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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 silicon crystal in the Bragg geometry was investigated.

Keywords: X-ray diffraction; forward diffraction; phase shifts; X-ray interferometer.

1. Introduction

The phase shift of diffracted and forward-diffracted X-rays by a periodic medium is one of the most fundamental problems for X-ray diffraction physics. 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 plays an important role, for example, in the X-ray standing-wave method (Batterman, 1964). The phase of forward-diffracted X-rays also undergoes a drastic change both inside and outside the strong diffraction region, and this is the basis for X-ray transmission phase plates (Hirano *et al.*, 1991, 1995). Thus, the phase shift of X-rays plays an essential role in many aspects of X-ray diffraction physics. However, there have been few methods of probing 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 and neutron interferometers developed by Bonse & Hart (1965*a*,*b*) are powerful tools for detecting the phase shift of X-rays and neutrons passing through an object. In the field of neutron diffraction physics, the phase shift of forward-diffracted neutrons by a crystal was observed by means of a neutron interferometer (Graeff *et al.*, 1978; Rauch, 1989). In the same way, we can probe the phase shift of forward-diffracted X-rays using an X-ray interferometer (Hirano & Momose, 1996). The basic idea is to insert a periodic medium, which is adjusted to 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. In the previous paper, we investigated the phase shift of forward-diffracted X-rays by a crystal in the Laue geometry (Hirano & Momose, 1996). Here we report on the phase shift in the Bragg geometry.

2. Theory

When X-rays impinge upon a perfect 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 is complex, being given by

$$n = 1 - \alpha = 1 - (\lambda^2 r_e / 2\pi V_c) F_0, \qquad (1)$$

where α is the deviation of the refractive index from 1, λ is the X-ray wavelength, r_e is the classical electron radius, V_c is the unit-cell volume and F_0 is the crystal structure factor of 000 reflection. The refractive index, however, requires a slight correction when the incident beam almost satisfies the 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 (see, for example, Batterman, 1964; Ishikawa & Kohra, 1991). 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

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

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 $\overline{hkl}$ reflections, respectively. Note that equation (2) is valid for both Bragg and Laue cases. Because the real part of the refractive index is related to the phase shift of the forward-diffracted X-rays, an additional phase shift caused by a diffraction correction is given by

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

where t is the length of the X-ray beam-path in 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 220 Bragg-case diffraction. Other calculating conditions were λ = 0.1 nm, C = 1 (σ polarization) and t = 38 μm (the thickness of the crystal slab is 10 μm).

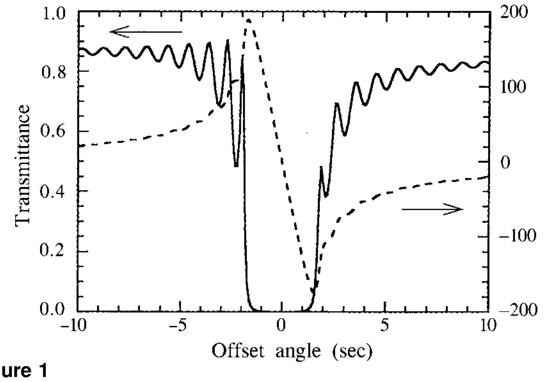

Figure 1
Calculated transmittance (solid line) and phase shift (dashed line). Calculations were made for the X-ray forward diffraction associated with the Si 220 Bragg-case diffraction. Other calculating conditions were λ = 0.1 nm, C = 1 (σ polarization) and t = 38 μm (the thickness of the crystal is 10 μm).

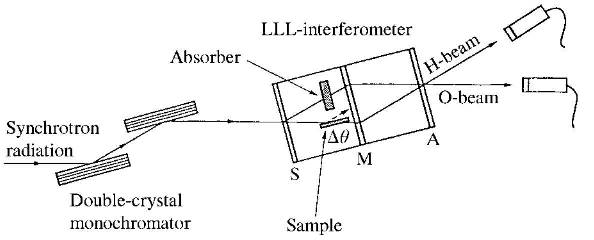

Figure 2
Schematic view of the experimental set-up. X-rays ($\lambda = 0.1$ nm), monochromated by a pair of Si(220) perfect crystals, are incident upon a triple Laue-case X-ray interferometer. In the interferometer, an Si(110) wafer of 400 µm thickness is inserted into one of the two coherent beam-paths. The wafer was adjusted to near the 220 Bragg diffraction condition. A silicon slab of 1.5 mm thickness was inserted into another beam path to balance the intensity of the two coherent beams. The intensities of the interfering outgoing beams (O-beam and H-beam) are measured by NaI scintillation counters.

