

Time-resolved X-ray diffraction from semiconductor crystals irradiated by ultrashort laser pulses

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Abstract

For ultrafast material analyses, we constructed the time-resolved X-ray diffraction system utilizing ultrashort X-rays from laser produced plasma generated by the 12TW-50fs laser at Nuclear Engineering Research Laboratory, University of Tokyo. Ultrafast transient changes in laser-irradiated GaAs crystals were observed as X-ray diffraction patterns. Clear subpeak appeared in the X-ray profile when the crystals were irradiated with 1.3 mJ/mm^2 laser pulses with 150 ps delay time. Numerical analyses were also carried out for the X-ray diffraction from the crystal with the laser-induced strain wave and compared with the experimental results.

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I. INTRODUCTION

Time-resolved pump-and-probe analysis is used for investigation of time-dependent transient phenomena. By development of ultrashort pulse generation techniques in the past decade, pico- to femto-second time resolution has been made available in dynamic analyses of material properties that have been performed in static manners so far. This has brought us the possibility of direct observation of ultrafast phenomena with the time resolution even less than 1 ps. For example, femtosecond evolutions of electronic states in solids [1] and photo-initiated chemical reactions [2] have been examined by laser pump-and-probe spectroscopy and laser-flash-photolysis using ultrashort lasers which have pulse durations less than 100 fs typically. In the field of X-ray application, as well, temporal changes of atomic arrangement have been investigated by means of the time-resolved X-ray diffraction mainly using synchrotron radiations with the picoseconds time resolution [3] [4]. Pico- to subpico-second X-ray pulses have become producible by the laser-based X-ray generation techniques that have been introduced by recent developments in ultraintense laser technologies such as a table-top tera-watt laser. The Thomson scattering of ultrashort laser pulses by relativistic electron beams produces energy-tunable sharply directed X-ray pulses [5]. An ultraintense laser focused on solid targets provides laboratory-scale sources of intense ultrashort X-ray pulses [6] [7] [8]. Those X-rays have been applied to observe ultrafast evolving diffraction patterns for organic films [6] and semiconductor crystals [4] [8] irradiated by the ultrashort laser pulses. It is expected that appropriate selection of a pump-pulse can induce and analyze various ultrafast phenomena such as lattice vibration and phase transition, which may result in experimental verification for the solid-state theoretical physics and molecular dynamics calculation.

Investigations of ultrafast phenomena such as pulse-radiolysis for radiation chemistry, and ultrafast laser-plasma interaction for laser acceleration have been one of the main topics at Nuclear Engineering Research Laboratory(NERL), University of Tokyo. In order for ultrashort X-ray generation, several experiments such as Thomson scattered X-ray generation

using the table-top tera-watt laser synchronized with the electron linear accelerators, and linac-based ultrashort X-ray generation with use of the subpicosecond electron linac [9] have been performed for the purpose of developing techniques to investigate ultrafast dynamics in materials [10]. Efforts have been also paid for the study of the laser produced plasma X-ray generation using the tera watt laser. In this study, we constructed the time-resolved X-ray diffraction system using the newly installed 12TW-50fs laser, and applied it to the dynamic analysis of GaAs(111) crystals in order to demonstrate performance of our system. The sample crystals were pumped by 50 fs laser pulses with fluence of 1.3 mJ/mm², and probed with the laser produced plasma X-rays from a copper target. Both the pump and probe pulses are produced from the same laser pulse in order to avoid the timing jitter problem during the repetitive data accumulation. We also report numerical results of the X-ray diffractions from the crystal with the laser-induced strain.

