after that it again enlarges but with the opposite orientation of the Poynting vector. A further interesting property can be found in Figs. 8(*e*) (σ) and in 8(*i*) (π). The values of Δk for the cases drawn imply $x_1 = x_4$ and $x_2 = x_3$. The dispersion surface in the plane of incidence is created by the three lines identical with the asymptotes. The last interesting property we wish to point out is illustrated in Fig. 8(*c*). The dispersion surface in the plane of incidence is identical for σ and π polarizations.

APPENDIX B

Reflection coefficient

We define the reflection coefficient according to James (1963) as

$$R = |Z_2|^2/|Z_1|^2 \qquad (13)$$

for the symmetrical reflection 440; for the extremely asymmetrical reflection 404 the formula used leads to zero. It was found (*e.g.* Bedyňska, 1973; Colella, 1982) that in cases of extremely asymmetrical reflections linearization of the coefficients of the dynamical equations is not permissible. We added to the coefficients A_{ii} from (1) and (2) the term $-(\Delta \,.\, \Delta)/k_m^2$ and calculated for $k = k_m$ the dispersion surface in this more accurate approach. The result of this calculation gives us in the 3 μm^{-1} neighbourhood of the Laue point a picture of the dispersion surface nearly identical with that calculated according to (5) (the shift in the values of $x_{1,2,3}$ is of the order of 10^{-5} μm^{-1}). We conclude that the dependences illustrated in Fig. 5 would stay unchanged in the more accurate approach. For this reason we can also expect that the resultant dependences shown in Figs. 6 and 7 are valid.

No great problems arose when we attempted to proceed in the obvious way to apply the boundary conditions and also to calculate the reflection coefficient with the linearized coefficients A_{ii}. The formula derived from (13) for Si[000, 440, 404] and the 440 reflection

$$R_{440} = B^2(B_{33} - B)^2/(B_{22}B_{33} - B^2)^2 \qquad (14)$$

does not lead to physically impossible or to apparently improbable results.

Several examples of the reflection coefficient R_{440} are shown together with the corresponding dispersion surfaces in Fig. 8. The angular scale given in Fig. 8(*a*) follows from (11). The reflection coefficient R_{440} equals one for the ranges of total reflection. The only point for which the calculation using the linearization is indefinite is $x = x_4$ and Δk such that $B_0 = 2B/3$ [Fig. 8(*e*) (σ) and Fig. 8(*i*) (π)]. From the continuity of R_{440} we established $R_{440} = 1$ at this point.

References

BEDYŇSKA, T. (1973). *Phys. Status Solidi A*, **19**, 365-372.

BUBÁKOVÁ, R. & PACHEROVÁ, O. (1978). *Acta Cryst.* A**34**, S233.

CHANG, S. L. (1984). *Springer Series in Solid-State Sciences*, Vol. 50. Berlin: Springer.

COLELLA, R. (1982). *Z. Naturforsch. Teil A*, **37**, 437-447.

DESLATTES, R. D. (1968). *Appl. Phys. Lett.* **12**, 133-135.

EWALD, P. P. & HÉNO, Y. (1968). *Acta Cryst.* A**24**, 5-15.

GRAEFF, W. & BONSE, U. (1977). *Z. Phys. B*, **27**, 19-32.

HÉNO, Y. & EWALD, P. P. (1968). *Acta Cryst.* A**24**, 16-42.

JAMES, R. W. (1963). *Solid State Phys.* **15**, 53-220.

PACHEROVÁ, O. (1979). Thesis. Czechoslovak Academy of Sciences.

PACHEROVÁ, O. & BUBÁKOVÁ, R. (1982). *Z. Naturforsch. Teil A*, **37**, 617-625.

PACHEROVÁ, O. & BUBÁKOVÁ, R. (1984). *J. Appl. Cryst.* **17**, 375-384.

PENNING, P. & POLDER, D. (1968). *Philips Res. Rep.* **23**, 1-12.

Acta Cryst. (1987). A**43**, 167-173

Random Elastic Deformation (RED) – an Alternative Model for Extinction Treatment in Real Crystals*

BY J. KULDA

Nuclear Physics Institute, Czechoslovak Academy of Sciences, 250 68 Řež near Prague, Czechoslovakia

(*Received* 30 *December* 1985; *accepted* 3 *September* 1986)

Abstract

A modification of the Hamilton-Zachariasen theory of extinction in imperfect crystals is reported. Unlike the generally adopted Darwin mosaic model the crystal is supposed to consist of elastically deformed domains so that individual reflection events can be treated within the quasiclassical approach developed by the author in an earlier study [*Acta Cryst.* (1984), A**40**, 120-126]. In this way a modified expression for the scattering cross section, taking into account multiple wave interference, is introduced into the

* This paper should be regarded as forming part of the Ewald Memorial Issue of *Acta Cryst.* Section A published in November 1986. The manuscript was received in its final form too late for inclusion.

