EUROPHYSICS LETTERS 20 October 1993

Europhys. Lett., 24 (3), pp. 211-216 (1993)

Direct Measurements of X-Ray Anomalous Transmission in Six-Beam Laue Diffraction.

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(received 20 July 1993; accepted 16 September 1993)

PACS. 61.10D – Theories of diffraction and scattering.
PACS. 61.10F – Experimental techniques (inc. apparatus, techniques and calculation methods for analyzing experimental results).
PACS. 78.70C – X-ray scattering.

Abstract. – Results of experimental measurements of transmitted-beam intensity under conditions of six-beam $(220, 242, 044, \bar{2}24, \bar{2}02)$ symmetrical Laue diffraction of synchrotron X-ray plane-wave radiation in a silicon crystal are presented along with a computer simulation. A new experimental arrangement with a six-beam collimator was used for the first time. The enhancement of the anomalous-transmission effect (in 3.3 times for $\mu t = 12$) in comparison with the (220) two-beam diffraction has been observed in a direct way. Experimental results agree with the plane-wave theory taking account of two-dimensional convolution.

Introduction. – An anomalous transmission of X-rays through a thick absorbing crystal is one of the fundamental phenomena of X-ray dynamical diffraction. Observed for the first time in 1941 by Borrmann [1] this effect has been studied in detail both theoretically and experimentally since then. The physical reason for the anomalous transmission is the excitation of the standing-wave field inside the crystal with intensity modulated along the diffraction vector and close to zero at the reflection planes. In 1965 Borrmann and Hartwig [2] observed a more complicate effect of anomalous transmission in the case of three-beam $(111, \bar{1}11)$ X-ray diffraction. Due to the three-beam interaction the minimum-absorption coefficient was much less than at two-beam (111) diffraction. As follows from theory [3, 4] if all reciprocal lattice vectors are lying in one plane, X-ray standing waves are modulated also in this plane. The structure of these fields offers new possibilities to reduce the interaction of X-rays with atoms and therefore to further decrease the X-ray absorption.

From this point of view the six-beam $(220, 242, 044, \bar{2}24, \bar{2}02)$ symmetrical Laue diffraction case is the most interesting [5, 6]. Twelve Bloch waves (six for each polarization state) with different absorption coefficients are excited in a crystal. The minimum absorption coefficient is limited by the Compton scattering and, as calculations show, one thousand times less than at non-diffraction interaction. As for the strongest process of photoelectric absorption, it decreases in ten thousand and more times [7]. The unique properties of the six-beam diffraction have attracted the attention of many authors (for references see [3] and [4]).

To study multiple-diffraction effects experimentally, a high collimation of X-ray beam both in vertical and horizontal directions is required. Moreover, fine angular rotations around two perpendicular axes should be provided. Such kind of experiments have been practically impossible so far, mainly because of the dramatic loss of intensity when using conventional X-ray sources. Instead, a more simple experimental arrangement based on the topographic observation of anomalous-transmission lines was used. Multiple-diffraction effects appeared as anomalies at cross points of these lines.

However, results obtained in this way showed that the anomalous-transmission patterns as well as the dependence of the intensity on crystal thickness disagree with predictions of plane-wave diffraction theory. It has been shown [8, 9] that the theory of X-ray spherical-wave multiple diffraction has to be used for a correct explanation of experimental results. It became clear that the anomalous-transmission effect was masked by optical effects of X-ray focusing or defocusing. So, the only way to observe the six-beam anomalous-transmission effect directly is the diffractometer analysis.

In this paper we propose new experimental arrangement and discuss first experimental results of angular-distribution measurements of anomalously transmitted intensity. The first quantitative measurements of the six-beam anomalous-transmission effect are the main result of our work.

Experiments and results. – The measurements were carried out with synchrotron radiation beam from vertical wiggler at SR source «Photon Factory». The experimental arrangement is shown in fig. 1. The SR beam was monochromized by a double-crystal monochromator with symmetrical Si(111) crystals. A symmetrical Si(220) crystal was used to reflect the X-ray beam to the following collimator and sample crystals. As we mentioned above, a high degree of the two-dimensional angular collimation of the X-ray beam should be provided to study the anomalous-transmission effect in a strightforward way.

