Time-resolved X-ray diffraction studies of laser-induced acoustic pulse generation in semiconductors using synchrotron radiation

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Abstract. Laser-induced lattice deformation generating acoustic pulses has been studied by time-resolved X-ray diffraction method using a brilliant synchrotron radiation source. We constructed a time-resolved X-ray diffraction system equipped with a synchronized femtosecond pulsed laser at an undulator beamline of SPring-8 facility, and observed laser-induced acoustic pulse in a semiconductor wafer of GaAs, where the laser irradiation causes initial strain of expansion with a response time of around 200 ps. High resolution X-ray diffractometry in asymmetric configuration combined with laser-pump X-ray probe method revealed that the strain is formed with coherent longitudinal acoustic phonons and lattice expansion along the surface normal. The laser power dependence shows the saturation of the lattice expansion ratio with the lengthened relaxation time for high power excitation.

1. Introduction
When the surface of a semiconductor plate is irradiated by a femtosecond pulsed laser, transient lattice deformation is generated around the surface and induces acoustic pulse propagating toward the rear surface. The pulse is then reflected at the rear surface and the deformation is again observed at the front surface at a certain delayed time. The repetitive reflection is known as acoustic pulse echoes. This phenomenon has been investigated by time-resolved optical monitoring method [1] which reveals the surface displacement and the shape. On the other hand, X-ray diffraction (XRD) is powerful method to investigate the lattice deformation. At the same time, a brilliant X-ray source is required when the XRD is combined with time-resolved technique to watch the transient lattice deformation in acoustic pulse echoes. Significant progress in recent development of synchrotron radiation (SR) X-ray sources has offered the opportunity to observe the laser induced acoustic pulse generation and the echoes by time-resolved (TR) XRD method [2,3]. The high quality of SR X-ray beam enabled to combine time-resolved technique and XRD method with high momentum resolution such as triple crystal diffractometry (TCD). By the TCD, where three crystals are the monochromator to generate a collimated and monochromatic incident X-ray beam, a sample, and an angle analyzer for diffracted beam, we can identify the type of lattice strain and know the direction and degree of lattice deformation [3]. We have been investigating the laser-induced acoustic pulse generation and the echoes in semiconductor wafers of gallium arsenide (GaAs) and silicon (Si) by TR-TCD at a SPring-8...
undulator beamline, equipped with a femtosecond pulsed laser system. Initial strains are expansion and shrinkage for GaAs(100) crystal and Si(111) crystal, respectively, and their echo pulses with several-tens repetition have the corresponding phase [2]. Since the lattice of GaAs expands with fast response time of about 200 ps, we conducted pump-probe method where the time-resolution is determined by the SR pulse duration of 40 ps (FWHM).

In this paper, we show the TR-TCD method with asymmetric configuration to investigate the initial strain generating acoustic echoes in a single crystal of GaAs, and report the deformation direction, phases of acoustic waves, and the laser power dependence of deformation.

2. Experimental
The TR-TCD measurements were conducted at an undulator beamline of BL19LXU of SPring-8 SR facility. Figure 1(a) shows the experimental setup. A 0.6 mm-thick, 76 mm-diameter, non-doped GaAs(100) wafer was aligned using a high precision diffractometer in the experimental hutch. The incident X-rays were monochromatized to be 21.3 keV, so that the X-rays are diffracted by the 620 plane of GaAs with a grazing exit angle of about 0.5°. In order to get a smaller energy bandwidth of incident X-rays, a Si 440 channel-cut crystal was used, in addition to a symmetric Si 111 double-crystal monochromator. The diffraction intensity from the sample is detected through a Si 111 analyzer crystal. The energy bandwidth and angle resolution were about ΔE/E = 9 × 10⁻⁶ and Δθ=30 μrad, respectively. These correspond to a resolution window of 3.2 × 10⁻⁴ (in the direction of Δθ) by 6.3 × 10⁻⁵ (ΔG). A diffraction intensity map in reciprocal space can be obtained by scanning the angles of diffractometers of D1 and D2.

A mode-locked Ti:sapphire laser with a regenerative amplifier, which has a pulse duration of 130 fs, wavelength of 800 nm, a pulse energy of 130 μJ, and repetition rate of 1 kHz, was used for irradiation of the GaAs. The laser beam diameter at the sample surface was about 1.8 mm, covering the X-ray diffracting area (100 μm by 100 μm). The penetration depth of the laser, 0.7 μm, is about 2 times larger than the extinction depth of X-rays. The timing of the laser shot was controlled by the laser trigger applied through an electronic circuits composed of RF phase shifter from the radio frequency (RF) signal of the master oscillator of the SR storage ring determining the SR X-ray pulse timing. The stability of the timing between the laser and SR pulses has been evaluated to be less than 5 ps [4], which is much better than the X-ray pulse duration of 40 ps (FWHM). The X-ray photons diffracted from GaAs 620 were detected by an avalanche photodiode. The counts only from the target SR pulse are picked up by using an electronic gate synchronized with the RF master oscillator of the ring. The data were accumulated for 10 s for each point in a reciprocal space map.

