Asymmetry factor in diffraction of x-rays in conditions of total external reflection

P. A. Aleksandrov, A. M. Afanas'ev, and S. A. Stepanov

Institute of Crystallography, Academy of Sciences of the USSR

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The problem of the dynamic diffraction of x-rays in conditions of total external reflection on planes perpendicular to the entry surface is solved with allowance for small misorientations of the reflecting planes. It is shown that for misorientation toward the incident beam one may obtain a continuous transition from diffraction in the Bragg geometry to diffraction in the Laue geometry for the same reflection by varying the angle between the incident beam and the surface of the crystal. It is shown that diffraction in conditions of total external reflection is uniquely sensitive to small misorientations of the reflecting planes.

In this article we develop the dynamic theory of x-ray scattering for diffraction by crystals in conditions of total external reflection.

A scheme of x-ray diffraction with total external reflection was suggested in Ref. 1. In this scheme the incident beam of x-rays is directed onto the crystal so that diffraction conditions are realized in the Laue geometry for planes perpendicular to the entry surface, and simultaneously makes a small angle φ with the surface (Fig. 1). The small angle of incidence – of the same order as the critical angle for total external reflection – gives intense specular reflection of the incident and diffracted wave from the surface of the crystal, and in principle this makes it possible to investigate the crystal structure of thin surface layers a few angstroms or thicker.

Afanas'ev and Melkonyan[2] constructed a dynamic theory of diffraction in conditions of total external reflection for perfect crystals. It was shown that the angle φ' of emergence of the specularly reflected diffracted wave depends on how accurately the Bragg condition is satisfied. In other words, there is a strict correspondence between the angles φ and φ' and the parameter α determining the deviation from the exact diffraction condition,

$$\alpha=\Phi^2-\Phi'^2, \tag{1}$$

where

$$\alpha=\frac{(\varkappa_0+K_h)^2-\varkappa_0^2}{\varkappa_0^2}\approx-2\sin 2\theta_B(\theta-\theta_B), \tag{2}$$

where $\varkappa_0$ is the wave vector of the incident x-ray wave and K_h is the reciprocal lattice vector.

Realization of relationship (1) opens up fundamentally new experimental possibilities. On this basis Imamov et al.[3] performed an experiment in which the intensity P_h^S of the specularly reflected diffracted wave was represented as a function of the angle Φ', and not as a function of the angle θ. In this method, relatively coarse measurements of the angles Φ and Φ', with errors of a few minutes of angle, reflect very small deformations of the crystal lattice, corresponding to changes of a fraction of an arc second in the parameter α.

Although in Refs. 2 and 3 the fundamental theoretical postulates were confirmed, there were also appreciable discrepancies between the theoretical predictions and the experimental measurements, and these have stimulated further development of the theory.

In this article the problem of diffraction in conditions of total external reflection by perfect crystals is solved with allowance for small misorientation of the reflecting planes from the direction of the normal to the surface.

It was found that misorientation by small angles φ of the order of the angles Φ, Φ' appreciably alters the angular dependence of P_h^S.

This fact may be important when experimental data are compared with theoretical calculations, since, for example, for silicon and germanium with Cu Kα radiation intense specular reflection arises at angles of incidence of $\Phi \leqslant 10'$-$20'$, whereas the accuracy with which the orientation can be set in cutting the crystals is $\varphi \sim \pm 30'$, and the accuracy with which the misorientation can be measured by known x-ray methods and thus choose the specimens is $\varphi \sim \pm 15'$.

Let us consider the scheme of diffraction shown in Fig. 1. For simplicity we shall assume that the incident radiation is polarized perpendicular to the plane of scattering formed by vectors $\mathbf{k}_0$ and $\mathbf{k}_h$ (σ-polarization).

Considering the conditions of continuity of the tangential components of the wave vectors at the entry surface of the crystal and the relation between vectors $\mathbf{k}_0$ and $\mathbf{k}_h$ and the misorientation of the reflecting planes through a small angle φ from the direction of the normal to the surface, we obtain the following relation between the angle of emergence of the specularly reflected diffracted wave Φ', the angle of incidence Φ, and the parameter α of de-

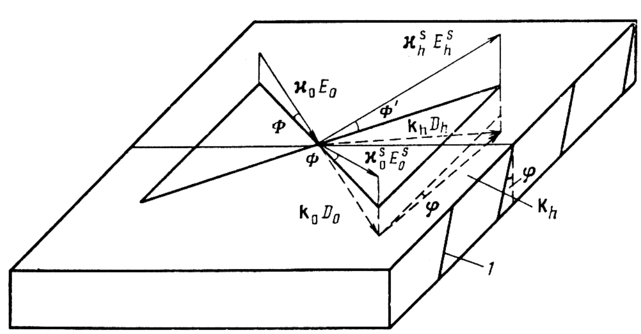

FIG. 1. Scheme of asymmetric diffraction in conditions of total external reflection. 1) Reflecting plane. Case $\varphi = 0$ corresponds to symmetric diffraction and is considered in Refs. 1-3.

