Extinction in mosaic crystals

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Exact formal solutions of the transport equations which govern the flow of current in a mosaic crystal set at a Bragg condition are given. These solutions are applied to a crystal cut in the shape of a parallelepiped. A formula is derived for the extinction coefficient for this crystal shape. We call this result the "*AB*-extinction formula". It will provide the experimentalist with an approximate method to correct for anisotropic extinction resulting from a crystal shape anisotropy. It is suggested that the exact results of the dynamical diffraction theory for slabs should be used to correct for primary extinction. The mathematical and conceptual errors in the theory given by Zachariasen are pointed out.

I. INTRODUCTION

Extinction is a central problem in the theory of diffraction in real crystal. It is a direct consequence of multiple Bragg scattering. One knows the fundamental equations which must be solved: the Schrödinger equation for neutron and electron diffraction and Maxwell's equations for x-ray diffraction. However, difficulties arise on two levels in developing a rigorous mathematical attack on the problem. First, a description of the type and geometrical array of imperfections which lead to incoherence of the waves emanating from various macroscopic regions of the crystal must be given; and then, once a model of the imperfections is assumed, a boundary-value problem must be solved.

There is known to be a manifold of imperfections that occur in real crystals, and their effects on the diffracted intensity is, in general, difficult to estimate. In addition, detailed information on the imperfection structure of a given crystal is generally not available. A method which has been widely used, and which has some experimental foundation, is the mosaic crystal model originally proposed by Darwin.[1] The idea is essentially that a real crystal is composed of many small perfect crystal regions, called mosaic grains, which are misoriented with respect to each other by small angles. The scattering cross section of each grain can be calculated using kinematical diffraction theory (Born approximation). Since it is assumed that there are many such randomly oriented grains in any macroscopic region of the crystal, the scattered waves can be added incoherently at any point distant from the region in which the scattered waves originate. The extent to which the basic physical requirements of this model of an "ideally imperfect" crystal can be relaxed to include primary extinction is not known. We will investigate this briefly in Sec. IV. The method by which coherent effects between relatively large adjacent grains is to be included in the theory is also not known.

An attempt to derive a general formula for extinction in real crystals which encompasses the entire range of crystalline perfection was made by Zachariasen.[2,3] Unfortunately, there are a number of conceptual and mathematical errors in those papers, some of which have been pointed out by Werner[4] and Zigan.[5] The purpose of this paper is to place the theory of extinction in mosaic crystals on firmer physical and mathematical ground; and in particular, to give the first exact solution to the extinction problem for any finite three-dimensional crystal. For completeness and clarity we review the general mathematical techniques developed previously (Werner and Arrott,[6] Werner, Arrott, King, and Kendrick[7]) in Sec. II. The exact solution for a particular case is given in Sec. III. The significance of this result, and the primary extinction problem are discussed in Sec. IV. Suggestions for the extension of the theory presented here, along with various conclusions, are given in Sec. V.

II. BASIC EQUATIONS

We assume that a real crystal is adequately described by Darwin's mosaic model. The derivations given here are intended for neutron diffraction; however, the results apply equally well to x-ray diffraction.

A. Differential equations

Consider a crystal of arbitrary (convex) shape (as shown in Fig. 1) immersed in a nominally collimated and monoenergetic beam of neutrons of uniform incident intensity J_0. J_0 is the number of neutrons/cm^2/sec/unit volume in k space incident on the crystal. We define $\bar{\sigma}$ as the probability per unit path length for Bragg scattering. The bar indicates it is an average quantity.

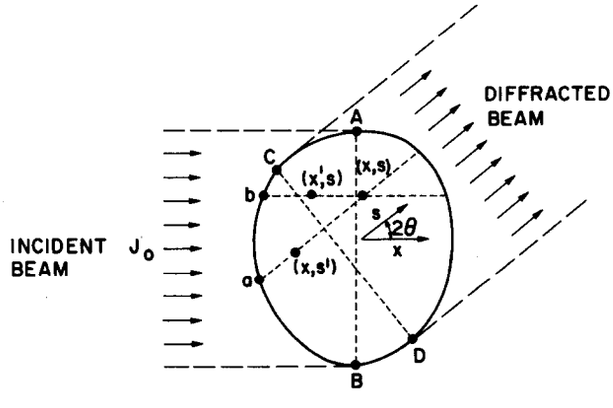

FIG. 1. Schematic diagram of a crystal of arbitrary (convex) shape. The coordinates (x,s) of an arbitrary point inside this crystal are given in the oblique coordinate system shown here, where 2θ is the scattering angle. The incident beam enters the crystal through the boundary between A and B on the left, and the diffracted beam leaves the crystal through the boundary between C and D on the right.