To solve these problems, we used a new method of collimation based on the six-beam anomalous-transmission phenomenon itself. A perfect immovable Si crystal with a thickness of 5 mm was adjusted for a six-beam Laue diffraction condition. A forward-transmitted beam was used as an incident to the sample. As calculations show [10], the angular ranges of anomalous transmission are very narrow in two directions ($\Delta\theta$ and $\Delta\varphi$, see fig. 1). The use of the effect under study for the angular collimation makes us sure that the degree of this collimation will be sufficient to measure the effect itself.

Perfect dislocation free FZ (111)-oriented silicon platelets with thicknesses of $t = 3$ mm and 5 mm were used as samples. The collimator crystal and the sample were placed on two precision diffractometers with vertical θ-axis [11]. The swivel tables on which the crystal were fixed allowed a fine (0.1 arcsec/step) rotation about the horizontal φ-axis which was parallel to the (044) reciprocal lattice vector.

The $\Delta\theta$-dependences of transmitted-beam intensities were measured at different values of $\Delta\varphi$. The result is a two-dimensional $(\Delta\theta, \Delta\varphi)$ intensity distribution data. The value of $\Delta\varphi = 0$ corresponds to the section through the centre of the six-beam diffraction angular region. Diffracted (220) and ($\bar{2}$02) beams were recorded simultaneously with the transmitted beam. Additional NaI detectors were set at (220) and (044) diffracted beams after the collimator to monitor the angular stability of the collimator crystal. The measurements were performed at

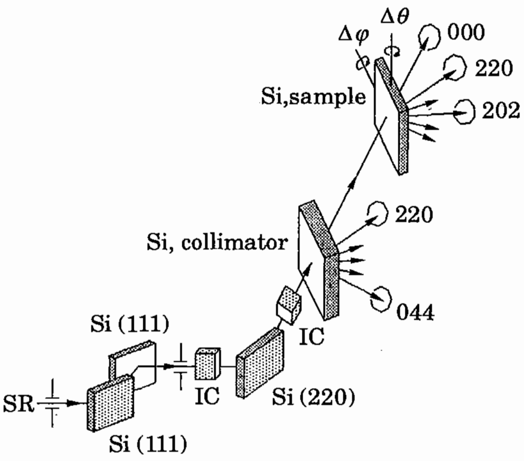

Fig. 1.

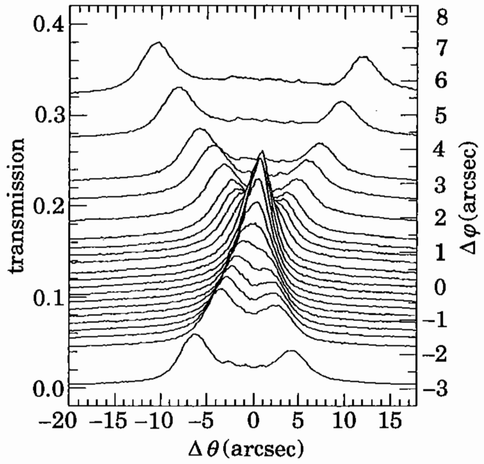

Fig. 2.

Fig. 1. – Experimental X-ray optics arrangement.

Fig. 2. – Experimental $\Delta\theta$-dependences of the six-beam anomalous-transmission coefficient measured for different values of $\Delta\varphi$. The angle $\Delta\varphi = 0$ corresponds to the centre of the six-beam diffraction region. The scale on the left side shows the value of the transmission coefficient.

wavelengths of $\lambda = 0.93$ Å and 1.15 Å, which give $\mu t = 12$ and 24 for the sample thickness of $t = 3$ mm and $\mu t = 19.4$ and 39.7 for $t = 5$ mm (μ is the absorption coefficient in the direction of the normal to the surface).

The discussion in this paper is restricted to the case of $\lambda = 0.93$ Å and $t = 3$ mm ($\mu t = 12$), where the effect of anomalous transmission is not so strong. It makes it possible to observe the details of the intensity distribution inside both the regions of six-beam and two-beam diffractions.

A set of $\Delta\theta$-dependences of the (000) beam intensity for different values of $\Delta\varphi$ is shown in fig. 2. These curves were measured with the angular step of $\Delta\theta = 0.2$ arcsec. To make the pattern more clear, the curves have been shifted with respect to each other in the vertical direction. The left vertical axis presents the scale of the transmitted coefficient. The values of the $\Delta\varphi$ angle correspondent to different curves are shown on the right. It is clearly seen that the peak of the six-beam diffraction in the central part of fig. 2 is three times more intense than the peaks of the two-beam 220 diffraction. This fact demonstrates that the enhancement of the anomalously transmitted intensity does exist. The effect is rather weak for the given value of μt. The most interesting question now is the agreement of experimental results with theory.