0108-7673/87/020167-07$01.50 © 1987 International Union of Crystallography

energy transport equations. Under the assumption of a constant magnitude of the strain gradient and of spherical isotropy of its orientation distribution, generally valid results formally compatible with the standard solution of Becker & Coppens [*Acta Cryst.* (1974), **A30**, 129-147] are obtained. A significant achievement is the physically correct description of the primary extinction effect.

1. Introduction

The problem of an adequate description of extinction effects in real crystals has been subject to extensive scrutiny since the advent of X-ray structure analysis. Darwin (1922) introduced, for crystals of intermediate perfection, the mosaic model assuming the crystal to be composed of perfect but slightly misoriented blocks. Reflecting power calculations were performed for special sample geometries by various authors until Hamilton (1957) and Zachariasen (1967, 1968) introduced the energy transport equations (ETE)

$$\begin{aligned} \partial I_0/\partial s_0 &= -\sigma(I_0 - I_G), \\ \partial I_G/\partial s_G &= -\sigma(I_G - I_0). \end{aligned} \tag{1}$$

Here I_0, I_G represent the intensities of the transmitted and reflected waves, respectively, and s_0, s_G are the corresponding beam paths. The angular dependence of the coherent scattering cross section σ is determined by convolution of the intrinsic rocking curve $\sigma(\varepsilon)$ of a single mosaic block with the misorientation distribution function $w(\Delta\theta)$.

Later, solutions of (1) were obtained for quite general crystal shapes by various authors, the standard references being the works of Cooper & Rouse (1970) and Becker & Coppens (1974) (henceforth BC). Since then the treatment based on the ETE has become a routine technique included in several structure refinement programs. However, this approach suffers from certain deficiencies, of which the most important in practice seems to be its failure to describe primary extinction. This arises from the fact that no useful solutions for finite-size mosaic blocks are available in the dynamical theory of diffraction because of the complexity of the phase boundary conditions. This problem has been by-passed in a first-order approximation by BC solving the ETE again, this time for individual blocks.

An entirely new approach, largely overcoming this problem, was introduced by Kato (1976, 1979, 1980). He starts under very general assumptions from the Takagi (1962, 1969) and Taupin (1964) equations. The transfer to the case of a crystal containing some random distribution of imperfections is performed by ensemble averaging the mutual phases of waves corresponding to different dynamical beam trajectories. As a price to pay for the general formulation, the results involve phase correlation lengths of higher orders which are difficult to calculate precisely, even for quite simple model cases. In practice, satisfactory results are obtained only for phase correlation lengths t^* not exceeding the primary extinction distance Λ, *i.e.* again for small primary extinction. On the other hand, in the limit of $t^* \ll \Lambda$ the Takagi-Taupin equations are transformed to a form equivalent to ETE except that the formulation employs incident spherical waves and an angularly independent cross section σ.

It can be concluded therefore that the use of the ETE in the limit of secondary extinction is physically correct. We shall start from this point and concentrate on the search for a new model which could replace the concept of mosaic structure and yield an analytical expression for σ, taking into account the primary extinction. In the next section we shall for this purpose briefly reconsider some important features of dynamical wave propagation in crystals. Our arguments will be based on the neutron diffraction case where nonrelativistic quantum mechanics (*e.g.* Messiah, 1962) is directly applicable. All the particular results have, however, analogous validity for X-rays. We shall also confine ourselves to the case of zero absorption; the extension to absorbing crystals can be accomplished to first order (no anomalous effects) exactly as in the treatment of BC.

2. Wave propagation in crystals

According to the standard treatment of the dynamical theory of diffraction [*e.g.* Rauch & Petrascheck (1978) for neutrons and Pinsker (1978) for X-rays] in the simplest, one-beam, case, an incident vacuum plane wave $u_0^v \exp(i\mathbf{kr})$ excites a single plane wave $u_0 \exp(i\mathbf{Kr})$ in the crystal interior. The nonzero interaction potential, represented by its matrix element $V_0 = (2\pi\hbar^2/m)(F_0/\Omega)$ (F_0 is the structure factor, Ω the elementary-cell volume and m the neutron mass) brings about only a slight reduction/increase of the wave-vector magnitude accompanied by a tiny change in flight direction to satisfy the continuity conditions on the crystal surface. Hereafter these effects will be neglected.