II. EXPERIMENT

Nowadays, ultraintense laser field more than 10^{17} W/cm² can be achievable by focusing a tera-watt laser. Interacting with matter, such an intense laser can produce hot plasmas that are sources of fast electrons. By decelerating these electrons in a solid target, X-ray emission such as bremsstrahlung and characteristic X-ray can occur. It is shown by the recent simulations [11] that under appropriate conditions the energy transfer from the intense femtosecond laser pulse into the target takes place in the time scale of the duration of the laser pulse. Therefore, the duration of the generated X-ray pulses can be ultrashort as well. Now we have a table-top tera-watt laser at NERL that can produce high power laser pulses with energy up to 600 mJ in 50 fs duration at 800 nm wavelength. We utilize it for the time-resolved X-ray diffraction experiments. A schematic view of the setup for the first experiment is shown in Fig.1. The laser pulse coming into the vacuum chamber is divided to the main pulse and the pump pulse in a ratio of nine to one by the beam splitter. The former is focused onto the copper target with the off-axis parabolic mirror (focal length =

162 mm) to generate the laser produced plasma X-rays that are used for X-ray diffraction from the single crystal as the probe-pulse. Focused by the spherical lens, the latter induces transient phenomena inside the crystal after passing through the delay-path with which the time interval between the pump- and probe- pulses can be controlled. For easy alignment, large wafers or disks of single crystals are employed as the samples for X-ray diffraction and mounted on the automatic positioner. The two-dimensional diffraction images are taken by X-ray imaging plates (BAS-SR, Fuji Photo Film, Tokyo, Japan) with the spatial resolution of 50 μm . The samples are surrounded by lead plates for shielding of the background X-rays scattered from the circumference. The X-rays may reach the sample surface only through the 3-mm-wide slit. Polyvinylidene chloride film is put in front of the parabolic mirror as a protection against ablated debris from the copper target. Another 1-cm-wide slit is also put at the exit of this window to cut the background X-rays from inside. Fig.2 shows the diffraction images and their horizontal profiles of X-ray intensities for GaAs wafer of (111) orientation taken with delay times of 0 ps in (a) and 150 ps in (b) with the laser fluence of 1.3 mJ/mm². Two lines on IP images in Fig.2 are composed of the characteristic X-rays, $K_{\alpha 1}$ and $K_{\alpha 2}$, of copper, and corresponding peaks can be seen in profiles. The accumulation time of these images was about 30 sec at 10 Hz repetition. The laser energy of 350 mJ per pulse was devoted to the X-ray generation. In Fig.2(b), we can see a subpeak besides the original peaks, which is due to deformation of the lattice of the GaAs crystal induced by the laser pulses. There is no subpeak in Fig.2(a). It means that the subpeak is not due to permanent changes induced by the pump-pulses but caused by transient ones. Changes of X-ray diffraction patterns as time are shown in Fig.3(a). At the delay time of 50 ps, in addition to two peaks that consist of $\text{Cu}K\alpha 1$ and $\text{Cu}K\alpha 2$, we can see the subpeak that results from the lattice deformation induced by the laser pulse irradiation. This subpeak appears on the side of smaller angles compared with the original Bragg angle, which implies lattice expansion inside the crystal. Then it goes closer to the main peak as time passes from 50 ps to 250 ps. An effect of the permanent changes caused by the laser into the crystal was observed in the X-ray diffraction patterns. After a few minutes irradiation, degradation

in the sample crystal was brought about, resulting in an irreversible change into the X-ray diffraction patterns. The crystals were translated on an automatic stage during the laser irradiation in order to avoid damage accumulation.

III. NUMERICAL CALCULATION

We also performed calculations of X-ray diffraction from laser irradiated crystal. Laser-induced strain derived by Thomsen et al. [12] was assumed and temporal evolution of X-ray diffraction patterns was calculated. The laser energy absorbed in the crystal makes the temperature increase distributed according to the absorption length ζ . Thus temperature increase ΔT can be described as

$$\Delta T(z) = \Delta T_s e^{z/\zeta}, \quad (1)$$

where z is the distance from the surface into the crystal and ΔT_s is the temperature increase at the surface just after the laser irradiation. This temperature distribution gives rise to a thermal stress given by

$$-3B\beta\Delta T(z), \quad (2)$$

where B is the bulk modulus and β is the linear expansion coefficient. Then thermal elastic equations are given by