Theory and discussion. – The theory of X-ray multiple diffraction in perfect crystals is elaborated rather well (see, *e.g.* [3, 4, 10]). In the Laue case of plane-wave diffraction in a plane-parallel thick crystal, let $\boldsymbol{k}_0$ be a wave vector and the angles $\Delta\theta$ and $\Delta\varphi$ determine deviations of $\boldsymbol{k}_0$ from the exact six-beam direction. The transmission coefficient $P_{\mathrm{T}}^{ps}(\Delta\theta, \Delta\varphi)$ defines the transformation of s-polarized plane-wave intensity at the entrance surface into the p-polarized one at the exit surface. It is determined by the formula

$$P_{\mathrm{T}}^{ps}(\Delta\theta, \Delta\varphi) = \sum_j [B_{0p}^j B_{0s}^j]^2 \exp[-\mu_j t], \tag{1}$$

where t is the crystal thickness, B^j_{0p} is $0p$-component of the eigenvector (standing-wave field) with number j.

The eigenvectors have 12 components (6 for each polarization state $p = \pi, \sigma$) and can be calculated as eigensolutions of the kinematical scattering matrix G^{ps}_{mn} that is determined by the Fourier components of the crystal susceptibility $\chi = \chi_r + i\chi_i$, polarization factors and deviations from the Bragg law taking account of $\Delta\theta$, $\Delta\varphi$ dependences. The eigensolution problem can be written in the following form:

$$\sum_{ns} G^{ps}_{mn}(\Delta\theta,\, \Delta\varphi)\, B^j_{ns} = \varepsilon_j B^j_{mp}\,, \tag{2}$$

where indices $m, n = 0, \dots, 5$ number the beams.

For the simplicity of calculation, without loss of exactness, we use the approximation based on the perturbation theory with a small value of (χ_i/χ_r). In this way eq. (2) was solved for a non-absorbing crystal (without χ_i). Absorption coefficients μ_j for each standing-wave field were evaluated as a diagonal matrix elements of the matrix M^{ps}_{mn} describing the absorption, namely,

$$\mu_j = \sum_{mp}\sum_{ns} B^j_{mp} M^{ps}_{mn} B^j_{ns}\,. \tag{3}$$

A comparison of theoretical calculations with experimental results can be carried out only with a detailed account of the experimental arrangement. In our case it means a convolution of the two-dimensional matrix-function $\widehat{P}_{\mathrm{T}}(\Delta\theta,\, \Delta\varphi)$ of the sample with a two-dimensional matrix-function $\widehat{P}_{\mathrm{C}}(\Delta\theta,\, \Delta\varphi)$ of the collimator crystal and summing over polarization states, namely,

$$I_{\mathrm{T}}(\Delta\theta,\, \Delta\varphi) = \frac{\displaystyle\sum_{s,\, s'} \int_R \mathrm{d}\xi\, \mathrm{d}\eta\, P^{s's}_{\mathrm{T}}(\Delta\theta + \xi,\, \Delta\varphi + \eta)\, P^{s\sigma}_{\mathrm{C}}(\xi,\, \eta)}{\displaystyle\sum_{s} \int_R \mathrm{d}\xi\, \mathrm{d}\eta\, P^{s\sigma}_{\mathrm{C}}(\xi,\, \eta)}\,. \tag{4}$$

Here $s, s' = \pi, \sigma$. We suppose in eq. (4) that the SR was σ-polarized, but the six-beam collimator changes the polarization and the radiation incident on the sample has both σ- and π-components.

The angular range of integration R in eq. (4) is determined by reflections of preceding crystals by taking account of slits and a non-monochromaticity of the incident radiation. The range R is known not so well. We carry out the integration within the rectangular $-6'' < \Delta\theta < 6''$, $-3.5'' < \Delta\varphi < 3.5''$. This range exceeds the width of total reflection range of the preceding (220) reflection and takes account effectively of the non-monochromaticity.

