Nondestructive Three-dimensional Observation of Crystal Defects in Single Crystal CaF_2 by Step-scanning White X-ray Section Topography

When we use single-crystalline materials for optics, it is important to understand the relationship between crystal defects and optical properties. A single crystal of calcium fluoride (CaF_2) is an attractive material for ultraviolet (UV) optics because of its high transparency to ultraviolet light and optical isotropy. The CaF_2 crystal has been used for the lenses of a deep UV optical lithography system. Large single crystals of CaF_2 are grown by the Bridgman-Stockbarger method and several types of crystal defects have occurred under the crystal growth process. We have found that the crystal defects within the lenses could cause serious problems in the performance of the deep UV image formation due to the extremely small inhomogeneity of the optical properties. Therefore, a technique for visualizing crystal defects deep within a single crystal is important.

X-ray topography is a valuable technique for investigating the crystal defects nondestructively. An in-house X-ray topography system is useful for observing defects near the surface of a crystal, but it is not practical for the defects deep within a thick crystal due to X-ray extinction by absorption. Data concerning crystal defects as well as defect at the surface are very important because the inhomogeneity of optical properties such as refractive index or birefringence emerges as an integrated value along the optical axis.

There are several reports of X-ray topography using high-energy and high-brilliance synchrotron radiation. Ludwig [1] demonstrated the visualization of three-dimensional distribution images of crystal defects that were reconstructed from a series of images that were recorded by tomographic acquisition using the monochromated synchrotron radiation. This technique is effective in measuring a nearly perfect single crystal, but it is not useful to observe an imperfect crystal, which has subgrain structures. Since the Bragg conditions for monochromatic X-rays on subgrains are different from each other, only a subgrain can be observed for an exposure.

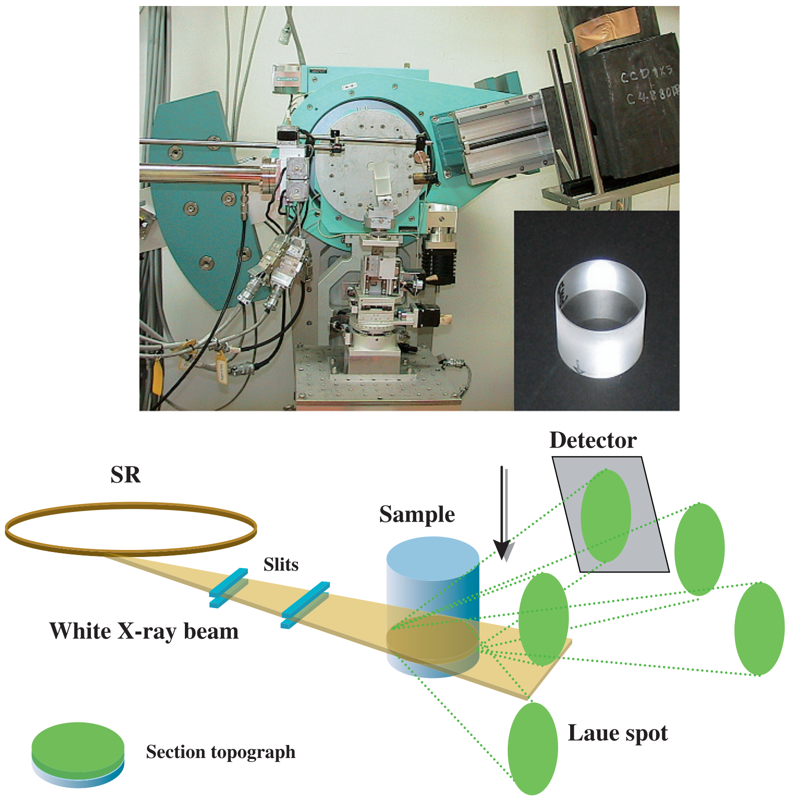

Fig. 1. Photographs of experimental setup and procedure of the step-scanning white beam X-ray section topography. The sample in the photograph was 30 mm in diameter and 30 mm in thickness.

We have developed the technique of step-scanning white X-ray section topography for the measurement of the three-dimensional distribution of subgrains and other crystal defects within bulky single crystal CaF_2. Using white X-rays enables the measurement of not only a perfect single crystal but also an imperfect single crystal for an exposure. Figure 1 shows the setup of the step-scanning white X-ray section topography system at beamline **BL28B2**. The white X-rays from the light source were shaped as a sheet-like beam of size between 10 mm and 30 mm (horizontal) × 0.1 mm (vertical) in accordance with the size of samples by the slits.