$$\sigma_{33} = 3 \frac{1-\nu}{1+\nu} B\eta_{33} - 3B\beta\Delta T(z), \quad (3)$$

$$\rho \frac{\partial^2 u_3}{\partial t^2} = \frac{\partial \sigma_{33}}{\partial z}, \quad (4)$$

$$\eta_{33} = \frac{\partial u_3}{\partial z}, \quad (5)$$

where η_{33} is the elastic strain tensor, ν is Poisson's ratio, u_3 is the displacement in the z direction, and ρ is the density. The initial condition is that $\sigma(z) = 0$ everywhere. These equations can be solved analytically. The solution is

$$\eta_{33}(z, t) = \Delta T_s \frac{1 + \nu}{1 - \nu} \left[e^{-z/\zeta} \left(1 - \frac{1}{2} e^{vt/\zeta} \right) - \frac{1}{2} e^{-|z-vt|/\zeta} \text{sgn}(z - vt) \right], \quad (6)$$

where v is the sound velocity which is given by.

$$v = 3 \frac{1 - \nu B}{1 + \nu \rho} \quad (7)$$

This solution gives two components, i.e., a strain wave propagating inward with the speed of sound and a thermal expansion of the lattice at the surface. X-ray diffraction patterns were calculated with this strain based on the kinetic diffraction theory. The result of the calculation is shown in Fig.3(b). Here also we can see the subpeak that behaves in the same manner as the experimental one, as well as peaks of characteristic X-rays. In the calculation, we determined lattice expansion at the crystal surface by the shift of the subpeak observed in the experiment. ζ was also fixed to be $0.3 \mu\text{m}$ so as to give the best fit to the experimental result. For comparison, we also calculated X-ray profiles for Si(111) and Ge(111) as well as GaAs(111), assuming the maximum strain at the crystal surface as 0.3% . Fig.4 shows calculated profiles at 150 ps delay time. In the case of Si(111), we can see a subpeak which is more distant from the original peak and smaller than one for GaAs(111), while there is no subpeak in the case of Ge(111). The difference that appears here is attributed to different absorption lengths of laser light and different attenuation lengths of X-ray in those crystals. X-ray can penetrate more inside into Si than GaAs because of its longer attenuation length for Si, which means that the diffracted X-ray profile from Si includes information of deeper atoms. On the other hand, laser pulse can also propagate deeper into Si because of the longer absorption length, making deeper lattice strained. The ratio of the laser penetration depth to the X-ray penetration depth into the crystals will affect the ratio of the subpeak to the main peak in X-ray profiles. In the case of Ge, therefore, no clear subpeak can be seen because the laser penetration depth is too small compared to the X-ray penetration depth and because most diffracted X-rays come from less perturbed region of the lattice.

IV. CONCLUSION

We constructed the time-resolved X-ray diffraction system utilizing the laser produced plasma X-ray generated with the 12TW-50fs laser at Nuclear Engineering Research Laboratory, University of Tokyo. The system was applied to the dynamic analysis for the laser-irradiated GaAs(111). Picosecond temporal evolution of X-ray diffraction was demonstrated. Clear subpeak appeared in the X-ray profile by 1.3 mJ/mm^2 laser irradiation at 150 ps delay time. We also performed the calculation of X-ray diffraction from the laser-irradiated crystal and confirmed the consistency with the experiment. We plan to apply this technique to the dynamic analysis of other materials such as metals, ferroelectrics, and organic substances.

