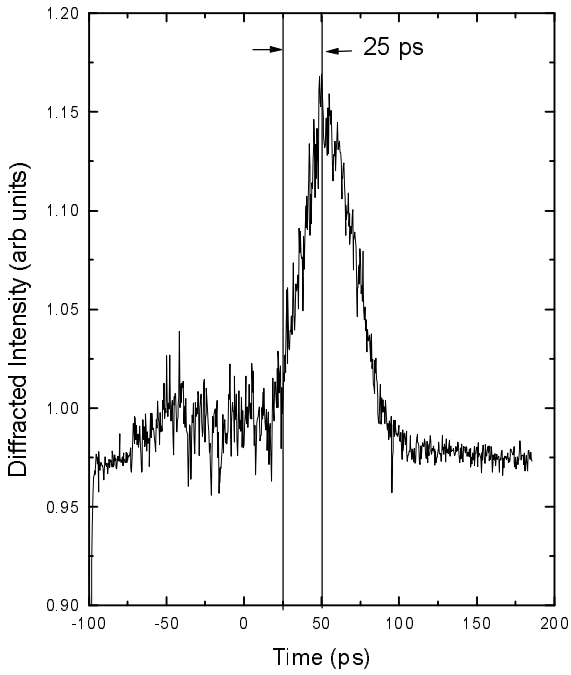

Figure 4. Laser-induced strain causes an increase of the bragg diffracted intensity on the negative side of the rocking curve.

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References.

1. J. Larsson et al., Applied Physics **A66**, 587 (1998)
2. C. Rose-Petruck et al., Nature **398**, 210 (1999)
3. C. Rischel et al., Nature **390**, 480 (1997)
4. J. Larsson et al., Opt Letters **22**, 1012 (1997)
5. A.H. Chin et al. Phys. Rev. Lett. **83**, 336 (1999)
6. A.M. Lindenberg et al, "Time-resolved X-ray diffraction from Coherent Phonons during a Laser-Induced Phase Transition", To appear in Phys. Rev. Lett.
7. P. Stampfli and K. H. Bennerman, Phys. Rev. **B 42** 7163 (1995)