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A ‘six-beam X-ray section topograph’ experiment was performed whose images are in excellent agreement with the images computer-simulated using the theory derived by one of the authors [Okitsu (2003). Acta Cryst. A59, 235–244] based on the Takagi–Taupin dynamical theory. X-rays whose wavefront was limited to 0.1 \( \times \) 0.1 mm were incident on a [111]-oriented floating-zone silicon crystal with thickness of 9.911 mm so that a six-beam condition was satisfied. The six reflection indices were 000 (forward diffraction), 440, 484, 088, 448 and 404. The plane of incidence of the 088 reflection was adjusted so as to be perpendicular to the direction of the incident horizontal linear polarization of the synchrotron radiation, which was monochromated into 18.5 keV (0.670 Å wavelength). While the simulations were performed under the assumptions that the incident X-rays were linearly polarized both horizontally and vertically, they were evidently different and only the simulated images under the assumption of horizontally polarized incident X-rays were in excellent agreement with the images obtained by the experiment.

1. Introduction

The possibility that the structure-factor phase information can be extracted from the analysis of three-beam X-ray diffraction profiles was pointed out by Ott (1938), Bijvoet & MacGillavry (1939) and Lipscomb (1949). Since the experimental work was published by Colella (1974), analysis of X-ray three-beam diffraction has come to be recognized as one of the most effective methods to solve the phase problem in crystal structure analysis (Post, 1977, 1979; Shen, 1986; Chang, 1986; Weckert et al., 1993; Weckert & Hümmer, 1997, 1998; Thorkildsen & Larsen, 1998; Larsen & Thorkildsen, 1998; Stetsko et al., 2001).

Incidentally, Kato (1976a,b, 1979, 1980a,b) derived the statistical dynamical theory to deal with the behavior of X-rays in a mosaic crystal based on the Takagi–Taupin equations (Takagi, 1962, 1969; Taupin, 1964). Thorkildsen (1987), Thorkildsen & Larsen (1998), Larsen & Thorkildsen (1998) and Thorkildsen et al. (2001) pointed out that a theory could be derived that can deal with the behavior of X-rays in a three-beam diffraction case in a mosaic crystal whose structure is to be solved if Kato’s procedure were applied to a Takagi–Taupin type dynamical theory extended to the three-beam case. Thorkildsen (1987) attempted to extend the Takagi–Taupin equations to the three-beam case. However, the extended theory neglects the effect of polarization. Okitsu (2003) has recently derived a Takagi–Taupin-type dynamical theory extended to \( n \)-beam (\( n \in \{3, 4, 6, 8, 12\} \)) cases that can deal with the effect of polarization correctly. Furthermore, a method to solve the theory numerically was also given by Okitsu (2003). The aim of the present paper is to verify the new theory by comparing the experimental result and the result computer-simulated based on the new theory.

The first computer-simulated section topograph image compared with an experimental section topograph image of a dislocation in a crystal was presented by Balibar & Authier (1967), based on an algorithm that can numerically integrate the Takagi–Taupin equations (Authier et al., 1968). These works were followed by studies on computer-simulated images of dislocations (Epelboin, 1974; Chukhovskii, 1974), planar defects (Epelboin, 1979), ferromagnetic domain walls (Nourtier et al., 1979) and strain centers (Green et al., 1990; Okitsu et al., 1992) on section topographs in the Laue geometry, and studies on computer-simulated dislocation images (Bedynska, 1973; Bedynska et al., 1976; Bubáková & Šourek, 1976; Gronkowski, 1980; Riglet et al., 1980) on topographs in the Bragg geometry. In the above works, algorithms with a constant integration step length (constant step algorithm)
were used. On the other hand, Epelboin (1983) developed an algorithm in which calculation step length was varied (varying step algorithm). In the varying step algorithm, the calculation step can be automatically changed so that the step length is small where the phase of the X-ray wave changes rapidly in the crystal. The circumstances related to the computer simulation using the Takagi–Taupin equations were reviewed by Epelboin (1985, 1987).

Recently, Heyroth et al. (2001) simulated three-beam pinhole topograph images based on an Ewald–Laue-type three-beam theory and compared them with experimental results performed at ESRF (Heyroth et al., 1999). On the other hand, in the present work, the simulations comparable with the experimental results obtained at SPring-8 were performed using the new Takagi–Taupin-type n-beam dynamical theory one of the present authors has derived (Okitsu, 2003).