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FIGURES

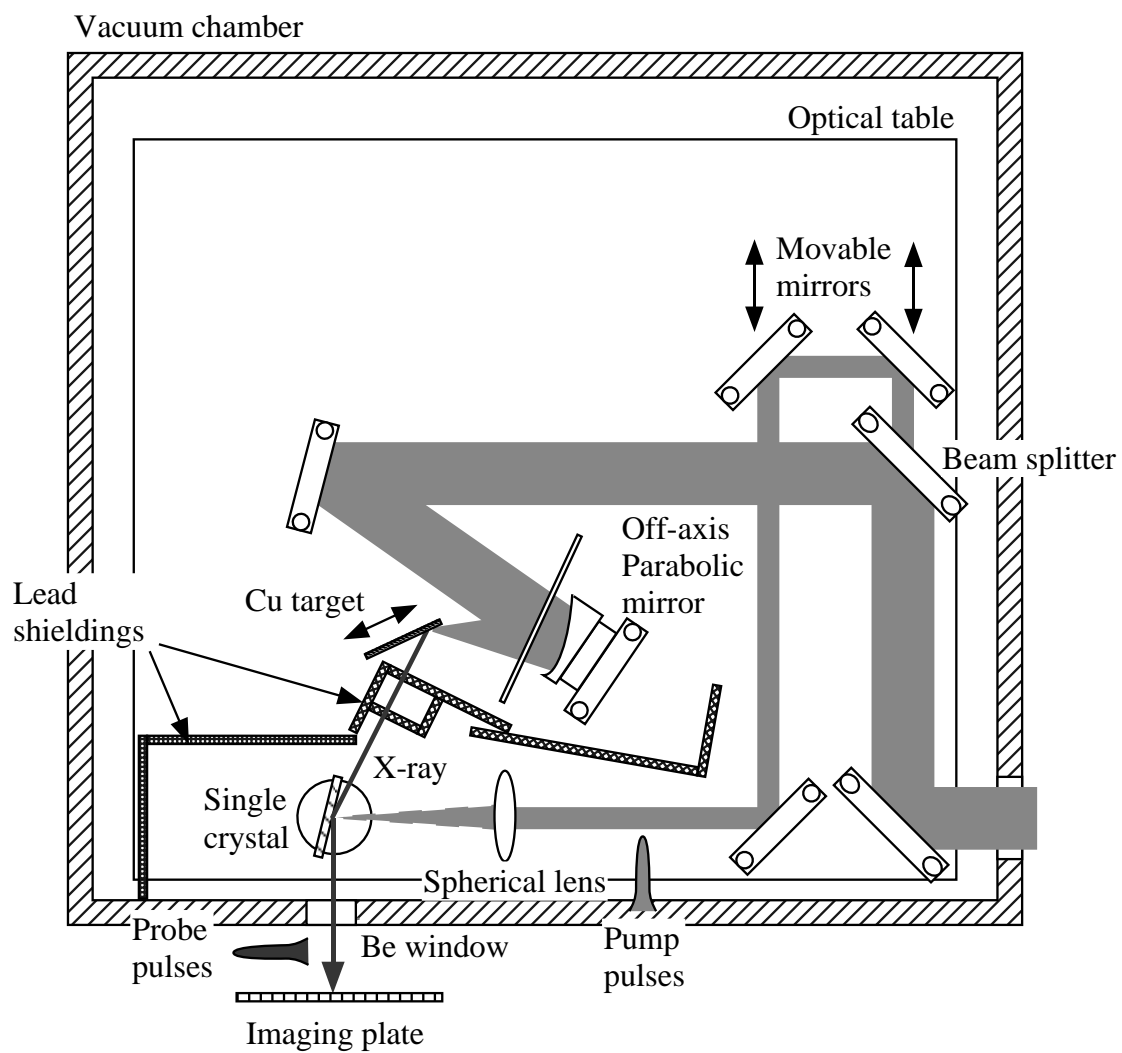


FIG. 1. Experimental setup for the time-resolved X-ray diffraction with 12TW-50fs laser.

