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On the Integrated Reflectivity of Perfect Crystals in Extremely Asymmetric Bragg Cases of X-ray Diffraction

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Abstract

In the case of grazing incidence, the integrated reflectivity calculated according to the theory appropriate for extreme asymmetry is smaller than that calculated according to the conventional theory. In the case of grazing emergence the results calculated according to each theory exhibit smaller differences than for the first case within a great range of extreme asymmetry. In the conventional theory the integrated reflectivity tends to zero for $\alpha - \theta_B$ tending to zero, whereas the theory for extremely asymmetric cases provides a non-zero value for the reflectivity for $\alpha - \theta_B$ tending to zero.

1. Introduction

The problem of the asymptotic behaviour of the integrated reflectivity of perfect and imperfect crystals has been discussed in some recent works. Mathieson (1976, 1977) has proposed that the very asymmetric Bragg case offers an experimental means of deriving extinction-free structure factors by extrapolation of integrated reflectivity data to the asymptotic limits. Wilkins (1978, 1980) systematically explored the variation of X-ray Bragg reflection properties of perfect and imperfect crystals with thickness and degree of asymmetry of reflection by direct numerical evaluation of the dynamical theory. In particular he showed that well-defined universal limits exist where the integrated reflectivity of an ideally imperfect crystal asymptotically approaches that of a perfect crystal of the same material under the same diffraction conditions. These calculations were based on the conventional dynamical theory. This means that they do not necessarily apply in the extremely asymmetric ranges where the conventional theory provides results which are either only partly correct or completely incorrect. The aim of the present work is to show the behaviour of the integrated reflectivity of a perfect crystal in the two extreme asymmetric limits which are characterized by grazing incidence and grazing emergence.

2. Theory

The integrated reflectivity of a perfect crystal on the glancing-angle scale is defined by

$$\rho_p^\theta = \int R_h(\Delta\theta)\,\mathrm{d}(\Delta\theta). \tag{1}$$

Here,

$$R_h(\Delta\theta) = \left| \frac{\sqrt{\chi_h/\chi_{\bar h}}}{\sqrt{\gamma_0/|\gamma_h|}} \frac{E_h^{(a)}}{E_0^{(a)}} \right|$$

is the reflectivity on the glancing-angle scale,

$$\begin{aligned} \gamma_0 &= \sin(\theta_B + \Delta\theta + \alpha) \\ \gamma_h &= \sin(\theta_B + \Delta\theta - \alpha), \end{aligned} \tag{2}$$

$\theta = \theta_B + \Delta\theta$ is the glancing angle, χ_h, $\chi_{\bar h}$ are the Fourier coefficients of the electric susceptibility, θ_B is the Bragg angle, α is the angle between reflecting lattice planes and crystal surface, $\Delta\theta$ is the departure of the incident beam from the exact Bragg law, $E_0^{(a)}$ is the amplitude of the incident wave and $E_h^{(a)}$ is the amplitude of the diffracted wave outside the crystal.

A more accurate form of the dynamical theory appropriate also in extremely asymmetric cases was first developed by Farwig & Schürmann (1967). Equivalent formulations and first calculations have been published by Kishino & Kohra (1971) and Bedyńska (1973).

The present calculations of the reflectivity $R_h(\Delta\theta)$ were carried out numerically according to the formulation of Bedyńska (1973), taking into account the results of Härtwig (1978*a*), using without further approximations the fundamental equation for the two-beam case in the form

$$\left.\begin{aligned} \left(\frac{K_{0i}^2 - k^2}{k^2} - \chi_0\right) E_{0i} - \chi_{\bar h} E_{hi} = 0 \\ -\chi_h E_{0i} + \left(\frac{K_{hi}^2 - k^2}{k^2} - \chi_0\right) E_{hi} = 0 \end{aligned}\right\}; \tag{3}$$

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the related equation of the dispersion surface; the boundary condition for the wave vectors; the four solutions g_i of the equation

$$g^4 + bg^3 + cg^2 + dg + e = 0; \qquad (4)$$

and the two boundary conditions for the amplitudes at the crystal surface. Here, $\mathbf{K}_{0i}$, $\mathbf{K}_{hi}$ are the wave vectors of the wave fields inside the crystal, $\mathbf{k}$ is the wave vector of the incident wave, g_i is the *Anpassungsfehler* defined by $\mathbf{K}_{0i} = \mathbf{k} - \mathbf{n}g_i$ k, b, c, d, e are functions of α, θ_B, $\Delta\theta$, χ_0, χ_h and $\chi_{\bar{h}}$, $\mathbf{n}$ is the unit vector normal to the entrance surface, $i = 1, 2, 3, 4$ is the index of the wave fields.

