Experimental Correction for Primary and Secondary Extinction.
I. Method

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Abstract

The energy transfer equations are used to solve secondary extinction in a parallel-sided crystal slab in Laue geometry. Practical formulas are given for determination of the reflectivity per unit length, $\sigma(\varepsilon)$, from the measured absolute powers of the diffracted and direct beams at rocking angle $\varepsilon = \theta - \theta_B$. The calculation assumes non-divergent beams, and the experimental tolerances are considered in detail. The primary extinction factor is defined by $y_p(\varepsilon) = \sigma(\varepsilon)/\sigma_{kin}(\varepsilon)$, where σ_{kin} is the kinematical reflectivity. When $y_p > 0.5$ it may be approximated by $y_p \simeq \exp[-(\alpha\delta)^2]$, where α is about 0.5 and δ is the average size of the coherent domains when measured in units of the extinction distance Λ, $\delta = D/\Lambda$. The extinction distance can be varied by changing the X-ray wavelength or polarization, and $y_p(\varepsilon)$ can be determined from two measurements.

1. Introduction

The explicit methods of counting for the defect structures of real crystals in X-ray and neutron diffraction have been based on the concept of mosaic crystal and energy transfer equations (Hamilton, 1957; Zachariasen, 1967; Cooper & Rouse, 1970; Coppens & Hamilton, 1970; Becker & Coppens, 1974*a,b*, 1975), although this has been realized to be an oversimplified approach, which can cover secondary extinction only (Werner, 1969, 1974). However, in recent years there has been considerable theoretical effort to bridge the gap between the infinite perfect crystal and the ideally imperfect mosaic crystal with more realistic calculations (Afanas'ev & Kohn, 1971; Kato, 1976*a,b*, 1979, 1980*a,b,c*). The basic difficulty with all theoretical calculations is that these use parameters which are inherently non-observable or are derived from specific geometrical models.

The practical procedures for making extinction corrections to the observed intensities of an X-ray or neutron diffraction measurement are based on least-squares fitting to the intensities which are calculated from a crystal model. The model includes theoretical structure factors and extinction parameters, which are highly correlated, and this may introduce artefacts as demonstrated by a few representative examples (Killean, Lawrence & Sharma, 1972; Cooper & Rouse, 1976; Lawrence, 1977; Schneider, 1977). At any rate, the results derived from various fitting schemes do not provide direct evidence of the validity of the theory.

The direct experimental approaches to the problem of extinction introduce corrections deduced from the reflection profile or are based on an extrapolation to the kinematical, zero interaction limit. The latter approach is discussed at length by Mathieson (1979), who classifies the methods as general, where the reflectivity per unit volume, Q, is reduced towards zero, and special, where the active volume of interaction is decreased.

Reflectivity is proportional to λ^3, where λ is the X-ray wavelength, and 'extinction-free' structure factors have been measured using energetic γ-rays (Schneider, 1977). Another extrapolation to $Q = 0$ is achieved by making the measurements with radiation polarized in the plane of diffraction and tuning the wavelength so as to make the scattering angle $2\theta \simeq 90°$; this is becoming feasible at the synchroton radiation sources (Mathieson, 1977*a*). The dependence of the integrated reflection on the polarization factor has been used also for determination of the parameters of Zachariasen's model (Chandrasekhar, Ramaseshan & Singh, 1969).

Changing the active volume is the traditional method of tackling extinction (James, 1962, ch. VI), and the recent variations include asymmetric Bragg reflection (Mathieson, 1977*b*) and tilting the crystal about the scattering vector in the symmetrical Laue case (Lawrence & Mathieson, 1977). Also, spreading the reflecting power diffusely in reciprocal space may be included in this category, and in addition to mechanical introduction of strains in the crystal radiation defects (Sanger, 1969) and thermal gradients (Seiler & Dunitz, 1978) have also been employed.

