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Femtosecond time-resolved x-ray diffraction from optical coherent phonons in CdTe(111) crystal

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Coherent phonons excited in a CdTe(111) crystal by 70 fs laser irradiation have been investigated by femtosecond time-resolved x-ray diffraction. The longitudinal optical phonon with a frequency of approximately 5 THz near the Brillouin zone center has been detected as modulation in intensities of x-ray diffraction. Atomic displacement in the [111] direction in the coherent longitudinal optical phonon has been estimated. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2968212]

Coherent phonons can be excited in solids by irradiation of laser pulses whose duration is sufficiently shorter than the vibrational period[1] and receive recently much attention because of possible application to coherent control of phase transitions. Excitation and relaxation of coherent phonons have been extensively investigated by using optical measurements (reflectivity and transmission) in various materials such as semimetals,[2,3] semiconductors,[4,5] and dielectrics.[6] However, changes of the crystal structure associated with coherent phonons have not been observed because the optical measurements are sensitive to susceptibility due to valence electrons and give indirect information for atomic positions.

X-ray diffraction is the most suitable technique to measure directly the atomic positions and crystal structures. Recently, ultrafast time-resolved x-ray diffraction using short-pulsed x rays has been performed on the laser- excited semiconductors and atomic motions associated with the acoustic phonon propagation are directly observed as the changes in diffraction angles.[7–11] However, atomic motions in optical phonons cannot be detected as the change in diffraction angles because the optical phonons do not change barycentric positions of the crystal lattice. Recently, Sokolowski-Tinten *et al.*[12] demonstrated that the coherent optical phonon can be detected as a modulation in the intensity of femtoseond time-resolved x-ray diffraction. The optical phonon (2.12 THz) in the semimetal nanolayer (50-nm-thick bismuth) has been reported. Several observations of optical coherent phonons of Bi using time-resolved x-ray diffraction are reported with x rays from both the laser plasma and an accelerator.[12–14] Optical coherent phonon oscillations are expected to be used as a monitor for synchronization of a femtosecond-laser pulse and ultrashort x-ray pulse generated from an x-ray free electron laser for laser-pump and x-ray probe measurements. We expect this technique to be applicable to other materials such as semiconductors and bulk materials. Changes in atomic positions and crystal structure associated with coherent phonons and transition from optical phonons to acoustic phonons can be revealed by using femtosecond time-resolved x-ray diffraction.

In this paper, we performed the femtosecond time-resolved x-ray diffraction on the 70 fs laser irradiated bulk sample of a semiconductor (CdTe) single crystal with a time step of 27 fs and detected the coherent longitudinal optical (LO) phonon (5 THz) at Brillouin-zone center. The coherent LO phonon is also confirmed by using an optical reflectivity measurement. We also describe how the optical coherent phonon affects x-ray diffraction intensities.

The changes in the diffraction intensity $I(h,k,l,t)$ caused by atomic motion in the unit cell are determined by the square modulus of the structure factor, where h, k, and l are Miller indices and t is time. The structure factor is expressed by

$$F(h,k,l,t) = \sum_{j=1}^{N} f_j \exp(-i\vec{G}_j \cdot (\vec{r}_j + \vec{\delta}_j)), \tag{1}$$

$$\vec{\delta}_j = \vec{u}_j \sin(\omega t + \varphi_j), \tag{2}$$

where f_j, $\vec{r}_j$, and $\vec{\delta}_j$ are the atomic scattering factor, the crystallographic atomic position, and the atomic-deviation vector of the jth atom in the unit cell, respectively. $\vec{G}$ is the reciprocal lattice vector and N is the number of atom in the unit cell. The $\vec{u}_j$ and φ_j are the maximum deviation vector and initial phase of the jth atom. In a coherent phonon, all vibrations are in phase.

The time dependence of the structure factor can be simply expressed in the first order approximation for the center symmetric atomic vibrations as follows:

$$F(h,k,l,t) \approx F_0 - i\left\{\sum_{j=1}^{N} f_j \vec{G}_j \vec{u}_j \exp(-i\vec{G}_j \vec{r}_j)\right\} \sin(\omega t), \tag{3}$$

where F_0 is the time-independent structural factor and initial phase is set to be zero. Because the diffraction intensity is proportional to the square modulus of the structure factor, the