The integration of (1) was also carried out numerically.

3. Results

The integrated reflectivity as a function of asymmetry was calculated for a special case: The 220 reflection of silicon, Cu $K\alpha_1$ radiation, $d = 1$ mm (thick crystal) and σ polarization. From these results the general behaviour of the integrated reflectivity in the two extreme asymmetric limits can be seen. For comparison, calculations were carried out also according to the conventional theory [with the anomalous dispersion parameter $\kappa = 0{\cdot}0347$, the parameter $g = -0{\cdot}0180 \times (\gamma_0 + |\gamma_h|)/\sqrt{\gamma_0/|\gamma_h|}$, the mean path length $A \simeq 15/\sqrt{\gamma_0/|\gamma_h|}$ and $d\theta/dy = 1{\cdot}323 \times 10^{-5}\sqrt{|\gamma_h|/\gamma_0}$ (Afanas'ev & Perstnev, 1969; Hirsch & Ramachandran, 1950; Pinsker 1974)].

3.1. *Grazing incidence*

Fig. 1 shows the integrated reflectivities ρ_p^θ calculated according to the conventional theory and to the theory appropriate for extreme asymmetry, as a function of $\alpha + \theta_B$ for the case of grazing incidence. For angles $\alpha + \theta_B < 0{\cdot}02$ rad ($\sim 1°$), the curve according to the theory for extreme asymmetry departs increasingly from that according to the conventional theory. After reaching its maximum at about $\alpha + \theta_B \simeq 6 \times 10^{-3}$ rad ($\sim 0{\cdot}34°$), it decreases with increasing asymmetry.

These considerable departures result mainly from three origins:

1. A strong specularly reflected wave E_m appears, which in comparison with the conventional theory leads to smaller amplitudes E_h (Kishino & Kohra, 1971; Brümmer, Höche & Nieber, 1976; Härtwig, 1978*c*).

2. The form of the dispersion surface changes distinctly, which considerably influences the departure from the Bragg law owing to refraction and the incident half-width of the reflection pattern (Rustichelli, 1975; Mazkedian & Rustichelli, 1975; Brümmer *et al.*, 1976).

3. The definition of the incident-beam direction cosine changes from $\gamma_0 = \sin(\theta_B + \alpha)$, which is an approximation holding as long as $|\theta_B + \alpha| \gg |\Delta\theta|$ (conventional theory), to that given in (2).

3.2. *Grazing emergence*

Fig. 2 shows the integrated reflectivities ρ_p^θ calculated according to the conventional theory and to the theory appropriate for extreme asymmetry, as a function of $\alpha + \theta_B$ for the case of grazing emergence. Within nearly the whole Bragg range the differences between the two curves are much smaller than in the case of grazing incidence. In the range of about $-5 \times 10^{-3} \lesssim \alpha - \theta_B \lesssim -2 \times 10^{-4}$ rad ($\sim -0{\cdot}7'$), the integrated reflectivity calculated according to the theory appropriate for extreme asymmetry is smaller than that calculated according to the conventional theory and for greater values of $\alpha - \theta_B$ it exceeds the latter. This behaviour results mainly from two sources:

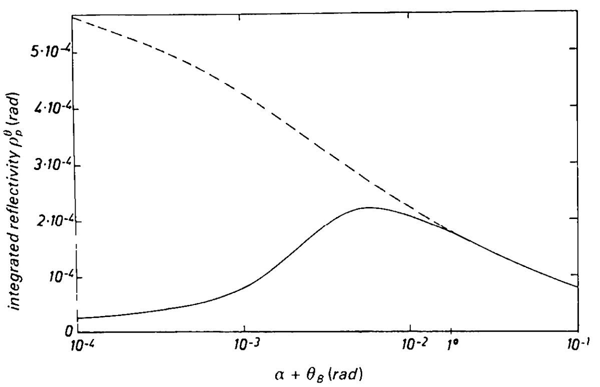

Fig. 1. Integrated reflectivity calculated according to the theory appropriate for extreme asymmetry (solid line) and according to the conventional theory (dashed line) in the case of grazing incidence. Amplitude of the incident plane waves $E_0^{(a)} = 1$.