The information contained in the reflection profile has been used in a synthesizing method (Bradaczek & Hosemann, 1968; Urban & Hosemann, 1972), which

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includes a parametrized extinction model, and in a direct determination of secondary extinction from energy conservation (DeMarco, 1967).

Several of the above methods have proved adequate for finding corrections for secondary extinction. Primary extinction can be eliminated at the limit of zero interaction, but this means by definition very weak intensities and often also cumbersome measurements. However, the recent theoretical formulations include measures of primary extinction which can be related to experimental quantities. The basic idea of the present work is to determine secondary extinction from the energy transfer equations and extrapolate to negligible primary extinction by varying the factors that determine the amplitude coupling between coherent waves. The recent theories of extinction are reformulated using only parameters which are observable in a typical diffraction experiment.

2. Transfer equations

(a) Amplitude coupling

When one diffracted wave of displacement D_g is excited by the direct wave D_d, transfer equations of Takagi–Taupin type can be written as (Takagi, 1962, 1969; Taupin, 1964)

$$\frac{\partial D_d}{\partial s_d} = i\kappa_{-g} D_g \exp\{iG(s_d,s_g)\} = i\kappa_{-g} D_g \varphi(s_d,s_g) \quad (1a)$$

$$\frac{\partial D_g}{\partial s_g} = i\kappa_g D_d \exp\{-iG(s_d,s_g)\} = i\kappa_g D_d \varphi^*(s_d,s_g) \quad (1b)$$

with

$$\kappa_g = (\lambda C/V_c) r_e F_g, \quad (1c)$$

where C is the polarization factor ($\cos 2\theta$ when the electric vector is in the plane of diffraction, 1 when perpendicular to that), V_c the unit-cell volume, $r_e = e^2/mc^2$ the electron scattering length, and F_g the structure factor. The equations apply to distorted crystals as the lattice phase is given by $G = 2\pi g \cdot u$, where g is the scattering vector and u the lattice distortion. Position in the crystal is given by the oblique coordinates (s_d,s_g) along the direct and diffracted beams, respectively.

A statistically homogeneous crystal can be characterized by two parameters: the average lattice phase $\langle \varphi \rangle = H$ is a measure of the long-range perfection, and the intrinsic correlation length τ is related to the short-range perfection (Kato, 1980b). A narrow wave which enters the crystal is split into two coherent waves, but these become incoherent when there is an irrevocable change in G. In the terminology introduced by Kato (1980b), the respective intensity fields leave the coherent channel. It is worth noting that local

distortion (such as thermal vibration) does not break the coherence of the waves, if the crystal is perfect as a whole. The volume where the wave coherence prevails is called the coherent domain, and it is determined either by the edges of the crystal or by changes in G. It is seen in Fig. 1 that the maximum area of coherent coupling between the waves is $2s_d s_s \sin \theta$, and the appropriate average is found by an integration over the exit surface. The 'size' of the coherent domain, as seen by this particular wave, may be defined as

$$D = (\overline{s_d s_g})^{1/2}. \quad (2)$$

Explicit calculation is possible for a given cross section of a perfect crystal, and detailed results are available for square and circular shapes (Olekhnovich & Olekhnovich, 1978, 1980). If the incoming ray is taken as a plane wave, reflectivity curves $R(\varepsilon)$, where $\varepsilon = \theta - \theta_B$ is the deviation from the Bragg angle, can be calculated. The result can be expressed in Bessel functions of even orders, and the argument of the functions is proportional to $\kappa_g D$. The detailed form of $R(\varepsilon)$ depends on the crystal shape, and even for a given shape there is no simple relationship between $R(\varepsilon)$ and $\kappa_g D$. On the other hand, the integral of $R(\varepsilon)$ is well defined by $\kappa_g D$, and this will be utilized in the following.

