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X-ray Diffraction Under Specular Reflection Conditions. Ideal Crystals

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Abstract

The theory of X-ray diffraction by ideal crystals under the conditions when the incident and diffracted beams are directed at small angles to the entrance surface of a crystal has been developed. Besides the diffracted wave propagating inside the crystal (*i.e.* Laue-case diffraction) there are two specular reflected waves arising from incident and diffracted waves respectively. Such a diffraction scheme has been recently put into practice [Marra, Eisenberger & Cho (1979). *J. Appl. Phys.* **50**, 6927–6933]. It is found that at small glancing angles of incidence there exist directions in which the intensity of the reflected diffracted wave is close to the incident wave intensity, while both the specular reflected wave and diffracted wave intensities are close to zero. The analytical expressions are obtained for the diffraction curve shape. It is shown that for diffraction curve measurements high collimation through the glancing angle of incidence of X-rays on the crystal, Φ, is sufficient. There is no need to provide collimation through parameter α denoting deviation from exact Bragg conditions. Owing to the rigid relation between α, Φ and the angle of emergence of the reflected diffracted wave from the entrance surface of the crystal, Φ',

$$\Phi^2 = \alpha + \Phi'^2,$$

when measuring the intensity of the reflected diffracted wave as a function of Φ, the intensity is obtained as a function of α. Measurement of Φ' with the accuracy of about 30″ corresponds to accuracy through α of about 0·1″. These facts sufficiently simplify the performance of experiment and open wide prospects for studies of crystal structure of thin subsurface layers with unique accuracy.

1. Introduction

The use of extremely asymmetric X-ray diffraction techniques requires the account of the specular reflection phenomenon. This problem has been studied in detail for both Bragg-case (Farwig & Schürmann, 1967; Kishino, 1971; Rustichelli, 1975) and Laue-case diffraction (Farwig & Schürmann, 1967; Kishino, Noda & Kohra, 1972; Bedynska, 1973, 1974; Härtwig, 1976, 1977). In the Laue case the specular reflection effect would essentially increase the intensity of anomalously transmitted waves in the T beam. In the Bragg case a decrease in penetration depth due to specular reflection leads to an appreciable increase in the integral reflection coefficient with the position and shape of the Bragg peak being essentially changed. Consideration of the specular reflection phenomenon does not appear to be restricted only to asymmetric diffraction schemes.

A new diffraction scheme has been recently described (Marra, Eisenberger & Cho, 1979). In this scheme, the incident-beam glancing angle was chosen in such a way as to allow the Laue-case diffraction condition to be realized. On the other hand, both the incident and diffracted beams made small angles with

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the crystal surface (see Fig. 1). Not only the incident but also the diffracted beams were reflected from the crystal. The angular dependence of the reflected diffracted beam contains valuable information on the crystal structure of the thin subsurface layers.

The present paper deals with the detailed analysis of diffraction scattering in such geometry for perfect crystals.

2. General considerations

We suppose the plane monochromatic wave of amplitude $\mathbf{E}_0$ and wave vector $\mathbf{x}_0$ to be incident on the crystal surface at a small glancing angle Φ. Keeping Φ small, one can choose the direction of incidence to provide strong diffraction scattering in a crystal from planes perpendicular to the surface of the crystal (see Fig. 1). At the same time, the diffracted wave angle with the entrance surface Φ' is also small. As a result, owing to the specular reflection, both the incident and diffracted waves are reflected from the crystal. The amplitudes of these waves are denoted by $\mathbf{E}_0^S$ and $\mathbf{E}_h^S$, respectively. As mentioned above, the X-ray wave field should consist of three waves at the entrance surface of a crystal:

$$\mathbf{E}(\mathbf{r}) = \exp(i\mathbf{x}_0\mathbf{r}) + \mathbf{E}_0^S\exp(i\mathbf{x}_0^S\mathbf{r}) + \mathbf{E}_h^S\exp(i\mathbf{x}_h^S\mathbf{r}), \quad (1)$$

where $\mathbf{x}_0^S$ and $\mathbf{x}_h^S$ are the wave vectors of the incident and diffracted waves respectively, being specular reflected.