Whenever the Bragg condition is satisfied (the two-beam case is assumed), inside the crystal there exist two symmetry-equivalent propagation directions of the transmitted and reflected waves, $\psi_0 = u_0 \exp(i\mathbf{Kr})$ and $\psi_G = u_G \exp[i(\mathbf{K}+\mathbf{G})\mathbf{r}]$ respectively. This degeneracy is removed by the wave-crystal interaction represented by the additional matrix element $V_G = (2\pi\hbar^2/m)(F_G/\Omega)$. The eigenstates corresponding to the two branches of the dispersion surface are described by wavefunctions of the type

$$\psi_{1,2} = u_0^{(1,2)} \exp(i\mathbf{K}_{1,2}\mathbf{r}) + u_G^{(1,2)} \exp[i(\mathbf{K}_{1,2}+\mathbf{G})\mathbf{r}]. \tag{2}$$

The ratio of the amplitudes $u_G^{(i)}/u_0^{(i)}$ is given by

$$c_{1,2} = Y \pm (Y^2+1)^{1/2}, \tag{3}$$

where $Y = a/(2\chi_G)$ and $\chi_G = V_G/[\hbar^2 k^2/(2m)]$ and a is defined by Kulda (1984).

If the transition from vacuum to crystal is instantaneous, which is the case normally considered in the dynamical theory, both eigenstates are excited simultaneously, the amplitudes being determined by the boundary conditions. During further propagation inside the crystal the small difference of the order of V_G in energy eigenvalues leads to interference causing the well known *Pendellösung* oscillations of intensity between the transmitted and reflected waves. Hence, for any point on the exit surface exact knowledge of the wave-field phase is required if the reflecting power is to be determined precisely. For this reason the dynamical theory provides really simple results only for infinite plane-parallel plates.

The situation changes substantially when the lattice orientation varies along the beam path in the sample on a scale exceeding the dynamical reflection width $W_D \simeq 1''$ as in the case of elastically deformed crystals. Here the parameter Y depends on the local displacement $\mathbf{U}(\mathbf{r})$ of the atomic positions through $a = a_0 - 2\lambda\partial(\mathbf{GU})/\partial s_G$ (Takagi, 1969). Typically the incident wave will not satisfy the Bragg condition when entering the crystal so that in fact only the one-beam case will be realized. According to the adiabatic theorem of quantum mechanics (Messiah, 1962), the wavefunction will follow continuously the variation of its parameters which all depend on Y. If, for instance, the starting value was $Y_0 \ll 0$ and because of (2) and (3) $\Psi = \psi_1 = \psi_0$, then, in the case of very slight deformation, along all the trajectory inside the crystal $\Psi = \psi_1$ and on the exit surface where $Y \gg 0$ we get $\Psi = \psi_1 = \psi_G$.

Obviously in this ideal case only one of the eigenstates is excited. At finite rates of Y variation, however, transitions to the other eigenstate are excited (Balibar, Chukhovskii & Malgrange, 1983; Kulda, 1984). Their probability may be calculated as

$$w = \exp\left[-(2V_G/\hbar)\int_{-1}^{1}(1-Y^2)^{1/2}|\partial Y/\partial s_0|^{-1}\,\mathrm{d}Y\right]. \tag{4}$$

Because the transition point is not fixed within the domain of $-1 \le Y \le 1$, no well defined phase relation between the old wave and the newly created one exists. Hence no *Pendellösung* oscillations take place and the reflection probability for an incident plane wave is given just by $r = 1 - w$. In the case of a constant strain gradient we have

$$r = 1 - \exp\left(-Q|\partial\Delta\theta/\partial s_0|^{-1}\right) \tag{5}$$

with

$$Q = F_G^2\lambda^3/(\Omega^2 \sin 2\theta).$$

To determine the integrated reflecting power ρ_θ one has to integrate r over the whole misorientation range $\delta\theta$ in the sample. For symmetric reflection on a plane parallel plate of thickness D, uniformly bent with radius R, we obtain for ρ_θ a particularly simple expression,

$$\rho_\theta = (D/R)\cot\theta[1 - \exp(-QR/\cos\theta)]. \tag{6}$$

We recall that the validity of (5) and (6) is restricted to cases where $\delta\theta > w_D$ (for details see Kulda, 1984) but does not require special assumptions concerning the crystal dimensions and shape. In fact, for crystals less than the extinction distance, $|\partial\Delta\theta/\partial s_0|$ becomes large and a power-series expansion in (5) and (6) leads to kinematical expressions $r = Q|\partial\Delta\theta/\partial s_0|^{-1}$ and $\rho_\theta = QD/\sin\theta$. Hence (6) is both simple enough and reasonably accurate for a wide range of degrees of crystal perfection. For these reasons it seems more promising to base a statistical model for extinction treatment in real crystals on elastically deformed rather than perfect lattice domains.