(b) Intensity coupling

Consider now two plane waves of unit amplitude and lateral width which enter the crystalline medium at an angle $\varepsilon = \theta - \theta_B$. The waves have an arbitrary phase
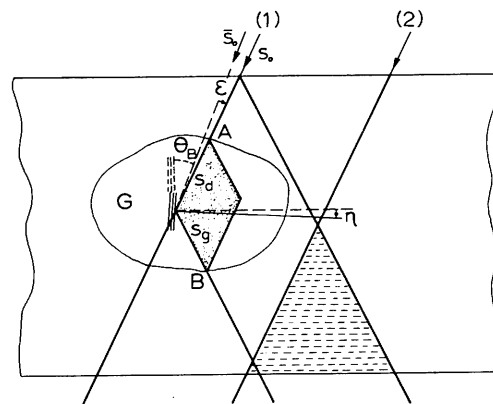

Fig. 1. Laue diffraction from a parallel-sided crystal slab. A coherent domain with a constant lattice phase G deviates from the average orientation, which is indicated by broken lines, by an angle η. The direction of the incident narrow wave, s_0, makes the angle ε with $\overline{s_0}$ which corresponds to the center of the reflection. Position in the crystal is measured along the direct and diffracted beams by s_d and s_g, respectively, and the area shaded with dots indicates the region of coherent coupling for the waves which enter G at A and exit at B. The region of interaction for two incoherent waves (1) and (2) is indicated by the area shaded with broken lines.

difference, and at each point the energy transfer equations hold,

$$\frac{\partial P_d}{\partial s_d} = -\mu_e^i P_d + \sigma_{-g} P_g \qquad (3a)$$

$$\frac{\partial P_g}{\partial s_g} = -\mu_e^i P_g + \sigma_g P_d. \qquad (3b)$$

If we assume a non-polar crystal, $\sigma_g = \sigma_{-g}$, and ignore the possibility of the Borrmann effect,

$$\mu_e^i = \mu_0 + \sigma, \qquad (4)$$

where $\sigma = \sigma(\varepsilon, s_d, s_g)$ is the reflectivity per unit length of the traversed crystalline medium and μ_0 the linear absorption coefficient. The region of interaction is that where the intensity fields of the two, originally narrow, waves overlap and where $\sigma \neq 0$ (Fig. 1). In the regions where $\sigma = 0$ the equations reduce to those of normal absorption. The observable reflectivity $\sigma(\varepsilon)$ is the integral of $\sigma(\varepsilon, s_d, s_g)$ over the region of interaction for the extended incident beam, and the integrated intensity is found by an angular integration.

3. Primary extinction

The coherent domain size, as defined in (2), does not refer to the mosaic-crystal model, but it rather measures the spatial extent of the amplitude coupling. However, for the sake of concreteness, it is useful to think of a crystallite of size D, which deviates from the average orientation of the crystallites by an angle η. The crystallites of this size have an average reflectivity curve $R_D(\psi)$, where ψ is the angle from the normal of the reflecting planes. An extended beam, which makes an angle ε with the direction of average orientation, sees a (normalized) distribution of crystallite sizes, $w_D(\eta)$, and the contribution to the reflectivity of the crystal may be written as

$$\sigma_D(\varepsilon) = \int R_D(\varepsilon - \eta) \, w_D(\eta) \, d\eta. \qquad (5)$$

In the following we will consider in detail cases where w_D varies with angle much slower than R_D, and in that case

$$\sigma_D(\varepsilon) = w_D(\varepsilon) \int R_D(\eta) \, d\eta. \qquad (6)$$

This formulation implies that there is a representative distribution of R_D as a function of the orientation angle η. It is more realistic to define $\sigma_D(\varepsilon)$ in a small range of angles of incidence where $R_D(\varepsilon - \eta)$ differs from zero,

$$\overline{\sigma_D(\varepsilon)} = \frac{1}{\Delta\varepsilon} \int \sigma_D(\varepsilon) \, d\varepsilon = w_D(\varepsilon) \int_{\Delta\varepsilon} R_D(\varepsilon - \eta) \, d\varepsilon \qquad (7)$$