Assuming $\Phi^2 \ll 1$, we have

$$\begin{aligned} x_{0z} &= x_0\Phi = -(x_0^S)_z \\ (x_h^S)_z &= -x_0\Phi'. \end{aligned} \quad (2)$$

The angles Φ and Φ' are connected by the equation

$$\Phi^2 = \Phi'^2 + \alpha, \quad (3)$$

where α is a parameter denoting the deviation from exact Bragg conditions:

$$\alpha = \frac{(\mathbf{x}_0 + \mathbf{K}_h)^2 - x_0^2}{x_0^2}. \quad (4)$$

Here $\mathbf{K}_h$ is the reciprocal-lattice vector.

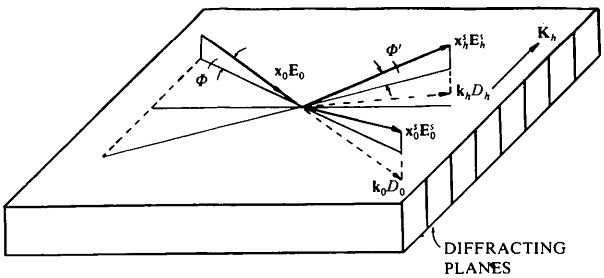

Fig. 1. The scheme of diffraction scattering of X-rays under specular reflection conditions.

Inside the crystal, the field consists of incident and diffracted waves:

$$\mathbf{D}(\mathbf{r}) = \mathbf{D}_0\exp(i\mathbf{k}_0\mathbf{r}) + \mathbf{D}_h\exp(i\mathbf{k}_h\mathbf{r}). \quad (5)$$

In the general case, the complex wave vector $\mathbf{k}_0$ differs from the vacuum wave vector $\mathbf{x}_0$ by a small value at normal $\mathbf{n}$ to the crystal surface:

$$\mathbf{k}_0 = \mathbf{x}_0 + \mathbf{n}x_0\delta. \quad (6)$$

$\mathbf{k}_h$ differs from $\mathbf{k}_0$ by the reciprocal-lattice vector $\mathbf{K}_h$:

$$\mathbf{k}_h = \mathbf{k}_0 + \mathbf{K}_h.$$

The value δ and the relationships between the amplitudes $\mathbf{D}_0$ and $\mathbf{D}_h$ are determined from the fundamental equations of the dynamic theory [for instance, see Pinsker (1978)], which can be written in a more convenient form as follows:

$$\begin{aligned} \frac{x_{0z}^2 - k_{0z}^2}{x_0^2} D_0 &= -\chi_0 D_0 - \chi_{\bar{h}} D_h \\ \left(\frac{x_{0z}^2 - k_{0z}^2}{x_0^2} + \alpha\right) D_h &= -\chi_h D_0 - \chi_0 D_h. \end{aligned} \quad (7)$$

For the sake of simplicity, these scalar equations are written for σ-polarized waves, *i.e.* for waves with the induction vectors perpendicular to the scattering plane formed by $\mathbf{x}_0$ and $\mathbf{x}_h^S$. In (7), χ_0, χ_h, $\chi_{\bar{h}}$ are the Fourier coefficients of electric susceptibility, the parameter α is defined from (4). The condition of consistency of set (7) gives the secular equation for determination of δ:

$$w(w - \alpha) - \chi_h\chi_{\bar{h}} = 0, \quad (8)$$

where

$$w = -\left(\frac{x_{0z}^2 - k_{0z}^2}{x_0^2} + \chi_0\right) \equiv \delta^2 + 2\delta\Phi - \chi_0. \quad (9)$$