The macroscopic region over which this average is made is assumed to be small in comparison to the size of the crystal. We assume $\bar{\sigma}$ is independent of position. If a neutron of initial wave vector $\mathbf{k}$ is Bragg scattered in a given mosaic grain, we assume that the diffracted wave vector is exactly $\mathbf{k}+\mathbf{G}$, where $\mathbf{G}$ is the reciprocal lattice vector. That is, we neglect refraction and finite size broadening effects. In addition, we assume the probability per unit path for this diffracted neutron to be rediffracted somewhere along its exit path is identical to $\bar{\sigma}$. That is,

$$\bar{\sigma}(\mathbf{k})=\bar{\sigma}(\mathbf{k}+\mathbf{G}). \tag{1}$$

$\bar{\sigma}$ will, of course, depend on the angular distribution of mosaic grains, commonly called W. It is clear that there will be dynamic interchange of neutron current between the incident and diffracted beams inside the crystal. If we define J_i as the *current density* in the incident direction and J_d as the *current density* in the diffracted beam direction at an arbitrary point (x, s) in the oblique coordinate system shown in Fig. 1, the balance relations are[8]

$$\frac{\partial J_i}{\partial x}=-\bar{\sigma}_t J_i+\bar{\sigma}J_d, \tag{2a}$$

$$\frac{\partial J_d}{\partial s}=-\bar{\sigma}_t J_d+\bar{\sigma}J_i. \tag{2b}$$

Here

$$\bar{\sigma}_t=\bar{\sigma}+\mu, \tag{3}$$

where μ is the probability per unit path for attenuation by all non-Bragg processes, i.e., true absorption, incoherent and inelastic scattering. These equations were first written by Hamilton[9] as a generalization to the *total current* equations of Darwin.[1] (Hamilton solved these equations numerically for certain special cases.) These equations must be solved subject to the boundary conditions

$$J_i=J_0, \text{ on the boundary between } A \text{ and } B \tag{4a}$$

$$J_d=0, \text{ on the boundary between } C \text{ and } D. \tag{4b}$$

Combining Eqs. (2a) and (2b) we have

$$\frac{\partial^2 J_d}{\partial x\,\partial s}+\bar{\sigma}_t\left(\frac{\partial J_d}{\partial x}+\frac{\partial J_d}{\partial s}\right)+(\bar{\sigma}_t^2-\bar{\sigma}^2)J_d=0. \tag{5}$$

This is the equation that must be solved. An identical equation holds for J_i.

For the coordinate system in which Zachariasen wrote Eqs. (2a) and (2b), an additional term must be added to the left-hand side. The coordinates used there were

$$t_1=x-x_b(s), \tag{6a}$$

$$t_2=s-s_a(x), \tag{6b}$$

where $x_b(s)$ is the value of x at the boundary point b, and $s_a(x)$ is the value of s at the boundary point a. That is, $x_b(s)$ or $s_a(x)$ are the equations of the boundary. From the usual rules for chain differentiation, the balance relations in this coordinate system are

$$\frac{\partial J_i}{\partial t_1}-\frac{\partial J_i}{\partial t_2}\frac{\partial s_a(x)}{\partial x}=-\bar{\sigma}_t J_i+\bar{\sigma}J_d, \tag{7a}$$

$$\frac{\partial J_d}{\partial t_2}-\frac{\partial J_d}{\partial t_1}\frac{\partial x_b(s)}{\partial s}=-\bar{\sigma}_t J_d+\bar{\sigma}J_i. \tag{7b}$$

The second term on the left-hand side of these equations was neglected by Zachariasen.

B. Integral equations

We now rewrite the pair of coupled differential equations (2a) and (2b) in integral form. This can, of course, be done in a strictly mathematical manner; however, we will do it in a more physical way as was done previously by Werner and Arrott.[6] Consider the line connecting the point a on the boundary and the point (x, s) in Fig. 1. Neutrons arriving at the point (x, s) and traveling in the direction of the diffracted beam originated by diffraction from the incident beam at any of the points (x, s') along this line, but with a probability attenuated by the factor $\exp[-\bar{\sigma}_t(s-s')]$. Thus,

$$J_d(x, s)=\int_{s_a(x)}^{s} J_i(x, s')\exp[-\bar{\sigma}_t(s-s')]\,\bar{\sigma}\,ds'. \tag{8a}$$