$$= Q y_p(D) \, w_D(\varepsilon),$$

where $Q = (\lambda/\sin 2\theta_B)|\kappa_g|^2$ is the integrated diffracted power per unit volume, and $y_p(D) < 1$ is the primary extinction factor. We are interested in cases where

primary extinction is appreciable, say $y_p(D) \leq 0.99$, and model calculations show that the width of the corresponding $R_D(\psi)$ is very small. According to the calculations by Olekhnovich & Olekhnovich (1978, 1980), at this limit $\Delta\varepsilon \simeq 10\lambda|\kappa_g|$, and typically $\Delta\varepsilon$ is 0.1 to 1 mrad. This range is usually covered by the beam divergences and/or continuous rocking of the crystal. Under these conditions, the observable reflectivity of the crystal is

$$\overline{\sigma(\varepsilon)} = Q \int y_p(D) \, w_D(\varepsilon) \, dD. \qquad (8)$$

It was mentioned earlier that the expressions of reflectivity involve low-order Bessel functions. For instance, in the Laue case of Fig. 1 the coherent intensity field at (s_d, s_g) is (Kato, 1980c)

$$I_g^c = H^2|\kappa_g|^2|J_0[2\kappa_g H(\overline{s_d s_g})^{1/2}]|^2 \exp\{-\mu_e^c(s_d + s_g)\}, \qquad (9)$$

where J_0 is the Bessel function of zeroth order, and μ_e^c the effective absorption coefficient for coherent intensity fields. For a perfect crystal, H is just the Debye–Waller factor $\exp(-M)$, and we define the extinction distance Λ by

$$\Lambda^{-1} = \kappa_g H = \frac{\lambda C}{V_c} r_e F_g H \qquad (10)$$

(note the small difference from the definition by Kato, 1980b). The arguments of the intensity expressions become linear in the size of the coherent domain, when this is measured in units of Λ,

$$\delta = D/\Lambda = (\overline{s_d s_g})^{1/2} \frac{\lambda C}{V_c} r_e F_g H. \qquad (11)$$

The effective size of the coherent domain can be varied by changing λ, C or F_g, and this gives an experimental possibility for determination of primary extinction.

At small enough values of δ it follows from the properties of J_{2n} that

$$y_p(\delta) = Q^{-1} \int R_\delta(\psi) \, d\psi \simeq \exp\{-(\alpha\delta)^2\}, \qquad (12)$$

where the numerical value of α is about 0.5. According to model calculations this approximation is valid when $\delta < 1.5$ or $y_p > 0.5$ (Olekhnovich & Olekhnovich, 1978, 1980).

The primary extinction factor at a given average angle of incidence is weighted by the distribution of the domain sizes,

$$y_p(\varepsilon) \, w(\varepsilon) = \int y_p(\delta) \, w[\delta(\varepsilon)] \, d\delta = Q^{-1} \overline{\sigma(\varepsilon)}. \qquad (13)$$

If we assume that $w[\delta(\varepsilon)]$ is a Gaussian of width $2\Delta_\varepsilon$, centered at $\bar{\delta}_\varepsilon$, convolution resulting from substitution of (12) into (13) yields

$$y_p(\varepsilon) = \exp\{-(\alpha\bar{\delta}_\varepsilon)^2/(1 + \alpha\sqrt{2}\Delta_\varepsilon)^2\} = \exp\{-(\alpha'\bar{\delta}_\varepsilon)^2\}. \qquad (14)$$

Although idealized, this calculation suggests that the functional form of (12) can be used also for real crystals. The working formulas for the actual determination of $y_p(\varepsilon)$ will be based on this approximation.