Further, it is convenient to determine, from secular equation (8), not the δ value, as is usually the case, but the value u defined as follows:

$$k_{0z} = ux_0. \quad (10)$$

As a result we have

$$\begin{aligned} u^{(j)} &= \pm(\Phi^2 + w^{(i)} + \chi_o)^{1/2} \\ w^{(i)} &= \alpha/2 \pm (\alpha^2/4 + \chi_h\chi_{\bar{h}})^{1/2}, \end{aligned} \quad (11)$$

where $i = 1, 2$ and $j = 1, 2, 3, 4$. In the case of a thick crystal it is reasonable to consider only the waves with the in-depth-decreasing amplitudes. So we are interested in roots $u^{(j)}$ with

$$\operatorname{Im} u^{(j)} > 0. \quad (12)$$

To determine the E_0^S and E_h^S amplitudes, one has to use not only the wave-field-continuity conditions

$$\begin{aligned} E_0 + E_0^S &= D_0^{(1)} + D_0^{(2)} \\ E_h^S &= D_h^{(1)} + D_h^{(2)} \end{aligned} \quad (13)$$

but also the continuity conditions of the amplitude derivatives which are equivalent to the conditions of continuity of tangential components of the magnetic field vector:

$$\begin{aligned} \Phi(E_0 - E_0^S) &= u^{(1)} D_0^{(1)} + u^{(2)} D_0^{(2)} \\ -\Phi' E_h^S &= u^{(1)} D_h^{(1)} + u^{(2)} D_h^{(2)}. \end{aligned} \tag{14}$$

The amplitudes $D_0^{(i)}$ and $D_h^{(i)}$ are connected by dynamic equations (7).

From (7), (13), (14), it is easy to obtain

$$\begin{aligned} E_h^s = &-2\Phi\chi_h(u^{(2)} - u^{(1)}) \\ &\times [w^{(2)}(u^{(1)} + \Phi)(u^{(2)} + \Phi') \\ &- w^{(1)}(u^{(1)} + \Phi')(u^{(2)} + \Phi)]^{-1} E_0. \end{aligned} \tag{15}$$

Finally, for the intensity of a diffracted wave specular reflected from the entrance surface of a crystal we obtain

$$\begin{aligned} P_h^S &= (|E_h^S|^2/|E_0|^2) \times \Phi'/\Phi \\ &= 4\, \Phi\Phi' \,|\chi_h|^2 |u^{(2)} - u^{(1)}|^2 \\ &\times |w^{(2)}(u^{(1)} + \Phi)(u^{(2)} + \Phi') \\ &- w^{(1)}(u^{(2)} + \Phi)(u^{(1)} + \Phi')|^{-2} \end{aligned} \tag{16}$$

Equations (16) and (11) provide the complete solution of the problem of finding the reflection coefficient of an appropriate wave.

3. The analysis of a diffraction pattern

The reflection coefficient P_h^S of the diffracted wave from the entrance surface of a crystal has an appreciable value only when the glancing angle $\Phi \sim \Phi_0$, $\Phi_0 = (|\chi_0|)^{1/2}$ and parameter $\alpha \sim |\chi_h|$. Obviously, owing to small Φ the depth of X-ray penetration into a crystal should be small; therefore the curve $P_h^S(\alpha)$ provides information on the crystal structure of relatively thin subsurface layers.

Let us begin the analysis with the case when $|\alpha| \gg |\chi_h|$, *i.e.* far from exact Bragg conditions. In this case for roots $u^{(1,2)}$ one has approximately

$$\begin{aligned} u^{(1)} &= (\Phi^2 + \chi_0)^{1/2} \\ u^{(2)} &= (\Phi'^2 + \chi_0)^{1/2}. \end{aligned} \tag{17}$$

Taking into account (17), the following equation for reflection coefficient P_h^S is obtained:

$$P_h^S = \frac{4|\chi_h|^2}{(\Phi + \Phi')^2} \frac{\Phi}{|(\Phi^2 + \chi_0)^{1/2} + \Phi|^2} \times \frac{\Phi'}{|(\Phi'^2 + \chi_0)^{1/2} + \Phi'|^2}. \tag{18}$$