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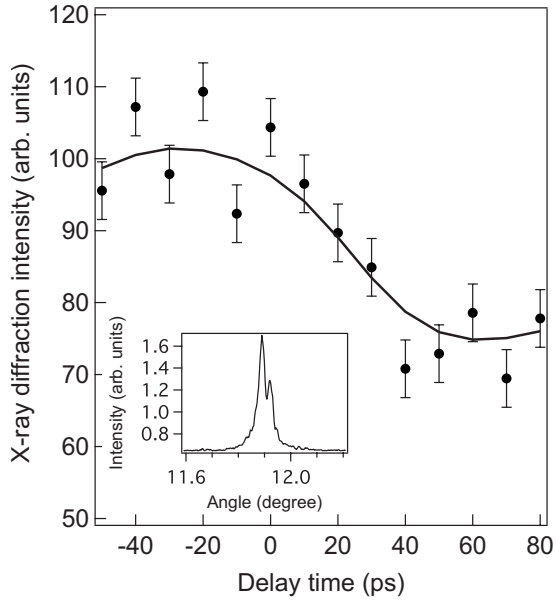

FIG. 1. Temporal change of intensities of x-ray diffraction from CdTe with (111) axis as a function of delay time (10 ps time step). The inset shows a rocking curve from CdTe(111) before the laser irradiation.

oscillation in diffraction intensities with frequencies of ω and 2ω is easily understood from Eq. (3). Since Eq. (3) has a term of inner product of the reciprocal vector and atomic deviation vector, only the longitudinal phonon mode can be detected.

Ultrafast time-resolved x-ray diffraction was performed on the 70 fs laser (wavelength centered at 790 nm) irradiated at a CdTe(111) crystal using laser-plasma x-ray pulses and a laser-pump and x-ray-probe technique. X-ray pulses of Cu K_α lines ($\sim$0.154 nm) were generated by focusing 70 fs laser beam onto Cu tape target (30 μm) at an intensity of 10^{17} W/cm^2 (70 kJ/cm^2). Diffracted x rays were detected by an x-ray charge coupled device camera at the symmetrical Bragg diffraction geometry (around 11.89 deg of diffraction angle). The sample used was a CdTe single crystal (30 $\times$30 mm^2 and 2 mm thickness). The CdTe(111) crystal was excited by 70 fs laser with the power density of 0.6 mJ/cm^2. Half of x-ray diffracted area on the CdTe surface was shielded from the laser irradiation to record both the x-ray diffracted signals from the laser perturbed and unperturbed areas at the same time.

The x-ray diffracted intensity from the laser perturbed area was normalized by the signal from the unperturbed area. At each delay time, the signal was accumulated by 400 times. The x-ray measured depth in CdTe with the x rays of Cu K_α is approximately 0.68 μm at the Bragg angle of 11.89 deg and the absorption depth of CdTe with a photon energy of 1.5 eV(=800 nm) is approximately 0.5 μm for normal incidence. Details of the experimental setup are described else where.[15,16] The energy density of the pump laser beam is kept to be low enough to avoid permanent damage of the sample.

Cu $K_{\alpha1}$ and $K_{\alpha2}$ lines were found in the x-ray diffraction from the pristine sample at 11.89 and 11.92 deg, respectively (Fig. 1 inset). Time-resolved x-ray diffraction, obtained with a time step of 10 ps, showed sudden decrease in the diffracted intensity when the 70 fs laser was irradiated on CdTe(111) at a laser-power density of 0.6 mJ/cm^2, as shown

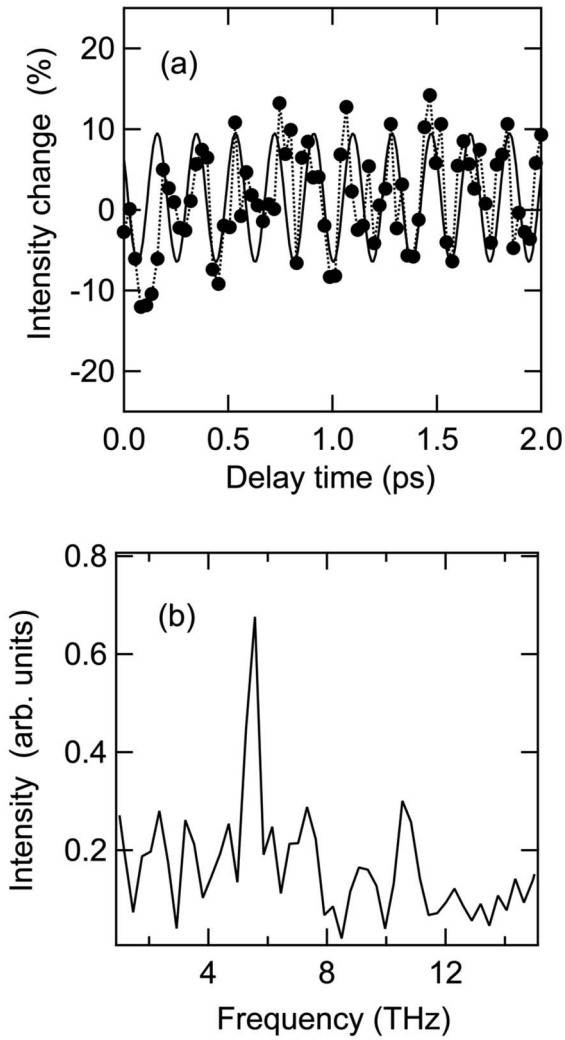

FIG. 2. Oscillation in x-ray diffraction intensity from the 70 fs laser irradiated CdTe(111) as a function of delay time (a). Circles and dotted lines are experimental results and the solid curve is oscillation with a frequency of 5.3 THz as a reference. Data have errors of $\pm$5%. Fourier power spectrum of the obtained data (b).