2. Experimental

Fig. 1 shows the experimental arrangement of ‘six-beam X-ray section topography’. The first-order undulator radiation from BL09XU of SPring-8 was monochromated into 18.5 keV with silicon monochromator crystals which gave two consecutive 111 reflections. The degree of horizontal polarization of the monochromated X-rays had been estimated to be 0.994. X-rays whose dimension was limited to 1 × 1 mm by a four-quadrant slit system were incident on a [111]-oriented floating-zone silicon crystal whose surfaces were mechanically and chemically polished. The thickness of the crystal was 9.911 mm. The crystal was mounted on a θ–ω–φ three-axis goniometer so that θ, ω and φ axes were parallel to [211], [111] and [011] directions of the crystal, respectively, as shown in Fig. 1. This situation in the reciprocal space is shown in Fig. 2. A two-axis swivel-type goniometer whose φ axis was mounted on the ω axis was equipped on a tangent-bar-type θ-axis goniometer. The θ axis was adjusted to be horizontal and perpendicular to the incident X-ray beam axis.

First, the forward-diffracted and 088-reflected X-rays were monitored with PIN photodiodes PIN000 and PIN088 in Fig. 1 rotating the crystal around the θ axis. Two of the present authors (YI and YY) found that a 088 reflection peak was observed at the angular middle position of θ between two intensity peaks of forward-diffracted X-rays. The two peaks of forward-diffracted X-rays were assigned to anomalous transmission arising from 440 and 404 reflections. The peaks split since the θ axis was not completely perpendicular to the [111] direction of the crystal. Then, the φ axis was adjusted so that the intensity of the forward-diffracted beam did not split but gave a single peak at an angular position of θ, at which the peak of 088-reflected X-rays was observed simultaneously, which resulted in the satisfying of the six-beam case of 000, 440, 484, 088, 448 and 404 reflections. Some dynamical diffraction parameters related to these reflection indices are summarized in Table 1. Under the condition of the six-beam case, the dimension of the incident X-rays was further limited to 0.1 × 0.1 mm, when the ‘six-beam X-ray section topograph’ images were recorded.

3. Computer simulations

The method to solve the new theory numerically in the case of ‘n-beam X-ray section topography’ is in a separate paper (Okitsu, 2003). The computer simulations were performed with the boundary conditions for the incidence of horizontally and vertically polarized X-rays. A crystal with thickness 9.911 mm was divided for calculation into 2000 layers of identical thickness in the [111] direction of the crystal. Each layer was divided into small hexagonal pyramids as shown in Fig. 3. The electric displacement amplitudes of X-rays at the apex of the small pyramid were calculated step by step by
The crystal was divided into small hexagonal pyramids for simulation. The lengths $l_1$ and $l_2$ were assumed to be 6.92 and 4.83 $\mu$m.

The extinction length of the forward diffraction was calculated to be 23.7 $\mu$m. $|R_{0}^{(i)}R_{1}^{(j)}|$ is small compared to this value.

4. Results and discussion

The `six-beam X-ray section topograph' images were simultaneously recorded on an imaging plate set 27.3 mm behind the crystal so as to be parallel to the (111) surface of the crystal. The pixel size of the imaging plate was $50 \times 50$ $\mu$m. Fig. 4 shows the images taken with an exposure time of 30 s. In the center of Fig. 4, a horizontal bar corresponding to 10 mm is drawn. A dark gray level shows a high intensity of X-rays.

Fig. 5[$X_{h}(z)$] summarizes the results of experiment (expansions of Fig. 4) and the computer simulation, where $X$ is $E$ or $S$ (experiment or simulation, respectively), $y$ is $h$ or $v$ (with the incidence of horizontal or vertical polarization, respectively) and $z \in \{a, b, c, d, e, f\}$ corresponds to 000, 440, 484, 088, 448 and 404 reflections, respectively. Each calculation for obtaining $Sh(z)$ and $Sv(z)$ ($z \in \{a, b, c, d, e, f\}$) took three weeks of CPU time on a personal computer with a Pentium III 1 GHz processor. The original pixel size of all computer-simulated images shown hereafter is $13.37 \times 13.37$ $\mu$m.