Consider now how neutrons which contribute to $J_i(x, s)$ arrive at the point (x, s). The probability that a neutron arrives directly at the point (x, s) from the entrant point b is simply $\exp\{-\bar{\sigma}_t[x-x_b(x)]\}$. Neutrons traveling in the incident beam direction arrive at (x, s) by diffraction of the diffracted beam at any point (x', s) along the line connecting b and (x, s). Thus,

$$J_i(x, s)=J_0\exp\{-\bar{\sigma}_t[x-x_b(s)]\}+\int_{x_b(s)}^{x} J_d(x', s)\exp[-\bar{\sigma}_t(x-x')]\,\bar{\sigma}\,dx'. \tag{8b}$$

Inserting Eq. (8b) into (8a) we have the integral equation

$$J_d(x, s)=J_0\int_{s_a(x)}^{s}\exp\{-\bar{\sigma}_t[x-x_b(s')+s-s']\}\,\bar{\sigma}\,ds'+\int_{s_a(x)}^{s}\bar{\sigma}\,ds'\int_{x_b(s')}^{x}\bar{\sigma}\,dx'\,J_d(x', s')\times\exp[-\bar{\sigma}_t(s-s'+x-x')]. \tag{9}$$

A similar equation can be written for $J_i(x, s)$.

C. General solution of differential equations

We seek a solution of Eq. (5) of the form

$$J_d(x, s)=\exp[-\bar{\sigma}_t(x+s)]f_d(x, s). \tag{10}$$

Inserting this into (5), we find that we have a simpler equation to solve:

$$\frac{\partial^2 f_d}{\partial x\,\partial s}-\bar{\sigma}^2 f_d=0. \tag{11}$$

This equation is separable. If we denote the separation constant by K, the general solution is

$$f_d(x, s)=\int dK\,F_d(K)\exp[K\bar{\sigma}x+(1/K)\bar{\sigma}s], \tag{12}$$

where the amplitude function $F_d(K)$ is to be determined from the boundary conditions. Since J_i satisfies an equation identical to (5) we must have

$$f_i(x, s)=\int dK\,F_i(K)\exp[K\bar{\sigma}x+(1/K)\bar{\sigma}s], \tag{13}$$

where

$$J_i(x, s)=\exp[-\bar{\sigma}_t(x+s)]f_i(x, s). \tag{14}$$

Equations (2a) and (2b) require

$$F_i(K)=F_d(K)/K. \tag{15}$$

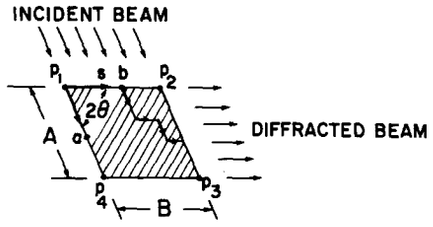

FIG. 2. Mosaic crystal cut in the shape of a parallelepiped. A typical path by which neutrons enter and leave the crystal after making three Bragg reflections is shown.

The contour of integration in the K plane is to be determined. Rewriting f_d in the form

$$f_d(x,s)=\int dK F_d(K)\exp\{\sigma(xs)^{1/2}[K(x/s)^{1/2}+(1/K)(s/x)^{1/2}]\}, \tag{16}$$

and using the expansion

$$\exp\left[y\left(\mu+\frac{1}{\mu}\right)\right]=\sum_{n=-\infty}^{\infty}\mu^n I_n(2y), \tag{17}$$

we have

$$f_d(x,s)=\sum_{n=-\infty}^{\infty}a_n\left(\frac{x}{s}\right)^{n/2}I_n[2\bar{\sigma}(xs)^{1/2}]. \tag{18}$$

Here

$$a_n\equiv\int dK K^n F_d(K), \tag{19}$$

and the I_n are the modified Bessel functions. Thus, the general solution of Eq. (5) is

$$J_d(x,s)=\exp[-\bar{\sigma}_t(x+s)]\sum_{n=-\infty}^{\infty}a_n\left(\frac{x}{s}\right)^{n/2}I_n[2\bar{\sigma}(xs)^{1/2}]. \tag{20}$$

The expansion coefficients a_n are to be determined from the boundary conditions. Note that the result given by Zachariasen[2] in his Eqs. (14a) and 14(b) is not the general solution.

