Investigation of the Phase Shift in X-Ray Forward Diffraction Using an X-Ray Interferometer

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The phase shift of forward-diffracted x rays by a perfect crystal is discussed on the basis of the dynamical theory of x-ray diffraction. By means of a triple Laue-case x-ray interferometer, the phase shift of forward-diffracted x rays by a diamond crystal slab was investigated. It was verified that the phase shift in x-ray forward diffraction actually follows the dynamical theory of x-ray diffraction. It is suggested that a "phase-sensitive" experimental method using an interferometer can be applied to explore a wide variety of diffraction phenomena of x rays, γ rays, and neutrons in a periodic medium. [S0031-9007(96)00156-1]

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The phase shift of diffracted and forward-diffracted x rays by a periodic medium is one of the most fundamental problems for x-ray physics and x-ray diffraction optics. It is well known that Bragg-diffracted x rays by a perfect crystal undergo a 180° phase shift within Darwin's selective reflection region. This 180° phase shift in the Bragg diffraction plays an important role, for example, in the x-ray standing-wave method [1] and in x-ray topographic observations of defects in crystals. The phase of forward-diffracted x rays also undergoes a drastic change both inside and outside the diffraction region. This phase shift in the forward diffraction was recently applied to develop x-ray transmission phase plates [2,3]. Thus, the phase shift of x rays plays an essential role in many aspects of x-ray diffraction physics. However, there has been no simple method to observe the phase shift of diffracted and forward-diffracted x rays. To gain deeper insights into x-ray diffraction phenomena in a periodic medium, it is necessary to develop a new method that is sensitive to the phase shift of diffracted and forward-diffracted x rays.

X-ray interferometers developed by Bonse and Hart [4,5] are powerful tools for detecting a phase shift of x rays passing through an object. To date, x-ray interferometers have been applied, for example, to precise measurements of the atomic scattering factors [6], absolute lattice parameter measurements [7], and phase-contrast imaging [8–10]. Here, we demonstrate that x-ray interferometers are also useful for investigating the phase shift in x-ray forward diffraction. The basic idea is to insert a periodic medium that is adjusted near a diffraction condition into one of two coherent beam paths in the interferometer. The phase shift produced by the periodic medium is detected by measuring the intensity of an interfering outgoing beam. This simple method differs from conventional x-ray diffraction techniques in that it is sensitive to the phase of forward-diffracted x rays. In this paper, we report on the first successful investigation of the phase shift in x-ray forward diffraction by means of this "phase-sensitive" method.

At first, we consider the phase shift of forward-diffracted x rays by a perfect crystal on the basis of the dynamical theory of x-ray diffraction. When x rays are incident upon a crystal, forward-scattered x rays are produced in the transmission direction. The phase shift acquired by the forward-scattered x rays on passing through a crystal depends on the refractive index n of the crystal. In general, n is slightly less than 1 and complex, being given by

$$n = 1 - \alpha = 1 - \frac{\lambda^2 r_e}{2\pi V_c} F_0 , \tag{1}$$

where α is the deviation of the refractive index from 1, λ the x-ray wavelength, r_e the classical electron radius, V_c the unit-cell volume, and F_0 the crystal structure factor of 000 reflection. The refractive index, however, requires a slight correction when the incident beam almost satisfies the Bragg diffraction condition and multiple scattering takes place in a crystal. Usually, x-ray multiple scattering in a perfect crystal is described by the dynamical theory of x-ray diffraction within the limits of the two-beam case [11]. According to this theory, when the incident angle is in the vicinity of, but not too close to, the diffraction condition, the diffraction correction to n is analytically given by

$$\Delta n = -\frac{r_e^2 F_h F_{\bar{h}}}{4\pi^2 V_c^2} \frac{\lambda^4 C^2}{\Delta\theta \sin(2\theta_B)} , \tag{2}$$

where θ_B is the Bragg angle, $\Delta\theta$ is the offset angle from the diffraction condition, C is the polarization factor [$C = 1$ for σ polarization and $C = \cos(2\theta_B)$ for π polarization], and F_h and $F_{\bar{h}}$ are the structure factors of the hkl and $\bar{h}\,\bar{k}\,\bar{l}$ reflections, respectively. Note that Eq. (2) is valid for both Bragg and Laue cases. The range of validity of Eq. (2) is defined by the following inequality:

$$|\Delta\theta| \gg r_e \lambda^2 |F_h| / \pi V_c \sin(2\theta_B) . \tag{3}$$

Because the real part of the refractive index determines the phase shift of the forward-diffracted x rays, an

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additional phase shift caused by a diffraction correction is given by

$$\delta = 2\pi\,\mathrm{Re}(\Delta n)t/\lambda = -\frac{\pi}{2}\left[\frac{r_e^2\,\mathrm{Re}(F_h F_{\bar{h}})}{\pi^2 V_c^2}\,\frac{\lambda^3 C^2}{\Delta\theta\,\sin(2\theta_B)}\right]t\,, \tag{4}$$

where t is the length of the x-ray beam path in the crystal. From Eq. (4) we can see that the phase shift is inversely proportional to the offset angle, and increases linearly with the thickness of the crystal.

