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Time-resolved Bragg coherent X-ray diffraction revealing ultrafast lattice dynamics in nano-thickness crystal layer using X-ray free electron laser

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Ultrafast time-resolved Bragg coherent X-ray diffraction (CXD) has been performed to investigate lattice dynamics in a thin crystal layer with a nanoscale thickness by using a SASE (Self-Amplified Spontaneous Emission)–XFEL (X-ray Free Electron Laser) facility, SACLA. Single-shot Bragg coherent diffraction patterns of a 100 nm-thick silicon crystal were measured in the asymmetric configuration with a grazing exit using an area detector. The measured coherent diffraction patterns showed fringes extending in the surface normal direction. By using an optical femtosecond laser-pump and the XFEL-probe, a transient broadening of coherent diffraction pattern profile was observed at a delay time of around a few tens of picosecond, indicating transient crystal lattice fluctuation induced by the optical laser. A perspective application of the time-resolved Bragg CXD method to investigate small sized grains composing ceramic materials is discussed.

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Key-words : Coherent X-ray diffraction, Time-resolved measurement, X-ray free electron laser, Semiconductor, Nanocrystal

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1. Introduction

Time-resolved (TR) X-ray diffraction (XRD) is a powerful method to reveal dynamical property of lattice motion in crystalline materials. TR-XRD, as the name suggests, combines time-resolved measurement and X-ray diffraction. Time-resolved measurement technique mainly includes the pump–probe method and fast detection systems with a short exposure time. The pump–probe method is usually used especially for phenomena with a response time faster than the electronic circuit speed of nanoseconds. For the pump–probe method, short pulsed X-ray sources are indispensable for probe beams, together with synchronized optical sources for excitation of target samples.

Recently developed accelerator-based pulsed X-ray sources have intensively contributed to the pump–probe experiment to study fast structural dynamics in materials. At synchrotron radiation (SR) facilities, the pulsed time structure of X-ray beam from a storage ring enables us to make a stroboscopic measurement with a time-resolution of a few tens of picoseconds to one nanosecond in FWHM. We have developed an ultrafast measurement system with optical laser-pump and X-ray probe system in a 3rd generation SR facility, SPring-8,[1)] and demonstrated time-resolved X-ray diffraction measurements. For phase change materials such as $Ge_2Sb_2Te_5$ and AgInSbTe, which are used in DVD optical recording devices, it was found that the amorphous-to-crystal phase changes occur in about 100 ns.[2)] As for the investigation of laser-induced lattice dynamics in semiconductor single crystals using optical laser-pump and SR X-ray probe method,[3)] we have observed acoustic pulses generation[4)] and the echoes in silicon and gallium arsenide.[5)] For faster lattice dynamics with sub-picosecond time-resolution, optical phonons[6),7)] and ultrafast melting[8)] have been observed using linac-based SR sources (at SPPS in LCLS) and optical laser slicing in the storage ring (SLS).

In 2009, the world's first SASE (self-amplified spontaneous emission)–XFEL (X-ray free electron laser) was generated at LCLS in Stanford, USA.[9)] Intense femtosecond pulses of SASE-XFEL offer a new opportunity to conduct femtosecond time-resolved X-ray diffraction. In 2011, the world's second SASE–XFEL facility named SACLA (SPring-8 Angstrom Compact free electron LAser) was also completed in the SPring-8 campus.[10)] SACLA produces intense hard X-ray pulses with a pulse duration of a few tens of femtoseconds and a shorter wavelength up to 0.0634 nm. The intense X-ray pulses are also useful to investi gate irreversible processes, through a single-shot pump–probe measurement,[11)] as well as to the investigation of small sized samples. Furthermore, SASE-XFELs have nearly full transverse coherence. This property offers the opportunity for coherent X-

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ray diffraction (CXD), which is a powerful lens-less imaging tool with nanometer–scale resolution. When CXD method is combined with TR-XRD, dynamical properties of small-sized crystal grains contained in ceramics may be investigated to reveal important features of ceramics, such as toughness and breaking mechanism.