D. Formal solution of integral equations

We look for a solution of the integral equation (9) in the form given by Eq. (10). Equation (9) then becomes

$$f_d(x,s)=\bar{\sigma}J_0\int_{s_a(x)}^{s}ds'\exp\{\bar{\sigma}_t[x_b(s')+s']\}+\bar{\sigma}^2\int_{s_a(x)}^{s}ds'\int_{x_b(s')}^{x}dx' f_d(x',s'). \tag{21}$$

This equation can be written in operational form as

$$f_d=\bar{\sigma}g_d+\bar{\sigma}^2 L f_d, \tag{22}$$

where L is the linear integral operator defined by

$$Lf=\int_{s_a(x)}^{s}ds'\int_{x_b(s')}^{x}dx' f(x',s'), \tag{23}$$

and g_d is the known function

$$g_d=J_0\int_{s_a(x)}^{s}ds'\exp\{\bar{\sigma}_t[x_b(s')+s']\}. \tag{24}$$

Equation (22) is of the Fredholm type. The solution is

$$f_d=(1-\bar{\sigma}^2L)^{-1}\bar{\sigma}g_d=\bar{\sigma}g_d+\bar{\sigma}^3Lg_d+\bar{\sigma}^5L^2g_d+\bar{\sigma}^7L^3g_d+\cdots. \tag{25}$$

Thus, the diffracted current density at any interior point (x, s) is

$$J_d(x,s)=\exp[-\bar{\sigma}_t(x+s)]\sum_{n=0}^{\infty}\bar{\sigma}^{2n+1}L^n g_d. \tag{26}$$

$L^n g_d$ means n successive applications of the operator L to the function g_d. The interpretation of this solution is straightforward: The first term in the series gives the contribution to the diffracted current density at (x, s) due to once-reflected neutrons, the second term gives the contribution to J_d from three-times reflected neutrons, and so on.

Equations (20) and (26) must be equal. We therefore have two mathematical prescriptions to calculate the diffracted current density and subsequently the extinction coefficient for a crystal of any geometry. Detailed calculations based on both techniques are carried out in Sec. III.

III. QUARTER-SPACE PROBLEM

Consider a mosaic crystal cut in the shape of a parallelepiped as shown in Fig. 2. The dimensions of the crystal are taken to be $A\times B\times C$. We illustrate the application of the solutions (20) and (26) by calculating the extinction coefficient for this crystal geometry.

A. Application of general solution of differential equations

The boundary conditions (4a) and (4b) require

$$J_i(0,s)=J_0, \tag{27a}$$

$$J_d(x,0)=0. \tag{27b}$$

Using Eqs. (13)–(15), we find that the boundary condition (27a) requires

$$\exp(-\bar{\sigma}_t s)\int dK[F_d(K)/K]\exp(\bar{\sigma}s/K)=J_0. \tag{28}$$

Using Eqs. (10) and (12), we find that the boundary condition (27b) requires

$$\int dK F_d(K)\exp(K\bar{\sigma}x)=0. \tag{29}$$

Expanding the exponentials in Eqs. (28) and (29), and using the definition of the expansion coefficients a_n given in Eq. (19) we find

$$\begin{aligned} a_n&=0, && \text{for } n\geq 0\\ &=(\bar{\sigma}/\bar{\sigma}_t)^{n+1}J_0, && \text{for } n<0. \end{aligned} \tag{30}$$

Thus, according to Eq. (20) the diffracted current density at an arbitrary point (x, s) is

$$J_d(x,s)=J_0\exp[-\bar{\sigma}_t(x+s)]\sum_{n=1}^{\infty}\left(\frac{\bar{\sigma}_t}{\bar{\sigma}}\right)^{n-1}\left(\frac{s}{x}\right)^{n/2}I_n[2\bar{\sigma}(xs)^{1/2}]. \tag{31}$$

In order to calculate the extinction coefficient, we must evaluate the *total neutron current* leaving the crystal across the exit boundary. We must integrate the expression (31) for J_d along the line connecting the corner points p_2 and p_3 (Fig. 2), that is

$$P_d=C\sin 2\theta\int_0^A J_d(x,B)dx. \tag{32}$$

Here, C is the height of the crystal, and the factor $\sin 2\theta$ occurs because the line connecting p_2 and p_3 is not normal to the diffracted beam direction. This integral is easily performed; we get

$$P_d = J_0 C\,\bar{\sigma}_t^{-1} \sin 2\theta \sum_{m=0}^{\infty} \left(\frac{\bar{\sigma}}{\bar{\sigma}_t}\right)^{2m+1} H_m(\bar{\sigma}_t A) H_m(\bar{\sigma}_t B), \quad (33)$$

where the function H_m is given by

$$H_m(u) = \frac{1}{m!}\int_0^u v^m \exp(-v)\,dv$$
$$= 1 - \exp(-u)\left(1 + u + \frac{u^2}{2!} + \cdots + \frac{u^m}{m!}\right). \quad (34)$$

Before discussing the significance of Eq. (33) and calculating the extinction coefficient, we will apply the integral techniques of Sec. II D to obtain the same result.

