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To cite this article: Masaki Hada and Jiro Matsuo 2011 IOP Conf. Ser.: Mater. Sci. Eng. 24 012010

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Evaluation of lattice motion in CdTe single crystal using in-air tabletop time-resolved X-ray diffractometer

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Abstract. A bulk single crystal sample of cadmium telluride (CdTe) was irradiated with a ~3 mJ/cm² infrared femtosecond laser and analyzed with time-resolved X-ray diffraction (XRD) performed on an in-air tabletop designed for an extremely short pulsed X-ray source. The integrated intensity of Kα XRD lines from the CdTe (111) plane was decreased by 4%–5% in the time scale of ~200 ps. The changes in integrated intensity of the XRD line were induced by the thermal effect of the CdTe lattice propagating at the acoustic velocity in the crystal.

1. Introduction

X-ray diffraction (XRD) is one of the most suitable methods for direct measurement of the atomic positions in a crystal. Recently, time-resolved XRD experiments have been performed on semiconductors and semimetals using laser-plasma-produced X-ray sources, and the structural dynamics in the femtosecond-to-picosecond timescale were revealed [1–7]. These laser-produced plasma X-ray sources consisted of high-power and low-repetition-rate lasers of above 100 mJ and 10 Hz. However, the utilization of such ultrafast pulsed X-ray has been limited because of the complexity of the huge vacuum systems and difficulty in managing a high-power laser. Very recently, compactly designed tabletop sub-mJ to several mJ and kHz repetition rate femtosecond lasers for generating hard X-ray in air with sufficient X-ray photon intensity were reported [8,9]. The tabletop X-ray sources in air reduce not only the cost of the time-resolved X-ray diffractometer, but also make it easier to develop and perform the time-resolved XRD measurements. In this study, we characterized the photoexcited thermal process in a cadmium telluride (CdTe) single crystal using the tabletop in-air time-resolved X-ray diffractometer. CdTe is a suitable material for the time-resolved XRD because the penetrating depth of the probing X-ray and exciting infrared light are almost on the same order. CdTe is also a promising material for solar cells, and its photoelectric and photothermal properties have been studied extensively [5,6,10-12]. This work is a first experiment in applying the extremely short pulsed in-air X-ray source with a tabletop laser to time-resolved XRD measurements. The high-intensity in-air X-ray source with a tabletop laser could be a desirable tool for laboratory-based ultrafast time-resolved measurements.

2. Experimental Setup

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Time-resolved X-ray diffraction measurements were performed in air using laser-plasma-induced X-ray source of Cu Kα radiation at 1 kHz, generated by focusing a millijoule femtosecond laser onto a rotating copper target in helium ambient. The experimental setup for the time-resolved XRD is shown in Fig. 1. The femtosecond optical pulses at a wavelength of 800 nm generated from the mode-rocked Ti: sapphire laser (Spectra Physics / Tsunami) and regenerative amplifier (Spectra Physics / Model Spitfire Pro XP) were split into pump pulse and probe pulse with a polarized beam splitter. The contrast of the pump and probe pulse intensity was changed continuously from 3:7 to 1:9 by rotating the polarization with a λ/2 waveplate placed before the beam splitter. The probe pulse at laser intensity of ~2 mJ/pulse was focused onto the rotating copper target with an infrared achromatic lens (f = 40 mm), as shown in detail elsewhere [8,9]. The probing X-ray radiation emitted over a range of 2π steradians was collimated using slits of 0.3 mm width and 1 mm height, and was focused on a CdTe single crystal. The X-ray incident angle was θ=11.88° at the symmetrical Bragg diffraction geometry. The pulse width of the pulsed X-ray was estimated to be about 200 fs, and the time-resolution was about 1 ps, determined by considering the geometry of the X-ray divergence angle. The diffracted X-ray at the sample was detected using a charge coupled device (CCD) camera. The X-ray intensity was monitored with a p-intrinsic-n (PIN) Si photodiode. The pump pulse passed through an optical delay line and was focused onto a CdTe single crystal at a laser intensity of about 3 mJ/cm². The area excited by the pump pulse was 1.7 mm in diameters and was larger than the X-ray-probed area.

![Experimental Setup for the time-resolved X-ray diffraction in air](image)

3. Results and Discussion
The Kα₁ and Kα₂ X-ray diffraction lines from CdTe (111) plane are shown in Fig. 2 (a); and a clear image with sufficient signal-to-noise ratio was obtained. Figure 2 (b) shows the changes in the integrated intensity of Kα X-ray diffraction lines as a function of the delay time. As shown in Fig. 2 (b), the intensity deceased by 4%-5% in the time scale of ~200 ps, consistent with the previously reported lattice dynamics in CdTe single crystal with time-resolved X-ray diffraction [6]. The probing depth of the CdTe crystal was about 0.68 µm, which is mentioned in the following passage. Therefore, the photo-excitation has propagated into the CdTe crystal at a velocity of about 3400 m/s, which corresponds well to the longitudinal acoustic velocity in CdTe crystal (3440 m/s) [13].