3. The RED model

We shall start from the following two basic assumptions concerning the macroscopic distortions in the crystal:

1. The crystal is built up of domains distorted by elastic deformation of random magnitude and orientation.

2. Local departure from the precise Bragg-angle value remains continuous on domain boundaries.

In other words, the true dependence $\Delta\theta(s_0, s_G)$ is approximated by a random-walk process in discrete time (Feller, 1970), the step length being determined by the domain size. The beam transport through the crystal will be considered as a series of independent reflections on those parts of the random-walk sequence which have proper orientation with respect to the incident plane wave (Figs. 1*a*, *b*). Each individual reflection will be treated *via* the reflection coefficient r derived in the preceding section (5).

In order to arrive at simple analytical results we shall complete the general assumptions by several further specifications: the domains will be of spherical shape with a constant diameter d; the deformation will be represented by uniform bending with a constant radius R; the sign of the deformation gradient will change at random with the transition probabilities p_+, p_- being independent of position in the crystal; and for reflections in individual domains the diffraction vector $\mathbf{G}$ will be parallel to the atomic position displacement $\mathbf{U}$.

As a consequence, the mean path per domain is $\bar{l} = 3d/4$ and the corresponding variation of $\Delta\theta$ is given by

$$\delta\theta = \bar{l}|\partial\Delta\theta/\partial s_0| = \bar{l}\cos\theta/R.$$

The probability distribution for various $\Delta\theta$ values, if it exists, has to be an invariant of the random-walk

process whose behaviour depends in a profound manner on the transition probabilities p_+, p_-. Here we shall give two examples.

The choice of constant values $p_+ = p_- = \frac{1}{2}$ results in an unrestricted random walk, in which case no invariant probability distribution for $\Delta\theta$ exists. Although this might be a good approximation to the real situation with multimodal misorientation distribution functions as observed frequently in experiments (Schneider, 1975), it is unsuited for further mathematical treatment.

For the rest of this paper we shall make use of another interesting possibility and fix the width of the total misorientation range to 2η so that $-\eta \le \Delta\theta \le \eta$ and we shall put

$$p_+ = (\eta - \Delta\theta)/(2\eta), \quad p_- = (\eta + \Delta\theta)/(2\eta). \quad (7)$$

Clearly, for $\Delta\theta = \eta$ one has $p_+ = 0$ and $p_- = 1$ and analogously $p_+ = 1$, $p_- = 0$ for $\Delta\theta = -\eta$, implying a bounded random walk with reflecting walls. In this case a stationary probability distribution exists and has the binomial form

$$P_N(j) = 2^{-N}\binom{N}{j}$$

with $N = 2\eta/\delta\theta$ and $j = (\eta + \Delta\theta)/\delta\theta$. The local misorientation angle $\Delta\theta$ will now oscillate about zero, achieving only extremely rarely values near $\pm\eta$. This behaviour is demonstrated in Fig. 2 on a sequence of 100 domains with $N = 10$ obtained with the help of a pseudorandom number generator.

As can be seen from Figs. 1(*a*) and 2(*a*), $P_N(j)$ refers in fact to the domain boundaries characterized by discrete misorientation values $\Delta\theta = j\delta\theta - \eta$. The probability for a wave to hit a properly oriented region is given by another distribution function $\tilde{P}_N(j)$ referring to the frequency of transitions between the states characterized by j, $j+1$ and *vice versa*, which can be expressed as

$$\begin{aligned}\tilde{P}_N(j) &= p_+ P_N(j) + p_- P_N(j+1) \\ &= \ldots = \tfrac{1}{2}[P_{N-1}(j-1) + P_{N-1}(j)].\end{aligned} \quad (8)$$

For large N we can use for both $\tilde{P}_N(j)$ as well as $P_N(j)$ the same Gaussian approximation

$$w(j) = (2/\pi N)^{1/2} \exp[-(2j-N)^2/2N] \quad (9)$$

with variance $N/4$. The mean recurrence time is, for any state, given by the reciprocal value of the stationary probability distribution function. For $j = N/2$ (*i.e.* $\Delta\theta = 0$), we obtain from (9) $\tau = (\pi N/2)^{1/2}$ steps. Hence for an incident wave corresponding to the maximum of the rocking curve the mean free path between subsequent reflections is $\bar{l} = \tau\bar{l} = (\pi N/2)^{1/2}\bar{l}$.