B. Application of solution of integral equations

For this problem the application of the result (26) is straightforward. We have

$$x_b(s) = 0, \quad (35a)$$

$$s_a(x) = 0. \quad (35b)$$

Therefore, the function g_d is

$$g_d = J_0 \int_0^s \exp(\bar{\sigma}_t s')\,ds'$$
$$= J_0\,\bar{\sigma}_t^{-1}[\exp(\bar{\sigma}_t s) - 1]. \quad (36)$$

Consequently, the diffracted current density at any interior point (x, s) due to once-reflected neutrons is

$$J_d^{(1)} = J_0 \exp[-\bar{\sigma}_t(x+s)](\bar{\sigma}/\bar{\sigma}_t)[\exp(\bar{\sigma}_t s) - 1]. \quad (37)$$

Applying the operator L to g_d, we get

$$L\,g_d = J_0\,\bar{\sigma}_t^{-1}\int_0^x dx' \int_0^s ds'[\exp(\bar{\sigma}_t s') - 1]$$
$$= J_0\,\bar{\sigma}_t^{-1}\{x\bar{\sigma}_t^{-1}[\exp(\bar{\sigma}_t s) - 1] - xs\}. \quad (38)$$

Therefore, the diffracted current density at the point (x, s) due to three-times-reflected neutrons is

$$J_d^{(3)} = J_0 \exp[-\bar{\sigma}_t(x+s)]\,\bar{\sigma}^3\,\bar{\sigma}_t^{-1}\{x\bar{\sigma}_t^{-1}[\exp(\bar{\sigma}_t s) - 1] - xs\}. \quad (39)$$

We find for $(2m+1)$-times-reflected neutrons

$$J_d^{(2m+1)} = J_0 \exp[-\bar{\sigma}_t(x+s)]\,\bar{\sigma}^{2m+1}\,\bar{\sigma}_t^{-1}$$
$$\times\left(\frac{\exp(\bar{\sigma}_t s) - 1}{\bar{\sigma}_t^m}\,\frac{x^m}{m!} - \sum_{k=1}^{\infty}\frac{x^m s^k}{m!\,k!\,\bar{\sigma}_t^{m-k}}\right). \quad (40)$$

The second term is zero for $m = 0$. To calculate the diffracted neutron current leaving the crystal which results from $(2m+1)$ Bragg reflections, we must integrate $J_d^{(2m+1)}$ on the boundary from point p_2 to point p_3 (Fig. 2). That is,

$$P_d^{(2m+1)} = C \sin 2\theta \int_0^A J_d^{(2m+1)}(x, B)\,dx. \quad (41)$$

We get

$$P_d^{(2m+1)} = J_0 C\,\bar{\sigma}_t^{-1} \sin 2\theta\,(\bar{\sigma}/\bar{\sigma}_t)^{2m+1} H_m(\bar{\sigma}_t A) H_m(\bar{\sigma}_t B), \quad (42)$$

where H_m is defined by Eq. (34). Since the total diffracted current is

$$P_d = \sum_{m=0}^{\infty} P_d^{(2m+1)}, \quad (43)$$

we see that this result is identical to Eq. (33), as it must be.

C. Extinction coefficient ϵ

In order to calculate the integrated diffracted current, one must know (or assume) the functional form of the mosaic distribution function $W(\Delta)$, and also make some assertions on the extent of the primary extinction within a mosaic grain. To start, we will assume that there is no primary extinction, and that W as a function of the mosaic grain orientation angle Δ is given by

$$W(\Delta) = 1/2\sqrt{3}\,\eta, \quad \text{for } |\Delta| \leq \sqrt{3}\,\eta$$
$$= 0, \quad \text{for } |\Delta| > \sqrt{3}\,\eta. \quad (44)$$

This distribution was used by Hamilton.[9] We call η the mosaic spread. In the absence of primary extention

$$\bar{\sigma} = QW, \quad \text{(Ref. 1)} \quad (45)$$

where

$$Q = \lambda^3 |F_G|^2 / V_{\text{cell}}^2 \sin 2\theta_B. \quad (46)$$

The symbols here have their usual meanings: λ is the neutron wavelength, F_G is the structure factor (including the Debye-Waller factor), V_{cell} is the volume of a unit cell, and θ_B is the Bragg angle.