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viation from the exact Bragg condition:

$$\alpha=(\Phi+\Psi)^2-\Phi'^2, \tag{3}$$

where

$$\Psi=2\varphi\sin\theta_B \tag{4}$$

is the effective angle of misorientation.

Taking account of misorientation, the fundamental equations of the dynamic theory can be transformed to the following form (see, e.g., Ref. 2):

$$\begin{gathered}(u^2-\Phi^2)D_0=\chi_0 D_0+\chi_{\bar h} D_h,\\ [(u+\Psi)^2-\Phi'^2]D_h=\chi_0 D_h+\chi_h D_0,\end{gathered} \tag{5}$$

where $u = k_{0z}/\varkappa_0$ is a parameter to be determined.

In this case the dispersion equation is not biquadratic as in Ref. 2, but a general quartic in u,

$$(u^2-\Phi^2-\chi_0)[(u+\Psi)^2-\Phi'^2-\chi_0]-\chi_h\chi_{\bar h}=0. \tag{6}$$

Equation (6) is most conveniently solved by the numerical method of tangents. From its four roots $u^{(i)}$ we must choose the two for which $\mathrm{Im}\,u^{(i)} > 0$ (thick-crystal approximation).

It is easy to prove that Eq. (6) always has two roots with a positive imaginary part and two with a negative imaginary part. In fact, when $\chi_{i0} = \chi_{ih} = \Psi = 0$, Eq. (6) becomes a biquadratic equation with real coefficients, which always has two roots with a positive imaginary part and two with a negative imaginary part. Let us consider the dependence of the roots of Eq. (6) on the parameters χ_{i0}, χ_{ih}, Ψ and suppose that for some values of χ_{i0}, χ_{ih}, $\Psi \neq 0$ the sign of the imaginary part of some one of the roots is reversed. Correspondingly for these values of the parameters, Eq. (6) has a real root. However, by direct substitution we can easily verify that for no physically possible values of these parameters can the real root simultaneously satisfy the real and imaginary parts of Eq. (6). This shows that Eq. (6) always has two roots with a positive imaginary part and two with a negative imaginary part.

In order to determine the amplitudes E_h^s and E_0^s we must use the condition of continuity of the tangential components of the electric and magnetic fields at the entry surface of the crystal.

The first conditions take the form

$$\begin{gathered}\Phi(E_0-E_0^s)=u^{(1)}D_0^{(1)}+u^{(2)}D_0^{(2)},\\ -\Phi' E_h^s=(u^{(1)}+\Psi)D_h^{(1)}+(u^{(2)}+\Psi)D_h^{(2)}.\end{gathered} \tag{7a}$$

In this case, the second conditions are equivalent, up to the first terms in the expansions in Φ and Φ', to the conditions of continuity of the normal components of the electric fields,

$$\begin{gathered}E_0+E_0^s=D_0^{(1)}+D_0^{(2)},\\ E_h^s=D_h^{(1)}+D_h^{(2)}.\end{gathered} \tag{7b}$$

Simultaneously solving (5) and (7) we get

$$E_h^s=\frac{-2\Phi W^{(1)}W^{(2)}(u^{(2)}-u^{(1)})E_0}{\chi_h[W^{(2)}(u^{(1)}+\Phi)(u^{(2)}+\Psi+\Phi')-W^{(1)}(u^{(2)}+\Phi)(u^{(1)}+\Psi+\Phi')]} \tag{8}$$

where

$$W^{(i)}=u^{(i)2}-\Phi^2-\chi_0,\ i=1, 2.$$

Finally,

$$P_h^s=\left|\frac{E_h^s}{E_0}\right|\frac{\Phi'}{\Phi}. \tag{9}$$

1. POSITION OF DIFFRACTION MAXIMUM WITH ASYMMETRY

To investigate Eq. (6) and its roots, we change from the variable u to variable δ,

$$\delta=u-\sqrt{\Phi^2+\chi_0}. \tag{10}$$

This variable characterizes the deviation of the normal component of vector $\mathbf{k}_0$ from the normal component of the usual refracted vector k, corresponding to reflection without diffraction:

$$\delta=(k_{0z}-k_z)/\varkappa_0.$$

Using Eq. (10), we can write Eq. (6) as follows:

$$\delta(\delta+2\sqrt{\Phi^2+\chi_0})[\delta(\delta+2\sqrt{\Phi^2+\chi_0}+2\Psi)+\beta]-\chi_h\chi_{\bar h}=0, \tag{11}$$

where

$$\beta=\alpha-2\Psi(\Phi-\sqrt{\Phi^2+\chi_0}) \tag{12}$$

is the parameter of deviation from the exact Bragg angle for asymmetric diffraction. For the parameter β we have

$$\beta=(\sqrt{\Phi^2+\chi_0}+\Psi)^2-(\sqrt{\Phi'^2+\chi_0})^2. \tag{13}$$

For sufficiently large values of the angles Φ, $\Phi' \gg \Phi_0 = \sqrt{|\chi_0|}$, relation (12) becomes the standard expression of dynamic theory (see, e.g., Ref. 4):

$$\beta=\alpha-\chi_0\left(1\mp\frac{\Phi'}{\Phi}\right) \tag{14}$$

(the sign + corresponds to $\Phi + \Psi < 0$).

In contrast with (14), relation (12) gives a finite displacement of the diffraction maximum in the limit $\Phi \to 0$ from the position of the maximum of kinematic diffraction. This result is the same as that in Ref. 5, and comes from allowance for the phenomenon of total external reflection ofr the incident wave.

The character of Eq. (11) depends on the sign of the misorientation Ψ. If we regard this as a parametric equation depending on the parameters Φ and $\beta(\Phi')$ with fixed values of the misorientation Ψ, then when $\Psi > 0$ Eq. (11) contains two singular points, $\Phi = \Phi_0$ and $\beta = 0$, corre-

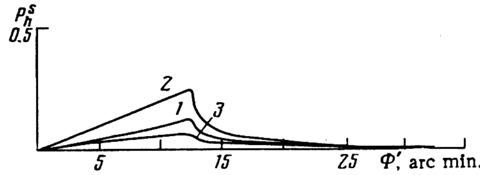
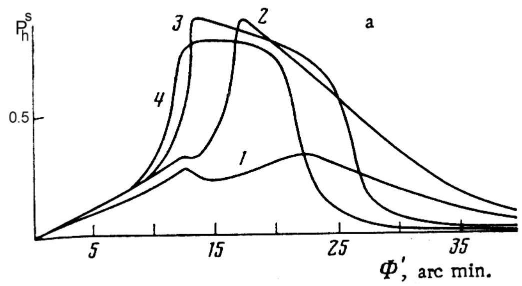


FIG 2. Diffraction pattern in Laue geometry for misorientation toward specularly reflected beam. Silicon, 220 reflections, Cu Kα radiation, φ = 30' (Ψ = 24'). 1) Φ = 5; 2) 10', 3) 15'.



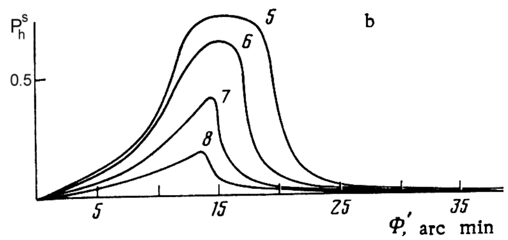


FIG. 3. Diffraction patterns in Bragg geometry for misorientation toward incident and specularly reflected diffracted beams; $\varphi = -30'$ ($\psi = -24'$). a) Formation of Darwin table with increasing angle of incidence: 1) $\Phi = 10$; 2) 13; 3) 17; 4) 20'. b) Transition from Bragg geometry to Laue geometry with further increase in angle of incidence (transition boundary $\Phi_{BL} = 27.5'$): 5) $\Phi = 22$; 6) 25; 7) 30; 8) 35'.

sponding to the boundary of total external reflection and the diffraction maximum; when $\Psi < 0$, another singular point arises in the equation, $\Phi = \sqrt{\Phi_0^2 + \Psi^2} \equiv \Phi_{BL}$. We shall now consider these two cases separately.

