

## Determination of Structure Factors by the Pendellosung Effect for Imperfect Crystals

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### Abstract

An investigation of thickness dependence of the X-ray integrated intensity of monochromatic linearly polarised X-ray radiation in a symmetrical Laue-case diffraction reveals that the method of determination of structure factors  $f$  from thickness oscillations can be extended to real crystals with a static distribution of Coulomb-type defects or dislocations. The  $fE$  value (where  $E$  is the static Debye-Waller factor) is determined from the oscillation period, while  $E$  is determined from the tilt-angle tangent of the oscillation axis of the reduced integrated intensity.

### 1. Introduction

The experimental study of electron density distributions aimed at determining the character of a chemical bond and peculiarities of an electronic crystal structure calls for the precise measurement of structure factors. As is known, the conventional methods based on measurements of integrated intensity of X-ray scattering by poly-crystal or single crystal samples in many cases do not allow one to determine the structure factors with sufficient accuracy. The precise measurement of structure factors can be performed on perfect crystals by employing the dynamic effect of thickness oscillations of intensity scattering in Laue-case diffraction (the Pendellösung effect) (Kato and Lang 1958; Hattori *et al.* 1965; Aldred and Hart 1973). The determination of structure factors by this method amounts only to the measurement of the oscillation period. For perfect crystals the above-mentioned effect is exhibited both in monochromatic and white radiation. The atomic scattering factors of silicon and germanium as a function of X-ray wavelength have been determined by the intensity beats of the polychromatic Pendellösung effect (Aristov *et al.* 1977; Takama *et al.* 1980, 1981). The method of sample inclination (Lawrence and Mathieson 1977) makes it possible to use the diffractometry procedure for studying dynamic thickness intensity oscillations (Somenkov *et al.* 1978; Olekhnovich *et al.* 1982).

Although the methods based on thickness oscillations are quite simple and reliable, and as the determination of the  $f$  factors by them is practically reduced to geometric measurement of the period, they have not found wide use since perfect crystals are obtained at present only for a very limited number of materials. A statistical dynamical theory of diffraction in real crystals has been developed by Kato (1980).

The present work shows the possibility of determining structure factors from the X-ray integrated intensity thickness oscillations for real crystals containing statistically distributed Coulomb-type defect clusters, dislocation loops or dislocations.

## 2. Experimental Procedure

A dynamical diffraction theory for single crystals with a statistical distribution of Coulomb-type defects has been developed (Gudzenko *et al.* 1983) with respect to coherent and diffuse scattering. In accordance with this theory the character of the diffuse component thickness dependence in Laue-case diffraction depends on the crystal thickness. In the range of a thin crystal ( $\mu t < 1$ , where  $\mu$  is the photoelectric absorption coefficient and  $t$  is the crystal thickness), the diffuse component is described by the kinematical theory, while in the range of a thick crystal ( $\mu t \gg 1$ ) the dynamical effect of anomalous transmission for diffuse scattering is revealed. This theory has been confirmed experimentally (Olekhovich *et al.* 1984).