Switching to the angular scale again, we can rewrite (9) in the form

$$\begin{aligned}w_{\mathrm{RED}}(\Delta\theta) &= w(j)\,\mathrm{d}j/\mathrm{d}\Delta\theta \\ &= (\pi\eta\delta\theta)^{-1/2} \exp(-\Delta\theta^2/\eta\delta\theta).\end{aligned} \quad (10)$$

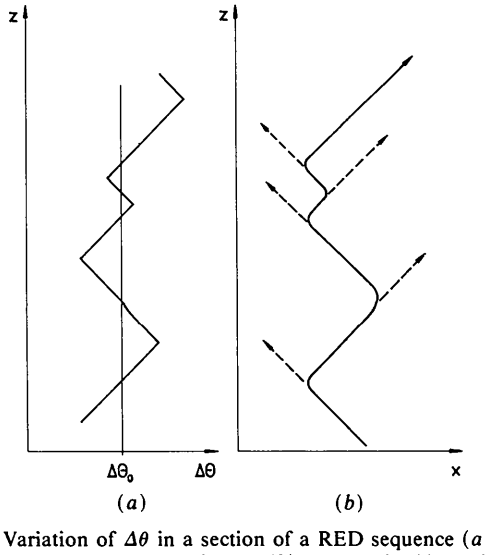

Fig. 1. Variation of $\Delta\theta$ in a section of a RED sequence (*a*) and the corresponding wave trajectory (*b*) characterized by an initial value $\Delta\theta_0$; broken lines indicate new waves created with probability given by equation (4); the z, x axes are parallel and perpendicular to the scattering planes, respectively.

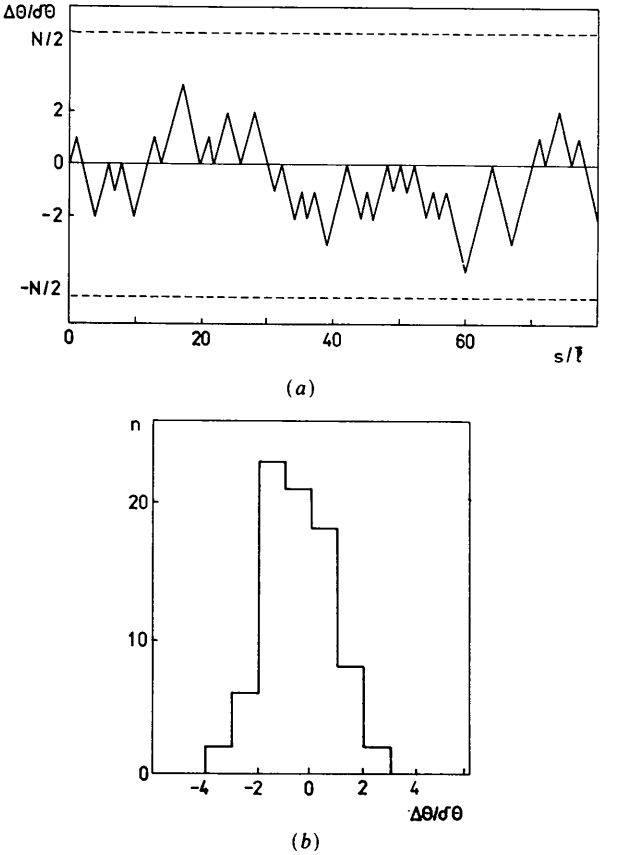

Fig. 2. A computer-simulated RED sequence with transition probabilities given by equation (7): (*a*) the $\Delta\theta$ variation along the beam path and (*b*) the abundance of particular domains in this sequence.

The angular parameters η, $\delta\theta$ can be expressed in terms of $\bar{t}$ and R so that

$$w_{\mathrm{RED}}(\Delta\theta) = (R/\bar{t}\cos\theta) \times \exp[-\pi(\Delta\theta R/\bar{t}\cos\theta)^2]. \quad (11)$$

In what follows w_{RED} will play a role analogous to the mosaic block orientation distribution function in the traditional treatment. Unlike the mosaic model the effective width of $w_{\mathrm{RED}}(\Delta\theta)$ [but not that of $w(j)$] here becomes angularly dependent through the factor $\mathrm{d}j/\mathrm{d}\Delta\theta = R/(\bar{t}\cos\theta)$. This takes into account the fact that Bragg reflection is less and less sensitive to angular deviations with increasing θ.

According to our assumptions the reflection probability for a wave hitting a properly oriented domain is given by expression (5). Hence the new expression for the coherent scattering cross section σ to be inserted into the ETE can be written as

$$\sigma_{\mathrm{RED}}(\Delta\theta) = rw_{\mathrm{RED}}(\Delta\theta)\,\mathrm{d}\Delta\theta/\mathrm{d}s_{0,G} = [1-\exp(-QR/\cos\theta)](1/\bar{t}) \times \exp[-\pi(\Delta\theta R/\bar{t}\cos\theta)^2]. \quad (12)$$

It is worth noticing that in the kinematical limit $QR \to 0$ and (12) reduces to

$$\sigma_{\mathrm{RED}}(\Delta\theta) = Qw_{\mathrm{RED}}(\Delta\theta),$$

which coincides with the conventional expression for type I crystals.