in Fig. 1. Time zero for the arrivals of both the laser beam and x rays is set at time when the integrated intensity began to decrease. No shift in the rocking curve was observed within delay times below 10 ps. The decrease in intensities of x-ray diffraction is due to thermal expansion, which is much slower than the non thermal melting.[10] The observed time scale (a few tens of pocoseconds) is reasonable for lattice heating, which should follow the slow electronic relaxation in CdTe.[17–19] Error of the data was estimated from deviation of diffracted x-ray intensities of 20 data sets.

At around the time zero, femtosecond time-resolved x-ray diffraction was examined. The delay time was controlled by changing the optical path length for the pumping laser beam with a step of 8 μm, which corresponds to 27 fs. A modulation was observed in the diffraction intensities, as shown in Fig. 2. The Fourier power spectrum of the modulation of diffracted intensities shows a distinct peak at 5.3 $\pm$ 0.4 THz. The obtained frequency is consistent with the LO phonon of CdTe at the Brillouin zone center.[20] The coherent LO phonon of CdTe has been measured using terahertz radiation from a 70 fs laser irradiated sample and its frequency has been obtained to be 5.1 $\pm$ 0.05 THz.[21] The fre-

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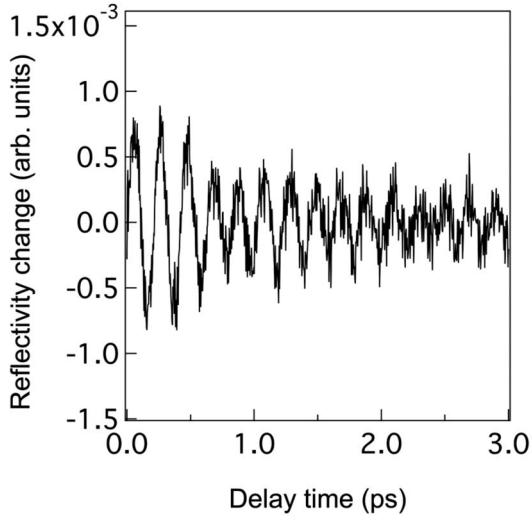

FIG. 3. Femtosecond time-resolved reflectivity measurement from the 80 fs laser irradiated CdTe(111) as a function of delay time.

quency obtained from time-resolved x-ray diffraction agrees with that obtained form the optical measurement within errors. Atomic displacement from the equilibrium position was estimated to be 0.8% of the distance of lattice planes in the [111] direction from the amplitude of the change in diffracted x-ray intensities.

We also studied coherent phonons by using femtosecond time-resolved reflectivity measurements on the CdTe(111) single crystal. Details of the experiments are described else where.[22] The observed time-domain spectrum is shown in Fig. 3. The value of the reflectivity change was estimated to be of the order of 10^{-6}. It shows clear damping oscillation (LO phonon) with a frequency of 5 THz. The results confirm that the oscillation obtained by time-resolved x-ray diffraction is due to LO phonon of CdTe.

The observation of 200 fs cycle oscillation in x-ray diffraction intensities suggests that the x-ray pulse width is shorter than 200 fs. There is no angular shift in rocking curves within 10 ps.

Feurer *et al.*[10] a performed time-resolved x-ray diffraction on the 80 fs laser irradiated thin layer of a CdTe crystal with a time step of approximately 140 fs. They observed a decrease in x-ray reflectivity according to modification of crystal structure due to nonthermal melting but did not observe the coherent phonon oscillation. This is because of their long time step compared with the phonon period and partly because of the permanent damage induced by their intense laser irradiation.

In conclusion, we performed femtosecond time-resolved x-ray diffraction experiment for laser-excited CdTe single crystal and successfully measured signal of coherent LO phonon (approximately 5 THz) near the Brillouin zone center in the [111] direction. It is shown that the optical phonon is detected as a modulation in intensities of x-ray diffraction even for bulk materials and semiconductors. The observation of 200 fs cycle oscillation in x-ray diffraction intensities suggests that the x-ray pulse width is shorter than 200 fs. Lattice deformation due to thermal expansion and acoustic phonons has not been observed within 10 ps. Femtosecond time-resolved x-ray diffraction during much longer time may reveal a whole dynamics of transition from optical phonons to acoustic phonons.

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