We performed TR-Bragg CXD measurement using SACLA. Because of SACLA's shorter wavelength with a pulse duration of a few tens of femtosecond, high Q (transfer wavevector) scattering data provided us information on ultrafast atomic-scale motion, i.e., ultrafast lattice dynamics of nanometer-sized crystals. X-rays can also be used for in situ measurement in the atmosphere, which will enable us to observe, e.g., the ceramics under destroying the crystalline phase.

In this paper, we describe the result of our experiment with optical laser pump and XFEL probe for a silicon thin crystal, which is known as a silicon-on-insulator (SOI) wafer. We also discuss the potentials of TR-Bragg CXD to investigate nano-sized crystal grain in ceramics.

2. Experimental

Figure 1 illustrates the experimental setup of ultrafast TR Bragg CXD with NIR (near infrared) pump and XFEL probe for an SOI sample performed at a SACLA beamline of BL3. Asymmetric diffraction experiments were performed in air. The SACLA XFEL was operated at a 10 Hz repetition rate with a duration of 10–30 fs[12] with a typical photon flux of 5×10^9 after a double crystal monochromator. The unfocused beam was guided on to the sample with a beam size of 200 μm in diameter. The photon energy was adjusted to 8.69 keV by tuning the accelerated electron energy and the undulator gap, in order to satisfy the diffraction geometry where the diffracted beam direction $\mathbf{k}_o$ is almost perpendicular to the surface normal, $\mathbf{n}$, of the silicon thin crystal as shown in **Fig. 2**. The SOI wafer is composed of 100 nm-thick Si(100) single crystal attached on 50 nm-thick SiO_2 layer and 1 mm-thick substrate of SiO_2. The configuration enables us to observe the coherent diffraction pattern extending in $\mathbf{n}_{100}$ direction with an area detector without need for rotating the sample angle.[4] Thus, lattice spacing and layer thickness along the surface normal are detectable by single-shot measurement. In the experiment, the incident X-ray wavevector, $\mathbf{k}_i$, satisfies the relation of $\mathbf{k}_o = \mathbf{k}_i + \mathbf{G}_{311}$, and the angle between $\mathbf{k}_o$ and $\mathbf{n}_{100}$ is around (90.0–0.5)°.

The Ti:sapphire pump laser system equipped with a chirped amplifier was operated at a repetition rate of 10 Hz. The wavelength and the pulse energy were set to 800 nm (NIR) and 2 mJ, respectively. The pump beam size is adjusted to be 1.5 mm (FWHM) on the sample. Both the XFEL and the Ti:sapphire laser beams were arranged to coincide at the sample surface with less than an accuracy of a few tens of microns. Timing between the pump and probe lasers was controlled using electronic circuit system and optical delay system. The initial delay time was tuned using a fluorescent reference sample, which was monitored by a fast photodiode and a 13 GHz oscilloscope.

A flat panel detector (Hamamatsu Co.) with a pixel size of $50 \times 50\,\mu m^2$ was mounted at a distance of 853 mm from the sample with a glancing angle of 52°. Mechanical shutters for XFEL and Ti:sapphire laser were controlled to pass a couple of shots with an exposure time of 333 ms. The exposure timing is also synchronized with the mechanical shutters with a repetition rate lower than 1 Hz. Each coherent diffraction pattern from the 311-reflection was then recorded with setting a delay time between XFEL and Ti:sapphire laser pulses.

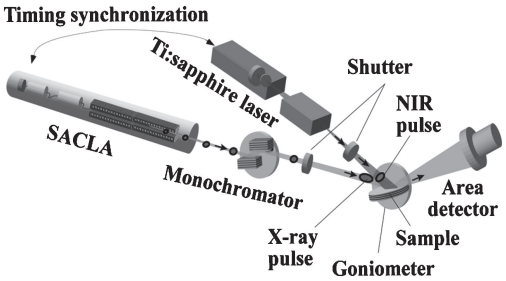

Fig. 1. Experimental setup of ultrafast TR Bragg CXD with Ti:sapphire laser pump- XFEL probe.