4. Solution of the ETE

Within our approximation σ_{RED} stays independent of coordinates s_0, s_G so that the original mathematical structure of the ETE is unaltered and we can use the BC solution for Gaussian mosaic distribution. The reflectivity term in square brackets in (12) renormalizes, however, its dependence on the kinematical reflecting power Q represented in the BC treatment by

$$x_{\mathrm{BC}} = \tfrac{2}{3}Q\alpha_G T. \quad (13)$$

Here α_G is the total mosaic width parameter of $w(\Delta\theta) = \alpha_G \exp(-\pi\alpha_G^2\Delta\theta^2)$ and T denotes the average beam path in the sample. In the RED approach an equivalent role is played by

$$x_{\mathrm{RED}} = \tfrac{2}{3}[1-\exp(-QR/\cos\theta)]T/\bar{t}. \quad (14)$$

Putting $A = \tfrac{3}{2}\bar{t}/T$ and assuming numerical equality $\alpha_G/\alpha_{\mathrm{RED}} = R/\bar{t}$ we can express it as

$$x_{\mathrm{RED}} = [1-\exp(-x_{\mathrm{BC}}A/\cos\theta)]/A. \quad (15)$$

The value of x_{RED} can now be used instead of x_{BC} to calculate the extinction correction with the help of the analytical approximation to the ETE solution given by BC (see also Kawamura & Kato, 1983):

$$y^{-1} = \{1+2{\cdot}12x+[ax^2/(1+bx)]\}^{1/2}. \quad (16)$$

In (16) a Gaussian distribution for $w(\theta)$ is assumed and a, b are given by

$$a = 0{\cdot}58+0{\cdot}48\cos 2\theta+0{\cdot}24\cos^2 2\theta,$$

$$b = 0{\cdot}02-0{\cdot}025\cos 2\theta.$$

The practical procedure in calculating y^{-1} can therefore be summarized as follows: first x_{BC} is computed, then corrected for primary extinction by means of (15) (x_{RED} results) and finally the x_{RED} value is inserted for x into (16) which supplies the extinction correction. The $[y(x_{\mathrm{BC}})]^{-1}$ dependence for various values of the parameter A is demonstrated in Fig. 3; the $A = 0{\cdot}01$ curves practically coincide with the BC result for pure secondary extinction.

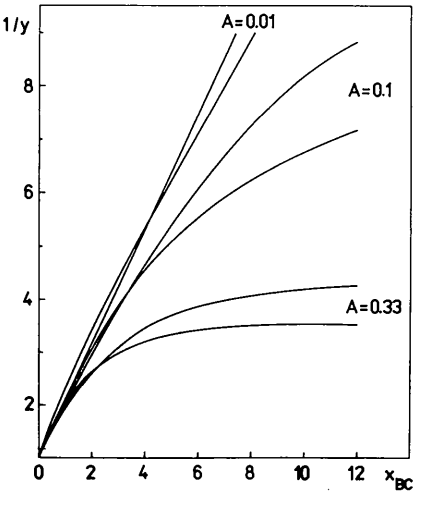

Fig. 3. The dependence of the extinction correction y^{-1} on x_{BC} for various values of the parameter A; for a given A the upper and lower curves correspond to $\sin\theta = 0{\cdot}2$ and $\sin\theta = 0{\cdot}7$, respectively.

The RED model contains two free parameters to be adjusted by fitting in the course of data processing. While α_{RED} has the conventional meaning, A represents the ratio of the mean free path between subsequent reflections to the averaged beam path in the sample and has no analogy in the mosaic model. Its value is a measure of proportion between the effects of primary and secondary extinction. Clearly for A small ($\bar{t} \ll T$) there exists a high probability of frequent returns into a given state in the random-walk sequence. This corresponds to multiple reflections on different domains which in turn have to be small to fit into a crystal of finite dimensions. A case of secondary extinction results and in the limit $A \to 0$ we obtain $x_{\mathrm{RED}} = x_{\mathrm{BC}}/\cos\theta$ from (15). The factor $1/\cos\theta$ arises from the above-mentioned θ dependence of the width of $w_{\mathrm{RED}}(\Delta\theta)$.

On the other hand for $A \gtrsim 1$ (*i.e.* $\bar{t}/T \gtrsim 1$), reflection on a single domain within the whole sample becomes more probable, with the intensity being affected

mainly by primary extinction. In this range (15) yields $x_{RED} \simeq 1/A$, implying the independence of the integrated reflecting power on Q and thus on the structure factor. This rather paradoxical behaviour corresponding to the saturation of the $1/y(x_{BC})$ dependence in Fig. 3 should not be surprising. It is an experimentally well established fact (Kulda, 1984, and references therein) that in the range of slight deformations $r=1$ [see equation (5)] and the integrated reflectivity is determined solely by variation of the local misorientation along the beam path in the crystal.