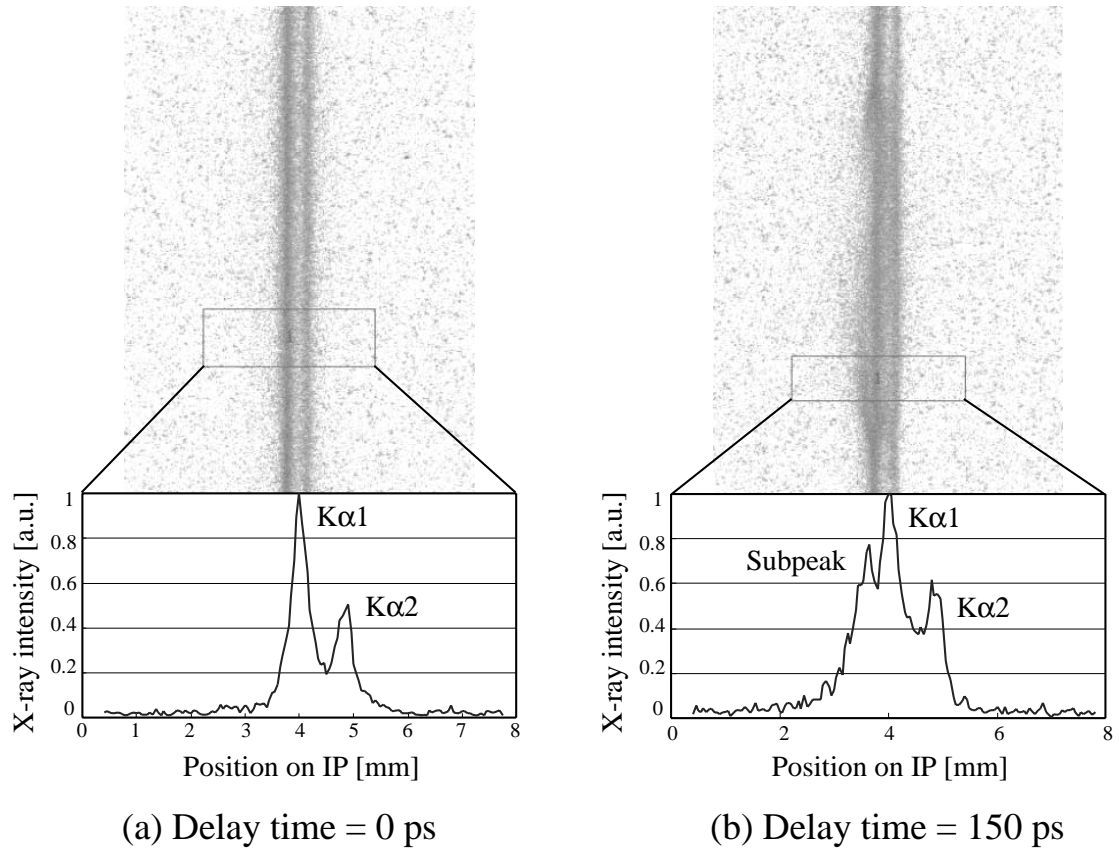


FIG. 2. Two dimensional images and horizontal profiles of diffracted X-rays from GaAs(111) surface. (a) Delay time between pump pulse and probe pulse is 0. (b) Delay time is 150 ps.