Rigorous calculations of the phase shift and the transmittance can be carried out by the dynamical theory of x-ray diffraction. For example, Fig. 1 shows the calculated $\Delta\theta$ dependence of the phase shift and the transmittance. Calculations were made for the forward diffraction associated with the Si 111 symmetric Laue-case diffraction. Other calculating conditions were $\lambda = 0.1$ nm, $C = 1$ (σ polarization) and $t = 76\ \mu$m (the thickness of the crystal slab is 75 μm). Figure 1 reveals some of the fundamental features of δ: (i) δ has an opposite sign on both sides of the reflection region, (ii) the maximum phase shift is achieved in the vicinity of the diffraction condition, and (iii) δ slowly approaches zero as the incident angle departs from the diffraction condition.

The phase shift of the forward-diffracted x rays by a nearly perfect crystal has been investigated by a recent measurement performed at a bending-magnet beam line, BL-15C at the Photon Factory (PF) at the National Laboratory for High Energy Physics. In the experiment we used a triple Laue-case (LLL) x-ray interferometer cut monolithically from a silicon ingot (Fig. 2). The interferometer has three parallel wafers which act as x-ray half mirrors. Each wafer is usually called the "beam splitter (S)," "mirror (M)," and "analyzer (A)."

When the incident x-ray beam satisfies the Bragg diffraction condition, the beam splitter creates two coherent beams, and, subsequently, the mirror and the analyzer recombine the interfering beams and produce two outgoing beams (O beam and H beam). We inserted a (001)-oriented diamond crystal slab of 1.09 mm thickness in one of the coherent beam paths. The diamond crystal was adjusted near to the asymmetric Laue-case 111 diffraction condition. In the experiment the wavelength of the incident beam was chosen to be $\lambda = 0.1$ nm. Monochromatic x rays were produced by a pair of silicon (220) perfect crystals.

At first, we measured the rocking curve of the diamond crystal without the x-ray interferometer. In Fig. 3(a) the solid line shows the forward-diffracted intensity, and the dashed line shows the diffracted intensity. We then measured the O-beam intensity using the interferometer while rotating the diamond crystal through the diffraction condition [Fig. 3(b)]. In Fig. 3(a) the forward-diffracted intensity is almost constant for $|\Delta\theta| > 10''$. In contrast, the O-beam intensity oscillates rapidly with respect to $\Delta\theta$ in Fig. 3(b). This O-beam intensity is explained by considering the phase difference produced between the two coherent beams in the x-ray interferometer. In the interferometer, one of the two coherent beams (reference beam) passes through the air of $n \approx 1$; the other (object beam) passes through the crystal of $n \approx 1 - \alpha + \Delta n(\Delta\theta)$. Therefore, the phase difference produced between the reference beam and the object beam is given by $\psi(\Delta\theta) = 2\pi\,\mathrm{Re}[-\alpha + \Delta n(\Delta\theta)]t/\lambda$. Assuming that the absorption of x rays by air is negligible, the resultant O-beam intensity is expressed as

$$I(\Delta\theta) \propto |1 + \sqrt{T(\Delta\theta)}\,\exp\{i\psi(\Delta\theta)\}|^2 = 1 + T(\Delta\theta) + 2\sqrt{T(\Delta\theta)}\,\cos[\psi(\Delta\theta)], \tag{5}$$

where $T(\Delta\theta)$ is the transmittance of x rays at the diamond crystal. This equation indicates that we can extract the phase-dependent term, $\cos[\psi(\Delta\theta)]$, by measuring both $T(\Delta\theta)$ and $I(\Delta\theta)$. In the present experiment, the

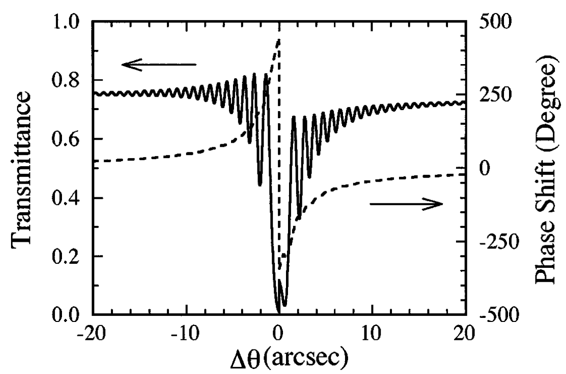

FIG. 1. Calculated transmittance (solid line) and phase shift (dashed line). Calculations were made for the x-ray forward diffraction associated with the Si 111 symmetric Laue-case diffraction. Other calculating conditions were $\lambda = 0.1$ nm, $C = 1$ (σ polarization), and $t = 76\ \mu$m (the thickness of the crystal plate is 75 μm).