2. ASYMMETRIC LAUE DIFFRACTION WITH INCLINATION OF REFLECTING PLANES TOWARD SPECULARLY REFLECTED BEAM ($\Psi > 0$)

If $\Psi > 0$ (assuming also that $\Psi > \Phi_0$), then in view of Eq. (13) it is impossible to direct the incident beam onto the crystal in such a way that the exact diffraction condition ($\beta = 0$) and the condition of total external reflection are simultaneously satisfied for the diffracted wave ($\Phi' < \Phi_0$). For misorientation toward the specularly reflected beam ($\Psi > 0$) there is therefore a transition to asymmetric diffraction in the Laue geometry, accompanied by a rapid reduction in the intensity of the reflected diffracted wave in comparison with the case of symmetric diffraction in the Laue geometry ($\Psi = 0$), and when $\Psi \gg \Phi_0$ for any angles of incidence we shall have $P_h^S(\Phi, \Phi') \ll 1$ (Fig. 2).

3. ASYMMETRIC BRAGG DIFFRACTION WITH INCLINATION OF REFLECTING PLANES TOWARD INCIDENT AND SPECULARLY REFLECTED DIFFRACTED BEAM ($\Psi < 0$)

When $\Psi < 0$, the diffraction pattern becomes more complicated. As easily seen from Fig. 1, at angles of incidence $\Phi < \Phi_{BL}$ the normal component of the vector k_h is negative:

$$\mathrm{Re}\, k_{hz} = \mathrm{Re}\, k_{0z} + K_{hz} < 0 \quad (K_{hz} = K_h\varphi < 0).$$

The diffracted beam therefore emerges through the entry surface of the crystal, corresponding to diffraction in the Bragg geometry. In this range of angles of incidence Φ the wave $\varkappa_h^S$ is the normal (not specularly reflected) diffracted wave in vacuum.

Depending on the angle of incidence Φ we can distinguish three characteristic regions.

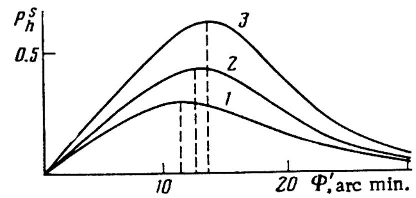


FIG. 4. Influence of small misorientations on intensity of curves of $P_h^s(\Phi')$ ($\Phi = 14'$). 1) $\Phi = 5$; 2) 0; 3) 5'.

1. For angles of incidence $0 < \Phi < \Phi_0$ the incident wave undergoes total external reflection, and correspondingly part of the intensity is transferred to the specularly reflected wave, and the intensity at the Bragg maximum is relatively low (Fig. 3, curve 1).

2. For angles of incidence $\Phi_0 < \Phi < \Phi_{BL} = \sqrt{\Phi_0^2 + \Psi^2}$ the intensity at the Bragg maximum may reach unity, and if $|\Psi| \gg \Phi_0$, then in this range of angles of incidence the curve of $P_h^S(\Phi')$ has a shape similar to a Darwin "table" with an angular width of several minutes (Fig. 3, curves 3-5).

3. For an angle of incidence $\Phi \approx \Phi_{BL}$ there is a transition to diffraction in the Laue geometry, and when $\Phi > \Phi_{BL}$ it is not the diffracted wave itself which reemerges from the crystal, but its specular component, the intensity of which rapidly decreases with increasing angle of incidence (Fig. 3, curves 7-8).

Thus we observe a continuous transition from diffraction in the Bragg geometry to diffraction in the Laue geometry for the same reflection plane as a result of a change in the angle of incidence of the x-rays on the crystal.

4. SENSITIVITY TO SMALL MISORIENTATIONS

The above results indicate that diffraction under conditions of total external reflection is uniquely sensitive to small deviations from the exact orientation of the reflecting planes. From the analysis in Secs. 2-3 it follows that the greatest differences should appear in the range of angles of incidence

$$\Phi_0 < \Phi < \Phi_{BL} = \sqrt{\Phi_0^2 + \Psi^2},$$

i.e., when $\Phi \approx \Phi_0$ (if $|\Psi| \ll \Phi_0$).

Figure 4 shows curves of P_h^S vs Φ' for $\Phi = 14'$, calculated for the 220 reflection in Cu $K\alpha$ radiation from single crystals of silicon ($\Phi_0 = 13.34'$) and misorientation of only 5' changes the intensity by 25%.

According to these data, in experiments on diffraction with total external reflection it is necessary to impose extra requirements on the precision of the orientation of the surface of the test crystals relative to the reflecting planes, and also on the precision of treatment of the surface. In particular, the discrepancies observed by Imamov et al.[3] between the theoretical calculations and experimental measurements are apparently due to slight deviations of the specimen surface from the (111) plane due to the cutting and treatment of the surface.

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