3. Experiment

The phase shift of the forward-diffracted X-rays by a silicon crystal has been investigated at a bending-magnet beamline, BL-15C at the Photon Factory at the Institute of Materials Structure Science. 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. The wafers are usually called the 'splitter' (S), 'mirror' (M) and 'analyser' (A). When the incident X-ray beam satisfies the diffraction condition, the splitter creates two coherent beams and, subsequently, the mirror and the analyser recombine the interfering beams and produce two outgoing beams (O-beam and H-beam). We inserted an Si(110) wafer of 400 µm thickness into one of the coherent beam paths. The wafer was adjusted to near the 220 Bragg diffraction condition. We inserted a silicon slab of 1.5 mm thickness into another beam path to balance the intensity of the two coherent beams. 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 Si (220) perfect crystals.

We measured the O-beam intensity while rotating the silicon crystal through the diffraction condition (Fig. 3). The O-beam intensity oscillates rapidly with respect to $\Delta\theta$ (Fig. 3). This oscillation is due to the phase shift produced between the two coherent beams in the X-ray interferometer. In the interferometer one of the two coherent beams (reference beam) passes through an absorber of $n \simeq 1 - \alpha$; the other (object beam) passes through the sample of $n \simeq 1 - \alpha + \Delta n(\Delta\theta)$. If we assume that the length of the beam path in the absorber is equal to that in the sample, the phase shift produced between the reference beam and the object beam is given by $\psi(\Delta\theta) = 2\pi \mathrm{Re}[\Delta n(\Delta\theta)]t/\lambda$. The resultant O-beam intensity is expressed as

$$\begin{aligned} I(\Delta\theta) &\propto \left| T_A^{1/2} + [T_S(\Delta\theta)]^{1/2} \exp[i\psi(\Delta\theta)] \right|^2 \\ &= T_A + T_S(\Delta\theta) + 2[T_A T_S(\Delta\theta)]^{1/2} \cos[\psi(\Delta\theta)], \qquad (4) \end{aligned}$$

where $T_S(\Delta\theta)$ is the transmittance of X-rays at the sample and T_A is that at the absorber. This equation indicates that we can extract the phase-dependent term, $\cos[\psi(\Delta\theta)]$, by measuring T_A, $T_S(\Delta\theta)$ and $I(\Delta\theta)$. In the present experiment, the observed $T_S(\Delta\theta)$ was almost constant with respect to $\Delta\theta$. Therefore, we can conclude that the oscillation of the O-beam intensity mainly originates from the phase difference $\psi(\Delta\theta)$.

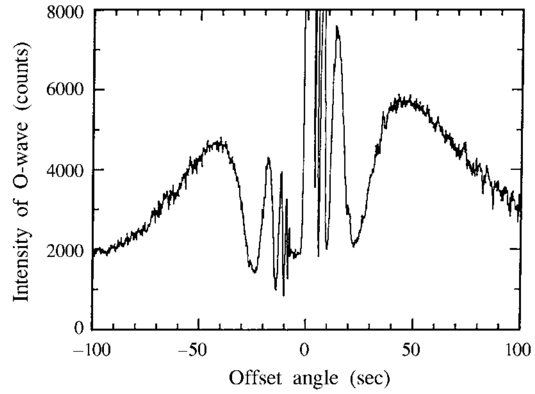

Figure 3
Measured offset angle ($\Delta\theta$) dependence of the O-beam intensity.

The experimental method using the interferometer described above is quite unique in that it can probe 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 (Kagan *et al.*, 1979) 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.

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