The single crystals of CaF_2 were grown along the (1 1 1) orientation. The samples were cut into cylinders from the ingots. The sizes of the samples were between 30 mm and 60 mm in diameter and between 30 mm and 50 mm in thickness. The sample was set on the goniometer composed of several stages.

Section topographs of the samples were recorded using a CMOS flat-panel detector or a CCD detector combined with a scintillator and a relay lens system. All cross-sections of the sample were recorded by repeating the exposure and translation of the sample along the horizontal and perpendicular directions to the top face of the sample. The exposure time of a section topograph was between 1 s and 15 s. Details of the experimental condition have been reported elsewhere [2].

Figure 2 shows an example of the section topographs corresponding to the cross section of the sample, which was 30 mm in diameter and 30 mm in thickness. It was clearly observed that the sample was composed of many subgrains. The subgrain boundaries were recognized as overlapping or separation of the images of the subgrains. Variations in the intensity of the subgrain images corresponded to crystal defects such as dislocations.

Figure 3 shows three-dimensional images reconstructed from the section topographs. Figure 3(a) is expressed by three horizontal planes, two vertical planes and a surface model of a subgrain shown as a yellow surface of the sample which was 30 mm in diameter and 30 mm in thickness. Figure 3(b) was expressed as a part of the sample which was 60 mm in diameter and 50 mm in thickness. It was easy to recognize the network structure of subgrains within the crystal. Furthermore, the width of the grain boundaries shown in Fig. 3(a) was thicker than that shown in Fig. 3(b). This indicates that the tilt angle between subgrains in Fig. 3(a) is larger than that in Fig. 3(b).

We have successfully visualized the three-dimensional distribution of the crystal defects of single crystal CaF_2. The step-scanning white X-ray section topography is a very useful and simple technique for analyzing the three-dimensional distribution of crystal defects within large single crystals.

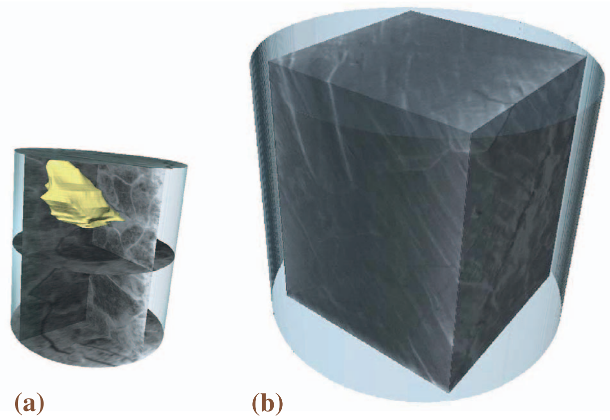

Fig. 3. Three-dimensional reconstructed image from section topographs. **(a)** 30 mm diameter and 30 mm thickness. **(b)** 60 mm diameter and 50 mm thickness.

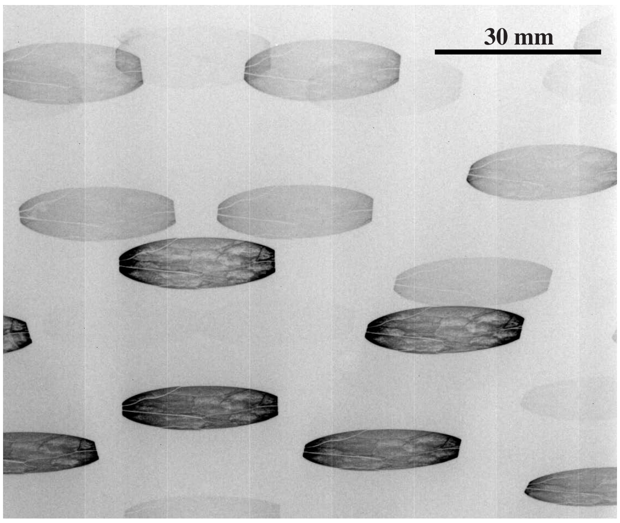

Fig. 2. Typical section topographs of the sample.

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References

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[2] *T. Mukaide, K. Kajiwara, T. Noma and K. Takada: J. Synchrotron Rad.* ***13*** *(2006) 484.*