From (18) and (3) it appears that with the deviation from the exact Bragg condition the reflected diffracted wave intensity sharply decreases. It can be easily seen that (18) is valid only when either Φ or Φ' are much greater than Φ_0. For exact Bragg condition $\alpha = 0$ and therefore $\Phi' = \Phi$, then

$$\begin{aligned} P_h^s(\alpha = 0) = |\,&\Phi[(\Phi^2 + \chi_0 + \chi_h)^{1/2} - (\Phi^2 + \chi_0 - \chi_h)^{1/2}] \\ &\times [(\Phi^2 + \chi_0 + \chi_h)^{1/2} + \Phi]^{-1} \\ &\times [(\Phi^2 + \chi_0 - \chi_h)^{1/2} + \Phi]^{-1}|^2. \end{aligned} \tag{19}$$

The corresponding curve $P_h^S(\Phi)$ is shown in Fig. 2. The calculations are made for the 220 reflection of Cu $K\alpha$ radiation from a germanium crystal, with allowance for absorption in the crystal.

It should be noted that the intensity of the reflected diffracted wave reaches a value close to unity for the angle $\Phi = (|\chi_0 - \chi_h|)^{1/2}$. If we neglect the imaginary parts of coefficients χ_0 and χ_h, the following expression is obtained from (19) at the glancing angle of incidence $\Phi = (|\chi_0 - \chi_h|)^{1/2}$:

$$P_h^S(\alpha = 0) = \left|\frac{2\chi_h}{\chi_0 + \chi_h}\right|. \tag{20}$$

Since for the 220 reflection χ_h slightly differs from χ_0, for P_h^S one obtains a value close to unity. At the same time the intensity of the specular reflected wave is small. Neglecting the absorption of X-rays in the crystal, one has (see formulae 13, 14):

$$P_0^S = \left|\frac{\chi_0 - \chi_h}{\chi_0 + \chi_h}\right|. \tag{21}$$

Thus, a rather interesting physical phenomenon takes place, that is a strong suppression of specular reflected wave intensity. In the case far from the exact Bragg condition the specular reflected wave intensity is close to unity for the same glancing angle of incidence $\Phi = (|\chi_0 - \chi_h|)^{1/2}$.

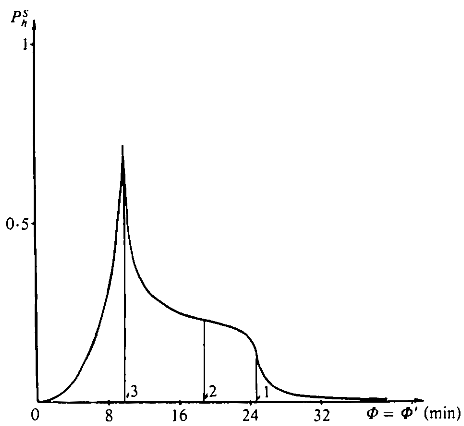

Fig. 2. The intensity of a specular reflected diffracted wave from the entrance surface of a Ge crystal (Cu $K\alpha$ radiation, 220 reflection) as a function of $\Phi = \Phi'$ for exact Bragg condition, $\alpha = 0$. 1, 2, 3 indicate the following values of Φ: (1) $\Phi = (|\chi_0 + \chi_h|)^{1/2}$; (2) $\Phi = (|\chi_0|)^{1/2} = \Phi_0$; (3) $\Phi = (|\chi_0 - \chi_h|)^{1/2}$.

Rather strong intensity of reflected diffracted wave is retained in the wide angular range $(|\chi_0 - \chi_h|)^{1/2} \le \Phi \le (|\chi_0 + \chi_h|)^{1/2}$. The value P_h^S sharply decreases outside this angular region.

In all diffraction experiments the dependence of the diffracted wave intensity on the parameter α is required to determine crystal structure parameters and coefficients of susceptibility (χ_0, χ_h). As in usual cases of normal incidence, in the given case the diffraction scattering disappears with the deviation from the exact Bragg condition by about some tens of seconds of arc. Usually, high collimation of the incident beam, providing small values of the parameter α, is required for diffraction studies in crystals.