Figure 1. (a) Experimental setup of TR-TCD using a pulsed X-ray SR beam. (b) Corresponding configuration in reciprocal space.
3. Results and discussion

Figure 2(a), (b) and (c) show the X-ray diffraction snapshots of GaAs 620 in reciprocal space, obtained for no laser irradiation, $\Delta t=180$ ps, and $\Delta t=1$ ms after the irradiation by the laser with a pulse energy of 5 mJ/cm$^2$, respectively. Asymmetric distribution along the [100]-axis is seen in figure 2(b), which is due to acoustic phonon generation. Figure 3 shows the plot of the peak position of the diffraction intensity distribution for $\Delta t= (i)1$ ms, (ii)180 ps, (iii) 300 ps, and (iv) 800 ps. It is found that the transient expansion occurs along a dashed vertical line parallel to the surface normal direction, and that the diffraction peak for about 1 ms after the irradiation is shifted towards an inclined dashed line showing the direction of $G_{620}$ with respect to the position without laser irradiation $O'$. The isotropic lattice expansion, $|\Delta d_{620}|/d_{620}$, is estimated from figure 3, to be $1.4 \times 10^{-5}$, which corresponds to the heating by 2.4 K. The maximum anisotropic transient expansion in the direction of the surface normal, is also evaluated to be $|\Delta d_{100}|/d_{100} =2.2 \times 10^{-5}$ at a delay time of 300 ps.

The fast lattice expansion is, then, observed by measuring the diffraction intensity as a function of the angle of analyzer crystal because $\Delta \theta$ direction is almost parallel to the [100]-direction as shown in figure 1(b). Figure 4 shows time dependence of the diffraction intensity profile in the $\Delta \theta$ direction, as obtained by changing an analyzer angle with a fixed sample angle. In order to discuss the lattice expansion and phonon oscillation separately, the shift, $\theta_p$ of principal peak in figure 4 is plotted in figure 5(a). The peak shift of the profile to minus angle direction indicates the lattice expansion whose maximal spacing is achieved at around 300 ps. The time dependence of the diffraction intensity profile with an axis of $\Delta \theta - \theta_p = (\Delta \theta')$ was drawn in order to pick up the acoustic phonon oscillation. The time-dependence of diffraction intensity at $\Delta \theta'$ is shown in figure 5(b). Acousto-optic effect produced by laser-induced longitudinal acoustic (LA) phonon and incident X-rays is shown as out of phase oscillation with a frequency of GHz region at side bands around the principal peak shifted due to lattice expansion. Figure 5(b) shows the diffraction intensity modulation depending on the off-set angle, which should have the relation of $\Omega = v|k||\Delta \theta'|$, where $\Omega$ is the angular frequency of the intensity modulation, $v$, the speed of sound, and $k$, the wavevector of X-rays [5].

From the relation between the off-set momentum and the oscillation frequency as shown in figure 5(c), the speed of acoustic phonon is roughly estimated using the above relation, to be $v=3400$ m/s, which is comparable with the speed of sound of GaAs in bulk.

![Figure 2](image1.png)

**Figure 2.** The snapshots of GaAs 620-Bragg diffraction intensity distribution for (b) $\Delta t=180$ ps and (c) $\Delta t=1$ ms. (a) is the Bragg spot without laser irradiation.

![Figure 3](image2.png)

**Figure 3.** The peak position of the diffraction intensity distribution moving with $\Delta t$ of (i)1 ms, (ii)180 ps, (iii) 300 ps, and (iv) 800 ps. An inclined dashed line indicates the direction of $G_{620}$. 
Figure 4. Time-dependent angular distribution of X-ray diffraction intensity for GaAs 620.

Figure 5. (a) The peak shift, $\theta_p$, and (b) oscillatory part at detuned angles $\Delta \theta' = \Delta \theta - \theta_p$, obtained from figure 4. In (b), the detuned angles are, $\Delta \theta' = (i) 46, (ii) 33, (iii) 26, (iv) 19, (v) -18, (vi) -25, (vii) -32, and (viii) -47 \mu$rad. Figure 5(c) shows the $\Delta \theta'$-dependence of the frequency of diffraction intensity oscillation, $\Omega$.

Figure 6. The laser power dependence of the lattice expansion measured for 400-GaAs diffraction. (a) The maximum expansion ratio and (b) relaxation time.

The laser power dependence of the lattice expansion obtained by the peak shift was also measured for 400-Bragg diffraction from a GaAs wafer. Figure 6 (a) and (b) show the laser power dependences of maximum expansion ratio and relaxation time, respectively. The maximum expansion ratio is almost proportional to the laser power for relatively small excitation; for the high power excitation, the ratio is saturated as the relaxation time becomes longer.

4. Conclusions
Laser-induced lattice deformation generating acoustic pulses in a semiconductor wafer of GaAs has been studied by TR-TCD using the SPring-8 SR source. The time-evolution of diffraction profile shows that the strain is formed with coherent LA phonon generation and lattice expansion along the surface normal. The maximum lattice expansion ratio is saturated for high power laser excitation.

References
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