The integrated diffracted current is defined by

$$R \equiv \int d^3k_0 \int d\Delta\, P_d. \quad (47)$$

In the kinematical limit, where $\bar{\sigma}_t A$ and $\bar{\sigma}_t B$ are small, the integrated diffracted current is

$$R_0 = I_0 QV, \quad (48)$$

which is independent of the functional form of $W(\Delta)$. I_0 is the total *current density* incident on the sample, namely

$$I_0 = \int d^3k_0 J_0. \quad (49)$$

V is the sample volume. For our problem, it is easy to establish Eq. (48) using Eq. (42). For our problem

$$V = ABC \sin 2\theta. \quad (50)$$

If we make the additional assumption that μ is large in comparison to $\bar{\sigma}$, then the integrated diffracted current can be modified by an absorption coefficient

$$\gamma(\mu) \equiv (1/V)\int \exp(\mu t)\,dv, \quad (51)$$

where t is the path length from the entrant point to the exit point for diffraction from the volume element dv. For our problem

$$\gamma(\mu) = [1 - \exp(-\mu A)][1 - \exp(-\mu B)]/\mu^2 AB, \quad (52)$$

and

$$R_0' = \gamma(\mu) I_0 QV, \text{ for small } \bar{\sigma}A \text{ and } \bar{\sigma}B, \text{ and } \mu \gg \bar{\sigma}. \quad (53)$$

We should point out two facts which result from these rather simple considerations:

(i) Factoring-out of an absorption coefficient γ which depends only on absorption (i.e., μ) and not on scattering (i.e., $\bar{\sigma}$) can only be done under the restrictive condition that $\mu \gg \bar{\sigma}$. This condition is seldom met in neutron diffraction in single crystals.

(ii) A calculation of the integrated diffracted current using only once-reflected neutrons gives a somewhat more accurate result than the kinematical formula (53), since it includes attenuation due to Bragg scattering. The result for our problem is

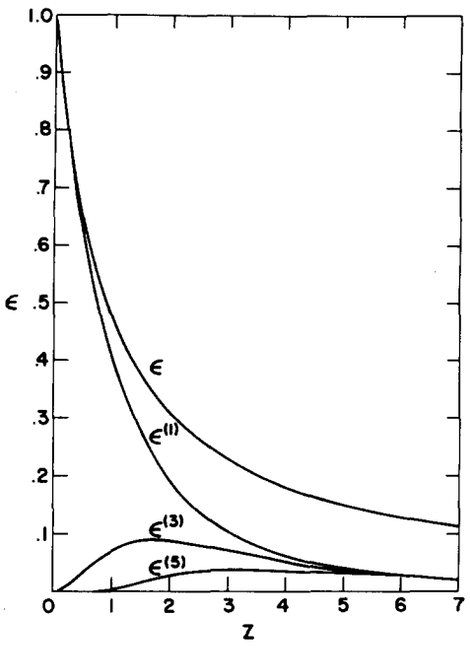

FIG. 3. Contributions to the extinction coefficient ϵ from once-, three-, and five-times-reflected neutrons as given by Eq. (64). The abscissa $z=\bar{\sigma}A=\bar{\sigma}B$.

$$R^{(1)}=I_0 C \sin 2\theta \int d\Delta\,(\sigma/\sigma_t^2)\,[1-\exp(-\sigma_t A)]\,[1-\exp(-\sigma_t B)]. \tag{54}$$

If we assume that the mosaic distribution is given by (44) and call

$$\sigma_0 = Q/2\sqrt{3}\,\eta, \tag{55}$$

then, the integral in (54) is simply

$$R^{(1)}=2\sqrt{3}\,\eta\, I_0 C \sin 2\theta[\sigma_0/(\mu+\sigma_0)^2] \times\{1-\exp[-(\mu+\sigma_0)A]\}\,\{1-\exp[-(\mu+\sigma_0)B]\}. \tag{56}$$

Note that when $\sigma_0/\mu \ll 1$ we recover Eq. (53).

By definition, the extinction coefficient ϵ is the ratio of the observed integrated diffracted current to that which would be observed if there were no absorption and no multiple Bragg scattering. Thus,

$$\epsilon \equiv R_0^{-1} \int d^3k_0 \int d\Delta P_d. \tag{57}$$

The integration over the distribution of incident wave vectors cancels in this expression (as long as the incident beam is nominally monoenergetic and collimated). Thus, if we simply set $J_0=1$ in all of the equations of Secs. III A and III B, the definition above reads

$$\epsilon \equiv \int d\Delta\, P_d/QV. \tag{58}$$

A sound method for computer calculation of ϵ is to calculate the individual contributions to ϵ from once-, three-times-, five-times-, ..., $(2m+1)$-times-reflected neutrons, and then form the sum. That is

$$\epsilon=\sum_{m=0}^{\infty}\epsilon^{(2m+1)}=(QV)^{-1}\sum_{m=0}^{\infty}\int d\Delta\, P_d^{(2m+1)}. \tag{59}$$