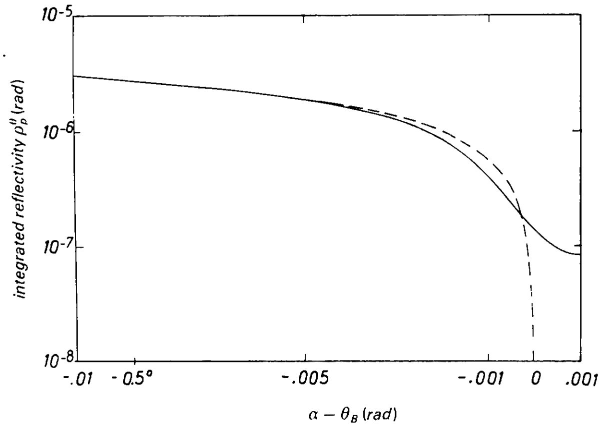

Fig. 2. Integrated reflectivity calculated according to the theory appropriate for extreme asymmetry (solid line) and according to the conventional theory (dashed line) in the case of grazing emergence. Amplitude of the incident plane waves $E_0^{(a)} = 1$.

1. At the exit surface a second strong diffracted wave appears, the wave E_{hd}. This leads, in comparison with the conventional theory, to smaller amplitudes E_h (Härtwig, 1978*b*). But, on the other hand, the diffracted wave E_h has a considerable amplitude still in the range of an extremely asymmetric Laue case with grazing emergence (Kishino, Noda & Kohra, 1972; Bedyńska, 1973, 1974), where the conventional theory predicts no E_h. Therefore, in the conventional theory the integrated reflectivity tends to zero for $\alpha - \theta_B$ tending to zero, whereas the theory for the extremely asymmetric case provides a non-zero value for $\alpha - \theta_B$ tending to zero.

2. The definition of the emerging-beam direction cosine changes from $\gamma_h = \sin(\theta_B - \alpha)$, which is also an approximation holding as long as $|\theta_B - \alpha| \gg |\Delta\theta|$, to that given in (2). But in the case of grazing emergence this change leads to an increase of the integrated reflectivity compared with the result of the conventional theory. So two opposite tendencies act now on the integrated reflectivity.

The consequences of the changes of the form of the dispersion surface are not so important now, because in the case of grazing emergence the departures from the exact Bragg law for the physically interesting region (*i.e.* for the maximum of the reflection curve) are much smaller than in the case of grazing incidence (now $\Delta\theta \simeq 10^{-5}$ rad and not $\sim 10^{-3}$ rad), but they too cannot be neglected (Härtwig, 1978*b*).

Despite the fact that the calculations were carried out for a special case, the obtained results may be generalized, because the discussed properties are independent of the chosen conditions.

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X-ray Diffuse Scattering in Dicalcium Barium Propionate

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Abstract

Diffuse X-ray intensities have been measured in the *hk*0 reciprocal plane of cubic dicalcium barium propionate, $Ca_2Ba(C_2H_5COO)_6$, with a diffractometer. The observed streaks run parallel to [$\bar{1}$10], passing through reciprocal-lattice points with $h + k = 8n$. Intensity profiles in two directions perpendicular to the streaks were measured and fitted theoretically under the assumption of one-dimensional Markov-chain-type correlations; the agreement between theory and experimental data is excellent, particularly for the [110] direction, giving correlation lengths of 24·5 (5) Å along [110] and 4·6 (1) Å along [001]. These lengths are compared with the crystal structure and suggest a model in which there are ordered domains elongated along [110] but rather short along [001].

Introduction

Recently, Stadnicka & Glazer (1980), hereafter SG, reported an accurate structure determination of di-

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