4. Secondary extinction

The energy transfer equations (3) can be solved for given boundary conditions and $\overline{\sigma(\varepsilon)}$. In the present work we will use Laue diffraction from a parallel-sided crystal of thickness T. In the symmetrical Laue case the diffracted power is (Zachariasen, 1945)

$$P_g^*(\varepsilon) = \tfrac{1}{2}P_0 \exp(-\mu_0 T/\cos \theta)$$
$$\times \{1 - \exp[-2\overline{\sigma(\varepsilon)} T/\cos \theta]\}$$
$$\simeq P_g(\varepsilon)/\{1 + r(\varepsilon) + \tfrac{1}{3}[r(\varepsilon)]^2\}, \quad (15a)$$

where

$$P_g(\varepsilon) = P_0 \exp(-\mu_0 T/\cos \theta) r(\varepsilon) \quad (15b)$$

is the diffracted power as corrected for secondary extinction, and $r(\varepsilon) = \overline{\sigma(\varepsilon)} T/\cos \theta$. The direct beam can be solved from the equations as well,

$$P_d^*(\varepsilon) = P_0 \exp(-\mu_0 T/\cos \theta) - P_g^*(\varepsilon) \quad (16a)$$
$$\simeq P_0 \exp(-\mu_0 T/\cos \theta)/\{1 + r(\varepsilon)\}, \quad (16b)$$

where the expression emphasizes the energy conservation. The series expansions are valid to second order in $r(\varepsilon)$.

If the effects of primary extinction are negligible,

$$\int R_D(\varepsilon - \eta) \, d\varepsilon = Q \quad (17)$$

for all values of D. From (5) we obtain the kinematical reflectivity

$$\sigma_{\text{kin}}(\varepsilon) = \int \int R_D(\varepsilon - \eta) w_D(\eta) \, d\eta \, dD$$
$$= \int R(\varepsilon - \eta) w(\eta) \, d\eta, \quad (18)$$

where $R(\varepsilon - \eta)$ is the average reflectivity of the domains oriented in an angle η, and $w(\eta)$ is the normalized abundance of these domains. In his theory for secondary extinction, Zachariasen (1967) distinguishes two cases on the basis of the wider, dominant distribution,

$$\sigma_{\text{kin}}(\varepsilon) = Qw(\varepsilon) \quad \text{type I} \quad (19a)$$
$$\sigma_{\text{kin}}(\varepsilon) = R(\varepsilon) \quad \text{type II.} \quad (19b)$$

The integrated intensity is found by an angular integration over the rocking angle of the crystal. In the symmetrical Laue case

$$E = \frac{1}{P_0} \int P_g(\varepsilon) \frac{d\varepsilon}{\omega} = \frac{1}{\omega} \exp(-\mu_0 T/\cos \theta) \int r(\varepsilon) \, d\varepsilon.$$
$$(20a)$$

If only secondary extinction is present, E can be related to the structure factor through the kinematical expression

$$E_{\text{kin}} = \frac{Q}{\omega} \frac{T}{\cos \theta} \exp(-\mu_0 T/\cos \theta). \quad (20b)$$

5. Discussion

The above formulation of the primary- and secondary-extinction effects involves a few assumptions which deserve closer discussion. These are related to the practical realization of the method and to a description of the crystal with distribution functions.

(a) Primary extinction

The case where primary extinction is appreciable was separated from (5) by the assumption that the reflectivity curves $R_D(\psi)$ of the responsible coherent domains are narrow in comparison with the total angular width of the reflection. This is only a matter of concreteness, as the convolution can be retained through (6) to (8), but the calculation gives estimates of the beam divergences that do not smear the effects of primary extinction.

The estimation of primary extinction is based on a simple relationship between the extinction factor y_p and the domain size. The approximation may be valid for $y_p > 0.5$, but the functional form (12) is smeared by the size distribution. A Gaussian distribution retains the form (12), but in the case of a real crystal the performance of the approximation can be judged only by the consistency of the results.