5. Discussion

For the sake of simplicity the above description of wave propagation was based on Hamilton–Zachariasen rather than Kato's theory. The beam transport in the sample was considered as a series of mutually incoherent reflections on well separated elastically deformed domains. Although the extent to which this concept coincides with reality is questionable, some experimental justification can be found in works on multiple reflections of neutrons in elastically deformed crystals (Mikula, Michalec, Chalupa, Sedláková & Petržílka, 1975; Kulda, Mikula, Vrána, Michalec & Vávra, 1981; Vrána, Mikula, Michalec, Kulda & Vávra, 1981; Kulda & Mikula, 1985). This simplification enabled us to take into account within a single domain multiple phase correlations of any order so that primary extinction is covered without limitation.

Contrary to the mosaic model the RED approach emphasizes the deformed parts of the crystal. The reason for this is not only a matter of mathematical convenience. As can be observed on diffraction topographs (*e.g.* Lang, 1978; Baruchel, Schlenker, Zarka & Petroff, 1978) the greatest part of the diffracted intensity (direct extinction contrast) originates from the surroundings of lattice defects, which are elastically deformed and not fine grained. Moreover, the subgrain boundaries are formed by dislocation networks accompanied by lattice strains so that the second assumption of RED is also close to reality.

On the other hand, it is clear that undisturbed parts are present in real crystals as well. They can be represented by $\Delta\theta = \text{constant}$ segments in the RED sequence corresponding to some p_0 transition probability ($p_- + p_0 + p_+ = 1$). It can be proved in an elementary way that the inclusion of such inactive segments will not change the stationary probability distribution $P_N(j)$ and the only correction has to be made in the definition of $\bar{t}$ (but not τ) replacing the previous one by

$$\bar{t} = (\pi N/2)^{1/2} \bar{l}/(1-p_0).$$

For fixed N and $\bar{l}$ the mean free path will become $1/(1-p_0)$ times larger and the parameter A will be rescaled at the same ratio indicating an increase of primary extinction. From the point of view of diffraction theory such an approach is valid as long as the deformation is slight and the adiabatic approximation holds. In the opposite case the traditional mosaic model completed by a proper dynamical description of diffraction in individual blocks would naturally be the most appropriate one.

For practical reasons we tried to follow approximately the lines of the BC treatment, one of the standard approaches to extinction model fitting. All the modifications necessary to adapt the correction procedure from the mosaic model to RED can be performed on the basis of (15). A practical consequence of the presence of a significant value of A will be a saturation of the $1/y(x_{BC})$ dependence above some x_{BC} value preventing extinction overestimation at very strong reflections. In the limit of pure secondary extinction the results of the fit should be insensitive to the model choice. The true form of the additional factor $1/\cos\theta$ in $w_{RED}(\Delta\theta)$ remains a matter of discussion. In § 3 it appeared in a completely correct way; however, both the mosaic model and RED up to this stage assume lattice-plane misorientations only. In reality, any deformation also involves changes of the interplanar spacing whose effect in the Bragg condition is complementary to misorientation, being proportional to $\tan\theta$. The θ independence of $w(\Delta\theta)$ takes this situation into account to a certain extent. Similarly, in the framework of RED it is possible to employ in (12) instead of $\cos\theta/R$ a more complicated expression combining both types of angular variation. The necessity of this step may depend on the sample type and the data being fitted, therefore it should be discussed together with further possible extensions of the RED model against a background of practical experience.

6. Concluding remarks

A new model for the description of diffraction phenomena in real crystals was presented. Analogously to the traditional treatment it is based on the solution of the energy transport equations. The crystal is assumed to consist of elastically deformed domains rather than perfect blocks. In this way the elementary reflections can be described within the quasiclassical approximation of the dynamical theory so that primary extinction can be covered with sufficient accuracy. In the simplest variant the RED model relies on two parameters being adjusted by fitting to a given intensity data set. One is equivalent to the conventional mosaic-width parameter for a Gaussian distribution while the other is completely new and represents the proportion of primary and secondary extinction. The comparison of the true efficiency of this new approach to other existing extinction models remains a matter for extensive

experimental tests. We hope to be able to report on some results of these in the near future.

I express my sincere thanks to Dr R. Michalec for his kind support, to Drs P. Mikula, Z. Kosina, V. Petříček and many other colleagues for helpful discussions and finally to Mr A. Dvořák for his help with the preparation of the manuscript.