For our problem, the first two terms of this series are

$$\epsilon^{(1)}=\bar{\sigma}_t^{-2}A^{-1}B^{-1}[1-\exp(-\bar{\sigma}_t A)]\,[1-\exp(\bar{\sigma}_t B)], \tag{60}$$

$$\epsilon^{(3)}=\bar{\sigma}_t^{-2}A^{-1}B^{-1}(\bar{\sigma}_0^2/\bar{\sigma}_t)\,[1-\exp(-\bar{\sigma}_t A)(1+\bar{\sigma}_t A)] \times[1-\exp(\bar{\sigma}_t B)(1+\bar{\sigma}_t B)]. \tag{61}$$

The mth term is

$$\epsilon^{(2m+1)}=\bar{\sigma}_t^{-2}A^{-1}B^{-1}(\bar{\sigma}_0/\bar{\sigma}_t)^{2m}H_m(\bar{\sigma}_t A)\,H_m(\bar{\sigma}_t B). \tag{62}$$

We will call Eqs. (60)—(62) the "AB-extinction" formulas. In these equations $\bar{\sigma}_t=\mu+\bar{\sigma}_0$. If one makes the common assumption that $W(\Delta)$ is a Gaussian function, namely,

$$W(\Delta)=[1/(2\pi)^{1/2}\eta\,]\exp(-\Delta^2/2\eta^2), \tag{63}$$

then the integrations in (59) should be done numerically. It is, of course, possible to expand the exponentials, and do the integrations over Δ term by term analytically; however, we do not pursue this here.

IV. DISCUSSION

In this section we discuss the significance and potential usefulness of the AB-extinction formulas. We also give a brief account of the derivation of the formula for $\bar{\sigma}$ [Eq. (45)] in order to provide some physical insight into the assumptions which underlie the validity of the basic transport equations [(2a), (2b), (8a), and (8b)].

A. AB-extinction formulas

If $A=B$, and the absorption is negligible, Eq. (59) takes on a particularly simple form:

$$\epsilon=\sum_{m=0}^{\infty}\left(\frac{H_m(z)}{z}\right)^2, \quad A=B,\ \mu=0 \tag{64}$$

where

$$z=\bar{\sigma}_0 A=\bar{\sigma}_0 B. \tag{65}$$

This function is shown in Fig. 3. The contributions to ϵ from once-, three-times-, and five-times-reflected neutrons are also shown. Note that for $z\leq 2$ (or for $\epsilon \geq 0.3$), the first three terms give a very accurate (better than 1%) approximation to the sum. For $z\leq 0.2$ (or for $\epsilon \geq 0.8$), the simple linear formula

$$\epsilon = 1-z, \qquad \text{for } z<0.2, \quad \mu=0 \tag{66}$$

is accurate to about 1%.

We would like to suggest a potentially very useful possibility. Since the dimensions A and B in this problem are precisely the average linear dimensions of the crystal along the incident and diffracted beams, respectively, it is likely that the AB-extinction formulas apply with reasonable accuracy to any simple polyhedral crystal, if we interpret A and B to be mean linear dimensions along these two directions. This suggestion can be checked by numerically integrating Eqs. (2a) and (2b) for various crystal shapes. We have not pursued this in any detail.

It is apparent that the extinction coefficient, in general, depends on both crystal shape and setting. For a sphere or a cylinder it is clear that the extinction depends on the scattering angle 2θ. Detailed numerical calculations were carried out by Hamilton[10] for a cylinder. We have found numerically (using Hamilton's results) that the extinction in a cylinder of diameter D can be calculated to an accuracy of better than 3% for

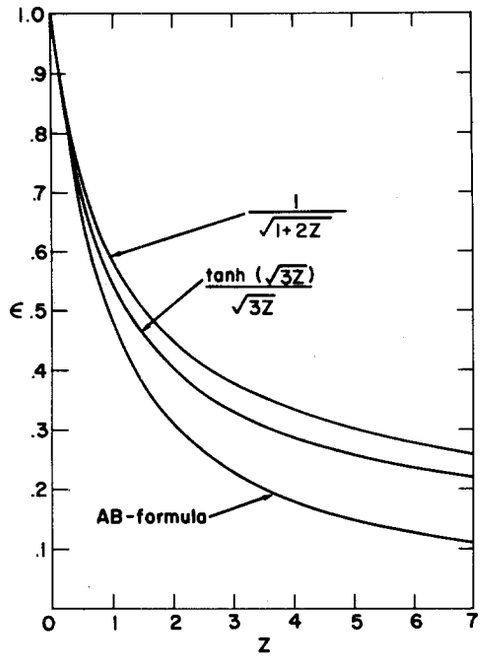

FIG. 4. Comparison of the extinction coefficient for the parallelepiped shown in Fig. 2 (for $A=B$) with the formulas suggested by Zachariasen.