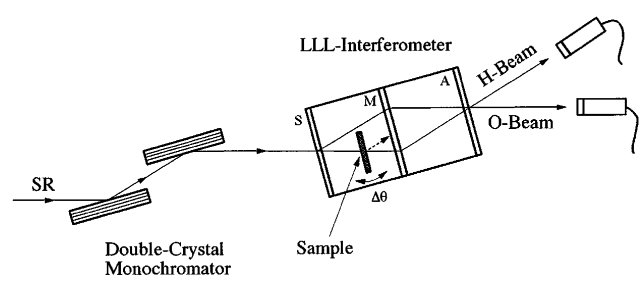

FIG. 2. Schematic view of the experimental setup. Monochromated x rays ($\lambda = 0.1$ nm) by a pair of silicon (220) perfect crystals are incident upon a triple Laue-case x-ray interferometer. In the interferometer, a (001)-oriented diamond crystal slab of 1.09 mm thickness is inserted into one of the two coherent beam paths. The diamond crystal is adjusted near the asymmetric Laue-case 111 diffraction condition. The intensities of the interfering outgoing beams (O beam and H beam) are measured by NaI scintillation counters.

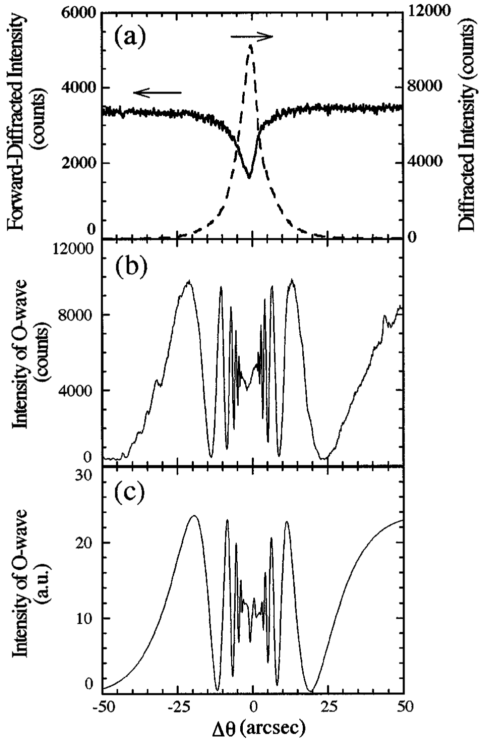

FIG. 3. (a) Rocking curves of the diamond crystal measured without the x-ray interferometer. The dashed line shows the intensity of the diffracted beam, and the solid line shows the intensity of the forward-diffracted beam. (b) Measured *O*-beam intensity with the x-ray interferometer. (c) Calculated *O*-beam intensity. In the calculation, the curve is convoluted with a Gaussian instrumental width function with a standard deviation of $1''$.

observed $T(\Delta\theta)$ was almost constant with respect to $\Delta\theta$. Therefore, we can conclude that the oscillation of the *O*-beam intensity originated mainly from the phase difference, $\psi(\Delta\theta)$. Figure 3(c) shows the calculated $\Delta\theta$ dependence of the *O*-beam intensity. In the calculation, the curve is convoluted with a Gaussian instrumental width function with a standard deviation of $1''$. There is general qualitative agreement between the calculated curve and the experimental data. This result confirms to us that the phase shift, $\psi(\Delta\theta)$, actually follows the dynamical theory of x-ray diffraction. Discrepancies between the experimental data and the calculation are due to distortion of the diamond crystal.

The experimental method using the interferometer described above is quite unique in that it can yield information concerning the phase shift of the forward-diffracted beam by a periodic medium. Although we have limited our interest in this paper to x-ray forward diffraction in the two-beam case, we would like to emphasize that a wide variety of diffraction phenomena of x rays, γ rays, and neutrons in crystals and synthetic multilayers can be explored by means of this phase-sensitive method. For example, time-domain interferometry of nuclear forward diffraction [12] will be one of the most interesting applications of this method. Another interesting application is phase-contrast imaging of imperfections and strain fields in a periodic medium.

In summary, we have discussed the phase shift in x-ray forward diffraction based on the dynamical theory of x-ray diffraction. Using an x-ray interferometer, we could obtain information concerning the phase shift of forward-diffracted x rays by a nearly perfect crystal. By comparing the experimental data with the theoretical calculation, it was verified that the phase shift in x-ray forward diffraction actually follows the dynamical theory of x-ray diffractions. Further, we have suggested that the phase-sensitive experimental method using an interferometer can be applied to explore a wide variety of diffraction phenomena of x rays, γ rays, and neutrons in a periodic medium.

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