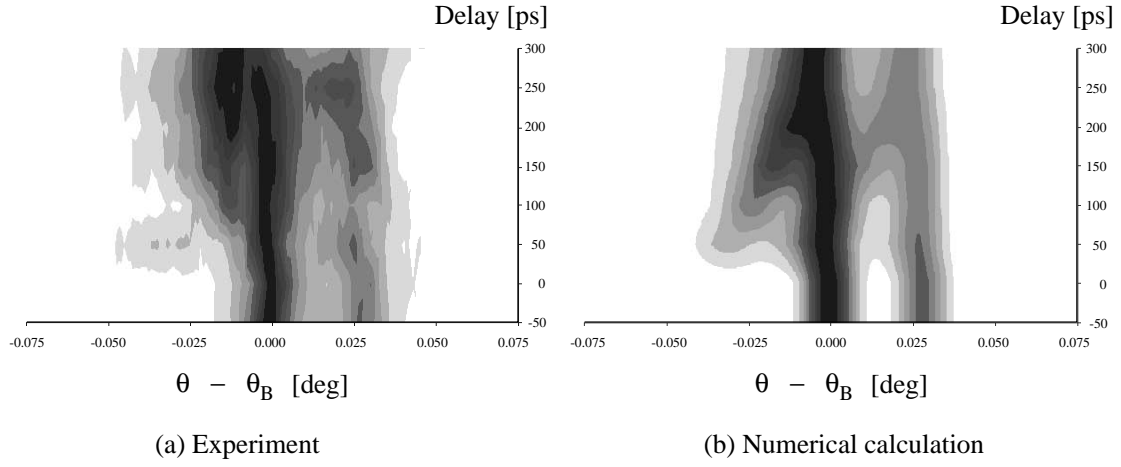


FIG. 3. Angular distributions of diffracted X-ray intensities at each time, shown as contour lines. (a) Experimental result. (b) Numerical calculation.

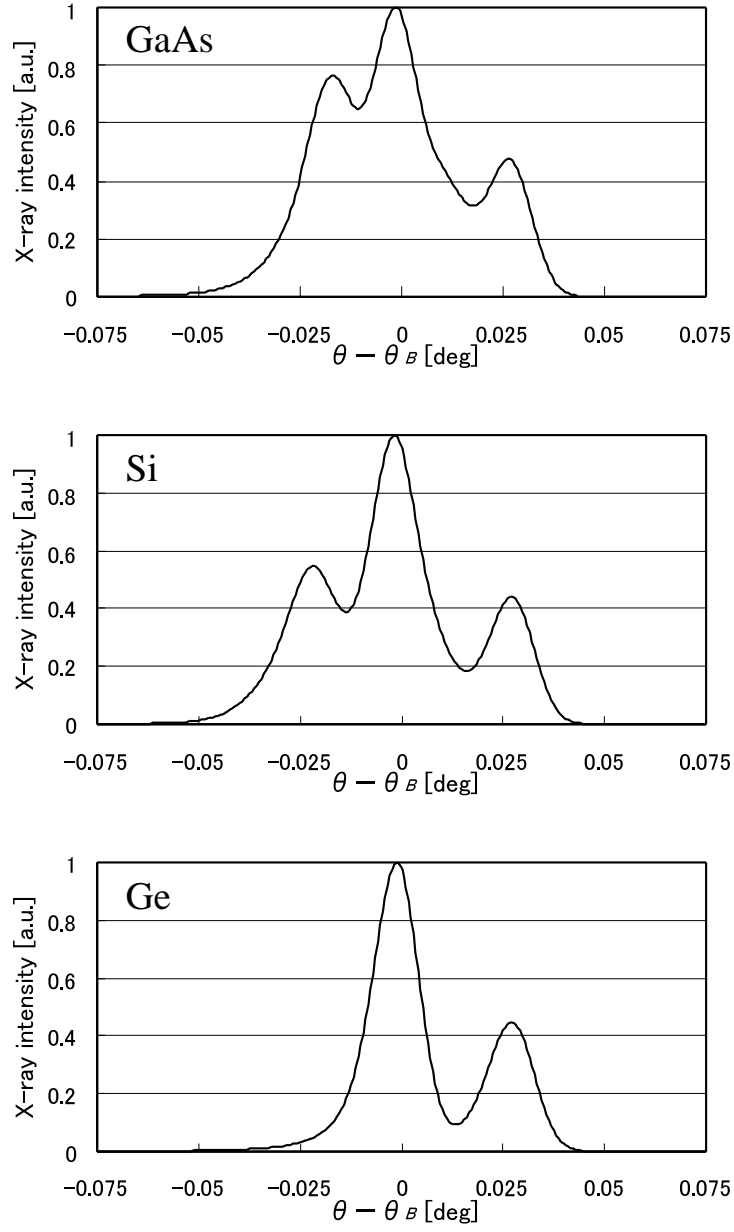


FIG. 4. Calculated X-ray diffraction intensities at the delay time of 150 ps. (a) GaAs(111). (b) Si(111). (c) Ge(111).