In the considered scheme, besides the above collimation rather narrow beams are indispensable in the vertical plane to ensure small glancing angles of incidence. Here the requirements for collimation are not so rigid and are of the order of several minutes of arc. However, ensuring a twice collimated beam through α and Φ is a complicated technical problem. From this fact it might seem that the theoretical analysis carried out in the present paper has a purely academic character and that the realization of such experiments can be expected only in the future. However, there is no need to provide horizontal collimation through α owing to the connection between α and Φ' (see formula 3). In fact, when measuring the intensity of the reflected diffracted wave as a function of Φ' (which varies at distances of about the total external reflection angle, *i.e.* some minutes of arc) one actually measures the intensity as a function of α. Typical curves of P_h^S as a function of Φ' (and consequently of α) are shown in Fig. 3. The segments in Fig. 3 present the variations of α in seconds of arc corresponding to variations of Φ' in the band of 4′. For example, the variation of Φ' in the band 8–12′ corresponds to variation of α by about 1·4″.

In the angular range of incidence $(|\chi_0 - \chi_h|)^{1/2} \le \Phi \le (|\chi_0 + \chi_h|)^{1/2}$, a strong intensity of the reflected diffracted wave is observed, with distinct peculiarities at the values of Φ' corresponding to the exact Bragg condition $\alpha = 0$. For instance, the curve 3, corresponding to the glancing angle of incidence $\Phi = (|\chi_0 - \chi_h|)^{1/2}$ has a narrow maximum near $\alpha = 0$, and with the deviation from $\alpha = 0$ by fractions of a second the intensity P_h^S sharply decreases. However, even these distinct peculiarities can be, in principle, measured experimentally.

As follows from (3), the variation of angle Φ' by the value $\Delta\Phi'$ corresponds to the variation of α by the value

$$\Delta\alpha = 2\Phi' \Delta\Phi'. \qquad (22)$$

As shown in a paper (Kov'ev & Matveev, 1981) on studies of total external reflection, it is rather easy to carry out measurements of Φ and Φ' with the accuracy of about some tens of seconds of arc. Taking into account the fact that Φ' itself is of the order of 10^{-3} min it can be easily seen from (22) that here appears an opportunity to carry out precision measurements of diffraction curves through α with an accuracy exceeding 0·01″. It is this fact that opens up wide prospects for experimental application of the considered technique.

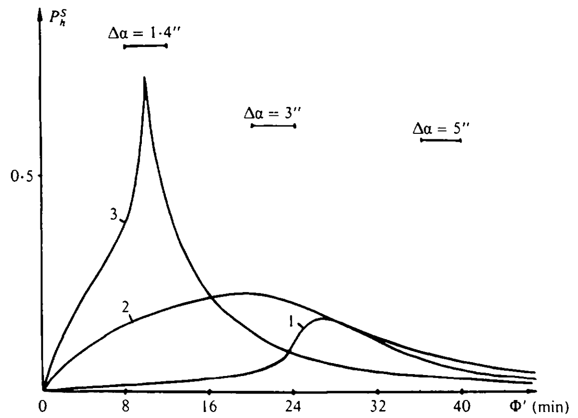

Fig. 3. The intensity of a specular reflected diffracted wave from the entrance surface of a Ge crystal (Cu $K\alpha$ radiation, 220 reflection) as a function of Φ' (and consequently of α) for the following values of glancing angle of incidence Φ: (1) $\Phi = (|\chi_0 + \chi_h|)^{1/2}$; (2) $\Phi = \Phi_0$; (3) $\Phi = (|\chi_0 - \chi_h|)^{1/2}$. The segments present the variation of α by the value $\Delta\alpha$ in s of arc corresponding to the variation of Φ' in the band of 4′. The variation of Φ' in bands 8–12; 20–24; 36–40′ is equivalent to $\Delta\alpha$ of about 1·4; 3; 5″ respectively (see formula 22).

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