The exciting depth and probing for the CdTe crystal with laser and X-ray were calculated with the extinction coefficient and mass attenuation factor. The laser intensity, Iₙ, after it passes a distance x in a medium is equal to:

\[ I_x = I_{i0} \exp(-\alpha x) \],

where \( I_{i0} \) is the intensity at \( x=0 \) and \( \alpha \) is called the absorption coefficient. The 1/e penetrating depth can
be defined as $1/\alpha$ from eq. (1). For many applications, the extension coefficient, $k$, which is equal to

$$ k = \alpha \frac{\lambda}{4\pi} , $$

where $\lambda$ is the laser wavelength, is more commonly used for characterization of the electromagnetic loss in materials. The refractive index $(n - ik)$ of the CdTe is $2.877 - 0.065i$ at the wavelength of 800 nm [14]; therefore, the $1/e$ penetrating depth of the infrared light is calculated to be about 0.9 µm with an incident angle of 45°. The linear absorption coefficient of CdTe is $1.42 \times 10^3$ cm$^{-1}$ for an 8 keV X-ray; therefore, the $1/e$ penetrating depth of the X-ray into the CdTe crystal with an incident angle of 11.88° is 0.68 µm [14].

In the thermal process, the change in diffraction line is attributed to the Debye–Waller effect. The intensity of diffraction lines ($I$) at a temperature $T$ is [15]

$$ I(T) = I(0) \exp \left[ -2 \cdot 2\pi^2 K^2 U(T) \right] = I(T_R) \exp \left[ -2 \cdot 2\pi^2 K^2 (U(T) - U(T_R)) \right] $$

where $K$ is scattering vector, $T_R$ is the room temperature, and $U(T)$ is the mean-square displacement parameter. The displacement parameter at the temperature $T$ assumes the thermal vibration as an isotropic harmonic oscillator. The energy of the vibration ($E$) in a crystal at a given temperature ($T$) can be described with the Maxwell-Boltzmann distribution as:

$$ f_{MB}(E) = e^{-E/k_B T} $$

where $k_B$ is the Boltzmann constant. Therefore, the distribution ($x$) of the atomic vibration in the crystal can be described with the following Gaussian distribution:

$$ f_{MB}(x) = \exp \left( -\frac{kx^2}{2k_B T} \right) $$

where $k$ is the force constant in an elastic body and is calculated with the linear compressibility ($B$) and the average atomic distance ($a_0$) of the material as:

$$ k = \frac{a_0}{B} . $$

The standard deviation of the distribution corresponds to the displacement by the thermal vibration:

$$ U(T) \sim \frac{k_B T}{k} . $$

The linear compressibility ($B$) and the average atomic distance ($a_0$) of the CdTe crystal is $7.41 \times 10^{-12}$ Pa$^{-1}$ and 2.81 Å, respectively [16]. Thus, thermal displacement at $T = 293$ K (room temperature) was calculated to be ~0.103 Å. The intensity loss of 4%-5% was corresponded to an isotropic thermal displacement of ~0.165 Å at a temperature of about 700 K. The CdTe crystal would be oxidized and significantly damaged when heated repeatedly to about 700 K in air. However, after the multiple
excitations, the CdTe crystal had no degradation on the surface [17,18]. Moreover, the optical pulse at an intensity of ~3 mJ/cm$^2$ induced a temperature increase in the CdTe crystal of 100 K at most. When the thermal vibration mode along the [111] direction was selectively excited with optical pulses, the thermal displacement caused by the temperature increase of about 100 K was enhanced to be about 0.147 Å in the [111] direction, which in the CdTe crystal corresponds to the longitudinal direction of thermal propagation. On the picosecond timescale, a longitudinal optical (LO) phonon will be excited selectively and an LO phonon will be coupled with a longitudinal acoustic (LA) phonon and expand the thermal effect into the depth area of the crystal [7,19]. The intensity loss with the anisotropic thermal displacement was calculated to be ~3% and agreed with the relative magnitudes of the experimental results. Thus, an anisotropic thermal vibration would have occurred in the CdTe crystal in the picosecond timescale and the thermal effect would propagate into the depth area of the crystal with acoustic velocity.

4. Conclusion

Time-resolved X-ray diffraction was performed on a CdTe (111) single crystal in air using a tabletop laser-plasma-induced X-ray source. The integrated intensity of Kα X-ray diffraction lines decreased by 4%–5% in the timescale of ~200 ps. The intensity decrease was consistent with anisotropically excited thermal vibrations calculated with the Debye–Waller factor, and the propagation time was agreed well with the acoustic velocity in the CdTe crystal. Thus, an X-ray source in air with a tabletop femtosecond laser would be a desirable tool for laboratory-based ultrafast time-resolved measurements. Further experiments will focus on varying the intensity of the photo excitation to reveal in more detail the thermal effect and thermal propagation in the CdTe crystal.

Acknowledgements

This work is supported by JST-CREST and the Kyoto University GCOE program. We would like to thank Dr. Rafael Manory of editassociates.com for help with preparing this material for publication.

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