(b) Inhomogeneity of $\sigma(\varepsilon)$

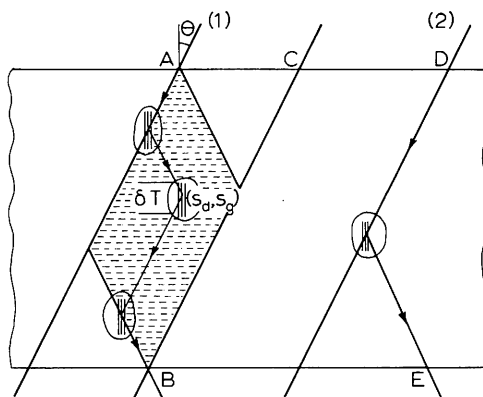The correction for secondary extinction was evaluated for a parallel-sided crystal slab in the

Fig. 2. 'Serial' and 'parallel' coupling of rays. For the ray (1) entering the crystal at A and exiting at B the reflecting domains within the shaded area are coupled 'in series', while the domain seen by the ray (2) is 'in parallel' with the domains experienced by (1).

symmetrical Laue geometry. The calculation involves the use of the average reflectivity $\overline{\sigma(\varepsilon)}$ for the whole direct and reflected X-ray beams. In the actual case the reflectivity experienced by the elemental rays varies, and the average should be calculated over the exit beam as counted by a detector. The conditions of homogeneity for a type-I crystal have been recently discussed by Mazzone (1981), and Schneider (1975) has demonstrated the effects of an inhomogeneous $\sigma(\varepsilon,s_d,s_g)$ on the diffracted intensity. This calculation can be generalized by terming the reflecting domains in the crystal as being 'in series' or 'in parallel'; see Fig. 2. For a ray travelling in the shaded area the total reflecting ratio $r_s(\varepsilon)$ is the sum of elemental reflecting ratios $\delta r(\varepsilon,s_d,s_g) = \sigma(\varepsilon,s_d,s_g).\ \delta T(s_d,s_g)/\cos\theta$, i.e. the effective reflectivity is the average reflectivity $\overline{\sigma(\varepsilon)}$ taken over the area of 'serial coupling'.

The intensities of 'parallel' rays are added, and from (15a)

$$\sum P_{g,i} = \tfrac{1}{2}P_0 \exp(-\mu_0 T(\cos\theta)\sum\{1 - \exp(-2r_{i,p})\}.$$
(21)

Writing $r_{i,p} = \bar{r} + \Delta r_{i,p}$ and expanding in power series

$$\sum P_{g,i} = \tfrac{1}{2}P_0 \exp(-\mu_0 T/\cos\theta)\{1 - \exp(-2\bar{r})$$
$$\times [1 + \sum\tfrac{1}{2}(2\Delta r_{i,p})^2]\}.$$
(22)

Accordingly, the 'parallel' inhomogeneities of $\sigma(\varepsilon,s_d,s_g)$ increase the effects of secondary extinction, and the reflectivity calculated from (15) and (16) remains smaller than the actual average $\overline{\sigma(\varepsilon)}$. The structure factor F is related to the average reflectivity through (20a), and so the inhomogeneities of $\sigma(\varepsilon)$ make the calculated values the lower bounds for F.

(c) Divergences of the beams

The reflectivity $\overline{\sigma(\varepsilon)}$ refers to beams of parallel rays, and in the energy transfer equations (3) $\overline{\sigma(\varepsilon)}$ is taken to be the same for direct and reflected beams. The divergences of the beams change in successive reflections, as illustrated by the Ewald construction in Fig. 3. The width of the reflecting domain in reciprocal space, i.e. the dimension perpendicular to $S = s_g - s_d$, is determined by the orientation distribution of the reflecting crystallites, and the length (parallel to S) by the thickness distribution of the crystallites. The power and the divergences of the reflected beam are determined by the section cut from the domain by the Ewald sphere. The figure shows the case of a low-angle reflection, which is of interest in a study of extinction effects. The inserts show the convolution by the beam divergence, and it is clear that the active volume of the reflecting domain increases in successive reflections. In a type-I crystal this is not substantial, and the figure even shows how the divergence of the incident beam could be retained by matching it to the length of the

domain; this corresponds to the averaging discussed in § 3. In a type-II crystal the effective reflectivity already in the second reflection is rather the reflectivity averaged over the domain.