The results of the calculations are shown in fig. 3 and 4. Figure 3 shows a two-dimensional $(\Delta\theta, \Delta\varphi)$ distribution of the forward-transmitted beam intensity for the sample crystal in the case of σ-polarized input beam (the so-called «intrinsic» curve). Polarization states π and σ were chosen as standard for the two-beam 044 diffraction. The central peak is three times more intense than the two-beam 220 diffraction peaks at tails of distribution that agree with the experimental curves of fig. 2. But there are significant differences between theoretical and experimental curves. Five two-beam diffraction peaks corresponding to all the reflections of the six-beam multiple diffraction case can be easily seen. High-order reflections have sharp peaks with rather high maximum intensity corresponding to the, respectively small, value of $\mu t = 12$. It is not so in the experimental curves.

Figure 4 shows three sections for $\Delta\varphi$ values of $-2.8''$, 0 and $4.4''$, in which theoretical curves calculated by taking account of the convolution by eq. (4) are compared in the same

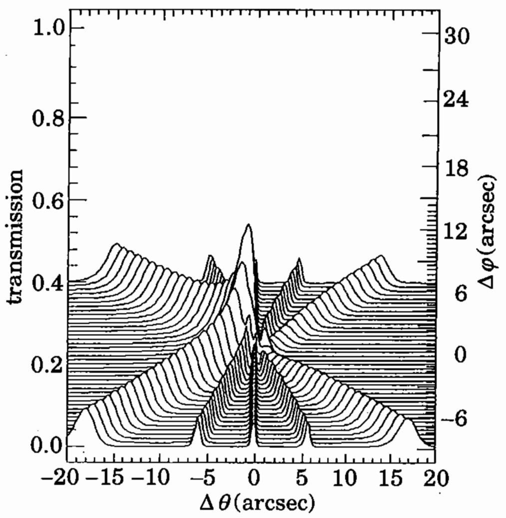

Fig. 3.

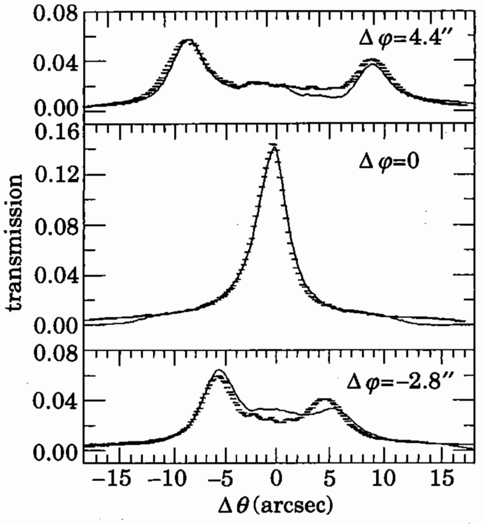

Fig. 4.

Fig. 3. – Theoretical two-dimensional $(\Delta\theta, \Delta\varphi)$ function of the anomalous-transmission coefficient calculated for experimental parameters and perfect σ-polarized plane wave.

Fig. 4. – Comparison of experimental (strip) and theoretical (solid lines) curves calculated with two-dimensional convolution ($\Delta\varphi = -2.8$; 0; 4.4 arcsec).

scale with the corresponding experimental curves. As follows from the figure, the convolution improved the correspondence very well. It may be further improved by optimization of the range of integration R, but it is not so important at this stage of consideration. The account of non-monochromaticity theoretically is not so simple a problem and it is beyond the scope of this paper. A more important problem is the further improvement of the collimation and the decrease of the non-monochromaticity of the incident beam experimentally. As a first attempt to obtain quantitative comparison between theory and experiment, the results obtained may be qualified as very good.

The good agreement between experimental results and theory obtained for the first time is the most important result of this work. It proves the effectiveness of the proposed new type of X-ray collimator. The six-beam collimator can be sucessfully used in different diffraction experiments (three-beam multiple scttering, X-ray standing waves, total external reflection and others). However, for respectively small μt value, the 220 two-beam tails of the angular distribution of the forward-transmitted beam intensity after collimator crystal are essential. As can be shown from our calculations, in this case the 242-reflected beam instead of the forward-transmitted beam is more effective. The measurement of a stronger enhancement of the anomalous transmission in the six-beam case requires higher values of μt. Such cases will be analysed elsewhere.

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We would like to express our acknowledgement to Profs. K. KOHRA and T. MATSUSHITA for their encouragement and support of our experiments at the Photon Factory. One of us (VGK) acknowledges the support of the Russian Foundation for Fundamental Research through Grant No. 93-02-2590.

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