3. Results and discussion

3.1 Single-shot CXD pattern

Figure 3 shows a single-shot CXD pattern of the thin silicon crystal layer of SOI in the asymmetric configuration without the pump laser irradiation. Although SASE-XFELs have an intrinsic pulse-to-pulse fluctuation in the beam intensity and the direction,

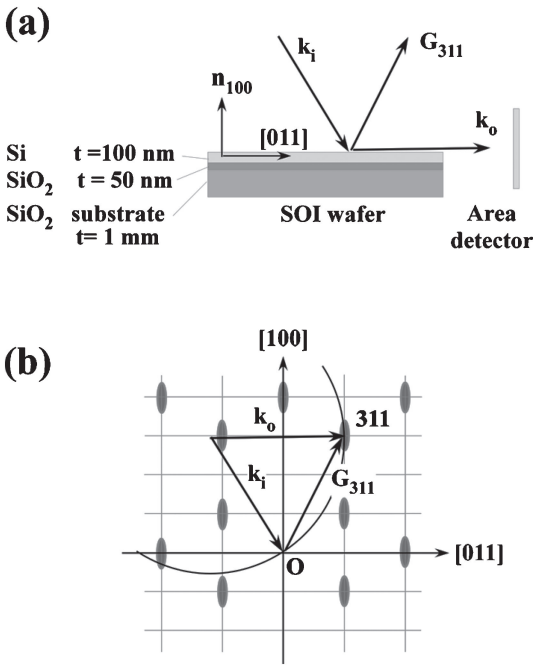

Fig. 2. (a) The structure of a thin wafer of Si (100) single crystal of SOI sample, and the geometry for the single shot CXD measurement. (b) The corresponding geometry in the reciprocal space.

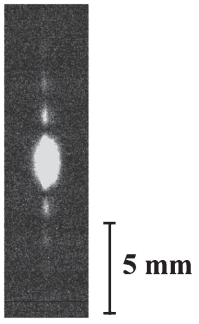

Fig. 3. Femtosecond single-shot diffraction pattern from a 100 nm thick SOI wafer with an X-ray free electron laser, SACLA.

the single-shot patterns such as Fig. 3 were obtained with a rate of about 1/10. The vertically broadened signal intensity can be attributed to the intensity distribution of crystal truncation rod. Here the CXD pattern in Fig. 3 is shown with a high sensitivity, where the diffraction intensity around the center of the Bragg spot is completely saturated, in order to show the fringe pattern in the vertical direction. The vertical fringe pattern provides the crystal lattice information in the (100) direction. In our experiment, the sample is illuminated by coherent waves, so the coherence length exceeds the thickness of the crystal. In this case, the vertical fringe pattern represents the squared Fourier transform of the outline shape of the crystal in the surface normal direction. As the atomic density has a rectangular distribution along the surface normal direction for SOI, the coherent diffraction profile has a form of a square of sinc function. The thickness of the crystal estimated from the interval of the fringe is about 100 nm.