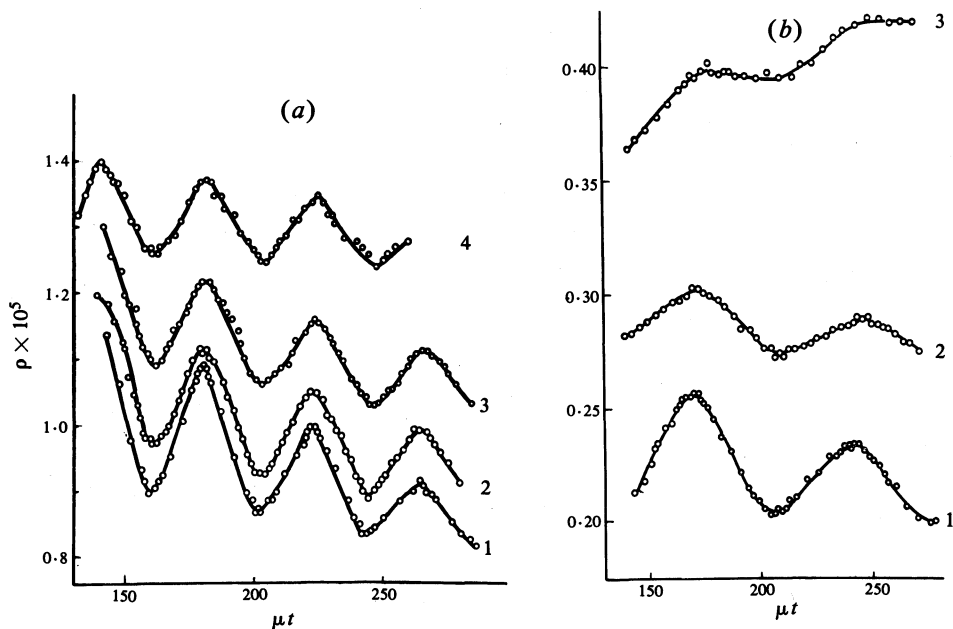


Fig. 1. Integrated intensity thickness dependence of reflections (a) 111 and (b) 333 for Si single crystals at the following  $N_d$  values: (1),  $8 \times 10^2$ ; (2),  $3 \times 10^3$ ; (3),  $1 \times 10^4$ ; and (4),  $2 \times 10^4 \text{ cm}^{-2}$ .

The same character of the thickness dependence of the X-ray integrated intensity in Laue-case diffraction has been found for crystals with a statistical distribution of low density dislocations  $N_d$  (lower than  $10^4$ – $10^5 \text{ cm}^{-2}$ ) (Olekhovich *et al.* 1983; Olekhovich 1986). In the range of a thin crystal the dynamic effect of the X-ray integrated intensity thickness oscillations has been observed. Fig. 1 illustrates the observed thickness dependence of the integrated intensity for the Si reflections 111 and 333 measured in linearly polarised monochromatic Mo  $K\alpha$  radiation. As can be seen the period of thickness oscillations increases with the dislocation density growth. The absolute value of the integrated intensity of the Bragg reflections also grows. Such behaviour of the integrated intensity results from a change in the relation between the coherent and diffuse components of scattering. This relation is determined by the static Debye–Waller factor. Its values, determined from changes in the thickness oscillations period and from the growth in the integrated intensity,

coincide (Olekhovich 1986). This implies that the X-ray integrated intensity for weakly distorted crystals with a statistical distribution of dislocations, as well as the Coulomb-type defects, can be described in terms of coherent and diffuse components. In the case of a thin crystal, when the effect of anomalous transmission does not appear, the expression for the integrated intensity in symmetric Laue-case diffraction of linearly polarised radiation can be written as (Gudzenko *et al.* 1983)

$$\rho = (1 - E^2)\rho_k + \frac{\rho_k E}{2A} \int_0^{2AE} I_0(x) dx, \quad (1)$$

where the first term defines the diffuse component, the second the coherent component of scattering,  $E$  is the static Debye-Waller factor,  $\rho_k$  is the integrated intensity calculated in terms of the kinematical theory,  $A = r_0 \lambda |f| t / V \cos \theta_B$  is the reduced thickness of the crystal (the notation has the usual meaning here), and  $I_0$  is the Bessel function of zero order of the first kind.