Figs. 5[$Sh(z)$] and $Eh(z)$ ($z \in \{a, b, c, d, e, f\}$) are in excellent agreement in detail whereas Figs. 5[$Sv(z)$] evidently differ from Figs. 5[$Eh(z)$], which reveals that the new theory and the computer program can correctly deal with the behavior of X-rays in the six-beam case taking into account the effect of polarization. The vertically striped patterns in the right-lower regions of Figs. 5[$Eh(z)$] ($z \in \{a, b, c, d, e, f\}$) are considered to be striations introduced when the crystal was grown. This shows that `n-beam X-ray section topography' will be a useful tool to estimate minute strain fields in nearly perfect crystals when coupled with the simulation.

During the calculation procedure until the simulated results shown in Figs. 5[$Sh(z)$] ($z \in \{a, b, c, d, e, f\}$) are obtained, topograph images are simulated for a crystal with small thicknesses up to the thickness of the real crystal (9.911 mm). Figs. 6(a), (b), (c) and (d) show `six-beam X-ray section topograph' images computer-simulated under the assumption that the thickness of the crystal is $p \times 9.911$ mm and the distance of the imaging-plate position from the crystal is $p \times 27.3$ mm, where the values of $p$ are 0.25, 0.5, 0.75 and 1, respectively. The features of Figs. 6(a), (b), (c) and (d) are almost identical in contrast to Pendellösung fringes which appear on two-beam X-ray section topograph images. Figs. 7(a), (b), (c) and (d) show expansions of forward-diffracted X-ray images in Figs. 6(a), (b), (c) and (d). Figs. 8(a), (b), (c) and (d) show X-ray intensity profiles along the horizontal black lines in Figs. 7(a), (b), (c) and (d). Figs. 7 and 8 show more clearly that X-ray intensity distribution almost does not change with propagation in the crystal. X-ray intensity peaks indicated by white arrows in Fig. 7(d) and by black arrows in Fig. 8(d) are observed also in Figs. 7(a), (b), (c) and Figs. 8(a), (b), (c). Further, similar black spots can be observed also in Figs. 5[$Sh(z)$], where $z \in \{a, b, c, d, e, f\}$, which shows that X-ray energy flow has a maximum value in these positions in the crystal for any reason. It can be considered that the enhanced Borrmann effect plays an important role rather than the extinction effect in the six-beam case.

Table 1

| h  | Bragg angle (°) | $|F_h|$ | $|X_h|$ | $|\chi_h|$ |
|----|----------------|--------|--------|--------|
| 000| 0              | 112.5000 | 2.8292 × 10⁻⁵ | 1.2600 × 10⁻⁸ |
| 440| 20.4270        | 48.8457 | 1.0854 × 10⁻⁶ | 1.1134 × 10⁻⁸ |
| 484| 37.1935        | 25.6233 | 4.4453 × 10⁻⁷ | 8.6938 × 10⁻⁸ |
| 088| 44.2689        | 20.7667 | 3.1832 × 10⁻⁷ | 7.6823 × 10⁻⁸ |
| 434| 37.1935        | 25.6233 | 4.4453 × 10⁻⁷ | 8.6938 × 10⁻⁸ |
| 040| 20.4270        | 48.8457 | 1.0854 × 10⁻⁶ | 1.1134 × 10⁻⁸ |

The crystal was divided into small hexagonal pyramids for simulation. Two horizontal bars correspond to 10 mm. Figure 4

Images obtained by the `six-beam X-ray section topograph' experiment.
Figure 5
Experimental and computer-simulated images of the ‘six-beam X-ray section topographs’, the arrangement of which is shown in Fig. 1. [Xs(z)] means an image by X (X ∈ [S, E], where S is the simulation and E is the experiment), with the y-polarized incident X-rays (y ∈ [h, v], where h is horizontal and v is vertical) and for z reflection (z ∈ {a, b, c, d, e, f}), where a, b, c, d, e and f correspond to 000, 440, 088, 448 and 404 reflections, respectively). A scale of 10 mm is drawn in the right-upper part of [Eh(a)] as a horizontal bar.
5. Conclusions

The new theory (Okitsu, 2003) based on the Takagi-Taupin equations (Takagi, 1962, 1969; Taupin, 1964) and the computer program to solve the new theory have partly been verified for 'six-beam X-ray section topography' by comparing the experimental and the computer-simulated images. It was particularly revealed that the new theory can take into account the effect of polarization correctly. However, further studies to verify the new theory are necessary since the present work gives just one example of the agreement between the experiment and the computer simulation based on the new theory. The new theory will give the theoretical basis to develop X-ray optical devices applying the \( n \)-beam cases, for example a transmission-type monochromator (Kikuta, 2002), which takes advantage of the enhanced Borrmann effect.

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