3.2 Pump–probe measurement

The SOI surface was then irradiated by the Ti:sapphire laser with a power density of 115 mJ/cm^2. Although hundreds of the Ti:sapphire laser shots made damages on the sample surface, as they were found with an optical microscope, several tens of the laser shots caused no obvious damage. We therefore performed single-shot pump–probe measurements at the same illuminated position on the sample for 28 times for each delay time. The position of the sample with an area of 20 mm by 10 mm is then scanned by a step size of about 1 mm. As described in Sec. 3.1, pulse-to-pulse fluctuation of the XFEL intensity and the direction is not negligible. We therefore accumulated the CXD data of 28 shots with recording the position and the intensity for each incident X-ray pulse by a beam position monitor. The fluctuation of beam position at around the sample was about 100 μm by 100 μm. The diffraction profiles for which the X-ray beam-illuminated positions lie within 10 μm by 10 μm were then picked up and are shown in **Fig. 4**. Figure 4(a) shows a CXD snapshot after the laser shot with an interval, τ, of 20 ps. For comparison to Fig. 4(a), the CXD snapshots obtained before laser irradiation, at almost the coincident condition, and after the long time interval, were obtained and shown in Fig. 4(b) $\tau = -20$ ps, (c) $\tau = 0$ ps, and (d) $\tau \gg 1$ s (denoted by ∞), respectively. It is seen that the CXD pattern at (a) $\tau = 20$ ps is different from the others: the profile seems slightly broadened in the vertical direction corresponding to the (100) surface normal direction of the Si thin crystal layer. The widths of CXD pattern with respect to the (100) direction were thus obtained by fitting with a Gaussian distribution function in order to confirm the broadening suggested above. From the fitted widths and the distance between the sample and detector, the divergence angles of the diffracted beams are evaluated and shown in the bottom of Fig. 4. It is clearly found that the divergence of the diffracted beam at (a) $\tau = 20$ ps is larger by about 13% in comparison with (b), (c), and (d). The result indicates transient crystal lattice fluctuation induced by the femtosecond optical laser irradiation. Our findings that the lattice fluctuation occurs around 20 ps are consistent with previous studies on semiconductors,[3] while the sample thickness is different from the SOI. Further measurement with fine time delay scan will reveal the response speed and the life time of this phenomenon. The TR-Bragg CXD, in addition, can provide information on propagation of melting or lattice fluctuation through the further analysis of the fine profile which is, for example, the fringes as seen in Fig. 3.

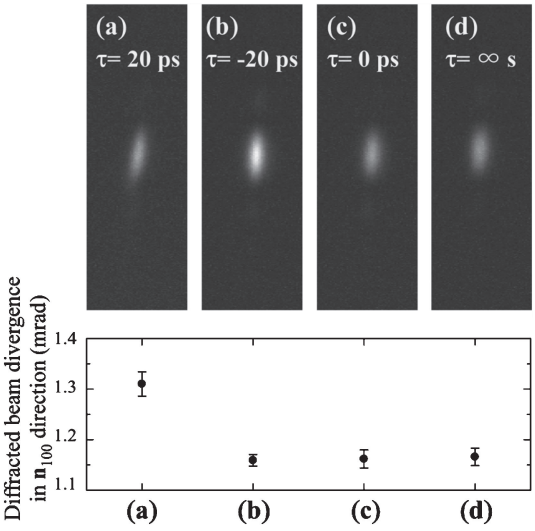

Fig. 4. Snapshots of 311 diffraction pattern of thin Si crystal for delay times of (a) $\tau = 20$ ps, (b) -20 ps, (c) 0 ps, and (d) $\gg 1$ s in the Ti:sapphire laser pump and XFEL single-shot probe measurement. The vertical divergences in (100)-direction of the diffracted beams are shown in the bottom.

In the experiment, we conducted the TR-Bragg CXD experiment for a 100 nm-sized thin single crystal layer, giving one-dimensional information in the surface normal direction, using the unfocused XFEL beam with a size of about 200 μm. For small crystal grains which are included in ceramics, a focused XFEL beam with a beam size of about 1 micron is then useful to obtain the diffraction intensity comparable with the present TR-Bragg CXD measurement.

4. Summary

We have demonstrated TR-Bragg CXD for a single thin crystal layer of silicon with a nanoscale thickness. Single-shot Bragg CXD patterns were successfully obtained in the asymmetric configuration with a grazing exit condition. CXD patterns recorded with an area detector shows diffraction intensity distribution in the surface normal direction. The thickness of the crystal estimated from the fringe interval is about 100 nm. The broadening of single-shot Bragg profile was observed at a delay time of 20 ps, indicating the transient lattice fluctuation induced by an optical laser. It is expected that the method is applicable with an X-ray focusing device to small sized crystal grains composing ceramic materials to investigate the initial breaking process for understanding of their toughness and stiffness.

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