From the analysis (1) it follows that the thickness oscillation period for a real crystal increases by a factor  $A$  when compared with that for a perfect crystal:

$$A = \frac{\pi V \cos \theta_B}{r_0 \lambda |f| E}. \quad (2)$$

The parameters in equations (1) and (2) are calculated at a given temperature with the thermal Debye-Waller factor. The thermal diffuse scattering contribution to the measured integrated intensity is rather small due to narrow reflections for crystals with low density dislocations. One can see that for determination of structure factors from the measured integrated intensity oscillation period it is necessary to obtain first the value of the static Debye-Waller factor. With this aim in view we consider the reduced value of the integrated intensity:

$$\rho_r = \frac{\rho}{\rho_k} 2A = (1 - E^2)2A + E \int_0^{2AE} I_0(x) dx. \quad (3)$$

As seen from (3) the reduced value of the diffuse component increases linearly with the crystal thickness, while the coherent one is an oscillating function. It follows that  $\rho$  oscillates around the axis, with the tilt-angle tangent being  $2(1 - E^2)$  (for the perfect crystal the oscillation axis is parallel to the abscissa). On determining the tilt-angle tangent of the oscillation axis one can find the value of the Debye-Waller factor.

Fig. 2 represents the thickness dependence of the reduced value of the measured integrated intensity for Si reflections 111 and 333, with a dislocation density of  $N_d = 8 \times 10^2 \text{ cm}^{-2}$ . It can be noted that the tilt angle of the oscillation axis increases with the reflection order. The values of the static Debye-Waller factor determined from the tilt-angle tangent of the oscillation axis are listed in Table 1. The oscillating period  $A$  was determined by the extrema positions of the reduced value of the integrated intensity (see Table 1).

From the values obtained for the oscillation period and the static Debye-Waller factor the atomic scattering factors  $f$  (Table 1) were determined employing (2). The values of  $f$  obtained from the thickness oscillation data for real crystals coincide (within experimental error) with the values obtained for a perfect crystal (Aldred and Hart

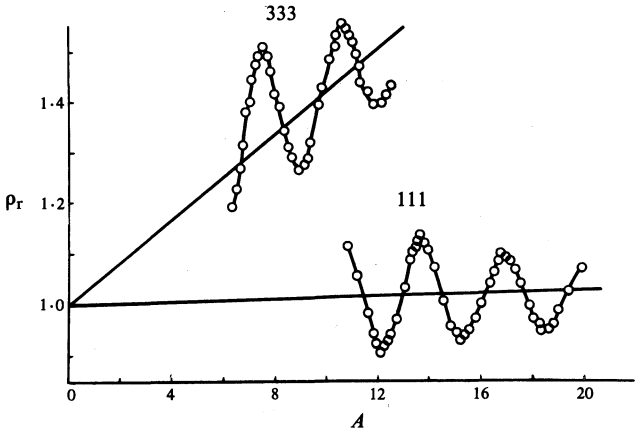


Fig. 2. Thickness dependence of the reduced integrated intensity value for Si reflections 111 and 333 at  $N_d = 8 \times 10^2 \text{ cm}^{-2}$ .

Table 1. Oscillation periods, static Debye–Waller factors and atomic scattering factors for Si reflections 111 and 333 ( $N_d = 8 \times 10^2 \text{ cm}^{-2}$ ) from the data measured in Mo K $\alpha$  radiation at 20°C

<i>hkl</i>	<i>A</i> (10 <sup>3</sup> nm)	<i>E</i>	<i>f</i>	
			Present work	Aldred–Hart (1973)
111	41.3	0.9994	10.70	10.703
333	71.9	0.990	5.87	5.867

1973). The accuracy of the *f* factor determination both for imperfect and perfect crystals depends mainly on the experimental error in the oscillation period, since the factor *E* can be determined with an error of less than 0.1%. This is due to the fact that *E* differs slightly from unity and the tilt-angle tangent of the oscillation axis of the reduced integrated intensity is proportional to  $(1 - E^2)$ . Consequently, to achieve the above-mentioned accuracy in determining *E*, it is sufficient to measure the tilt-angle tangent of the oscillation axis with an accuracy of up to 10%.

3. Conclusions

The results obtained reveal that the method of determining structure factors from the thickness oscillations of the X-ray integrated intensity can be extended to imperfect crystals with a statistical distribution of defects.

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