References

BALIBAR, F., CHUKHOVSKII, F. N. & MALGRANGE, C. (1983). *Acta Cryst.* A**39**, 387-399.
BARUCHEL, J., SCHLENKER, M., ZARKA, A. & PETROFF, J. F. (1978). *J. Cryst. Growth*, **44**, 356-361.
BECKER, P. & COPPENS, P. (1974). *Acta Cryst.* A**30**, 129-147, 148-153.
COOPER, M. J. & ROUSE, K. D. (1970). *Acta Cryst.* A**26**, 214-223.
DARWIN, C. G. (1922). *Philos. Mag.* **43**, 800-829.
FELLER, W. (1970). *An Introduction to Probability Theory and Its Applications*, Vol. 1, ch. 15. New York: Wiley.
HAMILTON, W. C. (1957). *Acta Cryst.* **10**, 629-634.
KATO, N. (1976). *Acta Cryst.* A**32**, 453-457, 458-466.
KATO, N. (1979). *Acta Cryst.* A**35**, 9-16.
KATO, N. (1980). *Acta Cryst.* A**36**, 171-177.
KAWAMURA, T. & KATO, N. (1983). *Acta Cryst.* A**39**, 305-310.
KULDA, J. (1984). *Acta Cryst.* A**40**, 120-126.
KULDA, J. & MIKULA, P. (1985). *Phys. Status Solidi A*, **92**, 95-100.
KULDA, J., MIKULA, P., VRÁNA, M., MICHALEC, R. & VÁVRA, J. (1981). *Nucl. Instrum. Methods*, **180**, 89-92.
LANG, A. R. (1978). *Diffraction and Imaging Techniques in Material Science*, edited by S. AMELINCKX & A. R. LANG, Vol. 2. Amsterdam: North-Holland.
MESSIAH, A. (1962). *Quantum Mechanics*, Vol. 2, ch. 17. Amsterdam: North-Holland.
MIKULA, P., MICHALEC, R., CHALUPA, B., SEDLÁKOVÁ, L., PETRŽÍLKA, V. (1975) *Acta Cryst.* A**31**, 688-693.
PINSKER, Z. G. (1978). *Dynamical Scattering of X-rays in Crystals*. Berlin: Springer.
RAUCH, H. & PETRASCHECK, D. (1978). *Neutron Diffraction*, edited by H. DACHS, pp. 303-351. Berlin: Springer.
SCHNEIDER, J. R. (1975). *J. Appl. Cryst.* **8**, 195-201.
TAKAGI, S. (1962). *Acta Cryst.* **15**, 1311-1312.
TAKAGI, S. (1969). *J. Phys. Soc. Jpn*, **26**, 1239-1253.
TAUPIN, D. (1964). *Bull. Soc. Fr. Minéral. Cristallogr.* **87**, 469-511.
VRÁNA, M., MIKULA, P., MICHALEC, R., KULDA, J. & VÁVRA, J. (1981). *Acta Cryst.* A**37**, 459-465.
ZACHARIASEN, W. H. (1967). *Acta Cryst.* **23**, 558-565.
ZACHARIASEN, W. H. (1968). *Acta Cryst.* A**24**, 421-424.

Acta Cryst. (1987). A**43**, 173-179

The Phases of Forbidden Reflections*

BY BEN POST†

Physics Department, Polytechnic University of New York, Brooklyn, New York, USA

AND JOSHUA LADELL

Philips Laboratories, North American Philips Corporation, Briarcliff Manor, New York 10510, *USA*

(*Received* 6 *June* 1986; *accepted* 30 *September* 1986)

Dedicated to the memory of Professor Paul P. Ewald

Abstract

The invariant phases of large numbers of germanium triplets, consisting of one forbidden and two permitted reflections, have been determined experimentally. The forbidden reflections include members of the forms {200}, {222}, {420} and {442}. Phase effects in triplets containing members of the ultra-weak (forbidden) {622} and {640} were detected but were too weak to provide reliable phase indications. The phases of all triplets which include a forbidden reflection and the $\bar{3}\bar{1}\bar{1}$ reflection are observed to be negative. The phases of individual forbidden reflections, whose indices are described as summing to $(4n-2)$, are equal to $(-1)^n$. The imaginary part of the dispersion correction to the atomic form factors is relatively large (0·89 for Cu $K\alpha_1$ radiation): it makes significant contributions to the structure factors and the phases of the vanishingly weak forbidden reflections.

I. Introduction

Reflections whose indices sum to $4n-2$ are 'forbidden' in diamond-type crystals. Their structure factors, calculated for atoms in positions 8(*a*) of space group $Fd3m$ (Henry & Lonsdale, 1952), equal zero. In 1921, however, W. H. Bragg detected intensities diffracted by the forbidden 222 reflection of a diamond crystal.

* This paper should be regarded as forming part of the Ewald Memorial Issue of *Acta Cryst.* Section A published in November 1986. The manuscript was received in its final form too late for inclusion.

† Work supported by the National Science Foundation, Division of Materials Research, Grant DMR 83115325, and in part by the Joint Services Electronics Program, Contract no. F49620-82-C-0084.

0108-7673/87/020173-07$01.50 © 1987 International Union of Crystallography