The above situation was recognized already by Darwin (1922), and the necessary modification of the transfer equations is discussed by Werner (1974). If the reflectivity is small, it can be taken as $\sigma(\varepsilon)$ for the direct beam, σ_{eff} for the reflected beam, and an approximate solution in the symmetrical Laue case is

$$P_d(\varepsilon) = P_0 \exp(-\mu_0 T/\cos\theta)/\{1 + \tfrac{1}{2}[r(\varepsilon) + r_{\text{eff}}]\}.\quad(23)$$

If r_{eff} is defined by $\int r(\varepsilon)\,\mathrm{d}\varepsilon = \Delta\varepsilon r_{\text{eff}}$, the integrated reflection from (20a) remains unchanged.

The problems of divergences are avoided when the incident beam is a spherical wave. There is no delicate double integration due to the angular scan and the convolution by the beam divergences, as pointed out by Kato (1980a). A first-order correction for secondary extinction would still be possible using the present method, as seen from (16) and (20), but only an approximate correction which assumes a constant y_p would be possible when the extinction distance is varied.

(d) General comments

A simple closed-form solution of the energy transfer equations is available only for a parallel-sided crystal in
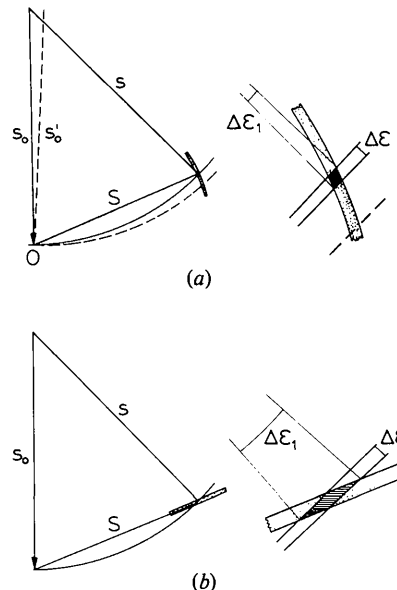

(a)

(b)

Fig. 3. Ewald construction for diffraction from (a) a type-I crystal and (b) a type-II crystal. The broken lines in (a) show the effect of variation of the incident-beam direction. The inserts illustrate the active volume of the diffraction domain, when $\Delta\varepsilon$ is the divergence of the incident beam. In (a) $\Delta\varepsilon$ can be matched to the length of the diffraction domain (along S) to make the divergence of the diffracted beam, $\Delta\varepsilon_1$, equal to $\Delta\varepsilon$, while in (b) $\Delta\varepsilon_1$ is large enough to cover the whole diffraction domain in the next reflection.

Laue geometry, but an iterative solution can be presumably found for any simple polyhedral crystal, when the so-called *AB*-extinction formulas of Werner (1974) are used. The present experimental correction for secondary extinction requires an absolute measurement of the transmitted intensity. This is difficult to measure when the crystal is bathed in the incident beam, while a measurement of the total transmitted power is sufficient for a crystal slab which intercepts the beam.

The evaluation of atomic scattering factors from the integrated reflections is based on the assumption that all elastic scattering is concentrated in the Bragg reflections. In other words, all atoms in the irradiated volume should belong to an environment where translational invariance holds over a sufficient range. The simplest model for such a crystal is the mosaic crystal, but it excludes lattice defects that occur in real crystals. A diffraction measurement should therefore commence with careful tests of background scattering, as the intensity outside the Bragg reflections should be due to inelastic scattering only. This requires extensive calculations of the scattering cross sections and a measurement of the incident beam power. It has been demonstrated (Suortti & Jennings, 1977) that the amorphous scattering can be appreciable in powders, and it is not clear without careful checks that the total irradiated volume of a single crystal contributes to the observed integral reflection.

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