$\bar{\sigma}D < 3$ using formula (64), if we set

$$z = \bar{\sigma}A = \bar{\sigma}D\xi(2\theta), \tag{67}$$

where the angle-dependent factor is

$$\xi(2\theta) = 0.734 + 0.324\exp[0.4624(2\theta)^2] \quad (2\theta \text{ in radians}). \tag{68}$$

Since the series given in Eq. (64) converges very rapidly (only the first few terms are necessary in the range $0 < z < 3$), these equations provide a simple and accurate method for calculating the extinction in a cylindrically shaped crystal.

To obtain a formula for a sphere, we note that the extinction coefficient for a sphere of diameter D is related to the extinction coefficient for a cylinder of diameter D' by

$$\epsilon_{\text{sphere}}(D) = (2/D)\int_0^{D/2} \epsilon_{\text{cylinder}}(D')\,dy, \tag{69}$$

where

$$D'(y) = (D^2/4 - y^2)^{1/2}. \tag{70}$$

The integral can easily be evaluated numerically.

The difficulty one faces in solving the transport equations for a cylinder or a sphere exactly is that the solution must be obtained in a piecewise sense. The reason for this is the fact there are discontinuities in the derivatives of the current densities along certain lines (planes). This problem was dealt with in the paper by Werner, Arrott, King, and Kendrick[7] and has been discussed in detail by Zigan.[5] A comparison of the exact results of this calculation with the suggested extinction formulas of Zachariasen is shown in Fig. 4. We note that if $\epsilon \geq 0.8$, the simple linear formula [Eq. (66)] works. However, when the extinction is large, the deviations between Zachariasen's formulas and the correct results are large.

B. Primary extinction and the formula for $\bar{\sigma}$

The transport equations written in Sec. II, and used in the solution of the problem in Sec. III, apply rigorously only to the secondary extinction problem in ideally imperfect crystals. It is incorrect to assume that they apply inside a perfect crystal region (as assumed by Zachariasen), since the current flow is not along the two directions **x** and **s**. The problem of current flow in perfect crystals has been discussed in many papers, and is a topic of great interest and importance in diffraction theory (see for example Kato[11]) and the review by Batterman and Coles[12], and the references given in these papers).

In order to see how we might modify the analysis of Secs. II and III to include the effects of primary extinction, and to gain some feeling of when these modifications might be valid, we will review the derivation of $\bar{\sigma}$. Derivations along somewhat different lines are given in numerous places, see for example James.[13] We envisage any macroscopic region of the crystal as being made up of many small perfect crystals, slightly misoriented with respect to each other. The macroscopic region under consideration is assumed to be small in comparison to the total crystal volume. The procedure used to calculate $\bar{\sigma}$ is to first calculate the cross section (in the Born approximation) of one of these small perfect crystal grains, and then to integrate this cross section over the angular distribution of mosaic grains.

The scattering cross section $d\sigma/d\Omega$ for a small spherical perfect crystal of radius R is given by Eq. (A8) in the Appendix. This cross section is related to the cross section per unit volume in k space by

$$\frac{d^3\sigma}{dk'^3}(\mathbf{k}\to\mathbf{k}') = \frac{d\sigma}{k^2 d\Omega}\,\delta(k'-k). \tag{71}$$

The delta function $\delta(k-k')$ requires that the scattering be elastic. We describe the orientational distribution of mosaic grains by

$$N(\alpha,\beta) = N_0 W'(\alpha,\beta) \tag{72}$$

(=number of mosaic grains per unit volume per unit orientational solid angle). $W'(\alpha,\beta)$ is the normalized mosaic distribution function; α is an angle in the horizontal scattering plane and β is an angle in the vertical plane. If δV is the average volume of a mosaic grain (which is assumed to be spherical) then

$$\delta V = \tfrac{4}{3}\pi R^3 = 1/N_0. \tag{73}$$

The probability for Bragg scattering per unit path, $\bar{\sigma}(\Delta)$, for a crystal orientation Δ, in a mosaic crystal composed of many of these small perfect crystals is

$$\bar{\sigma}(\Delta) = \frac{1}{\delta V}\int d\alpha\,d\beta\,W'(\alpha,\beta)\int d^3k'\,\frac{d^3\sigma}{dk'^3}(q_{\mathbf{G}}), \tag{74}$$

where

$$\mathbf{q}_{\mathbf{G}} = \mathbf{k}' - \mathbf{k} - \mathbf{G}. \tag{75}$$

k is the incident wave vector, **k'** the wave vector of the scattered beam, and **G** the reciprocal lattice vector. For spherical grains we can perform the integration over **k'**. For a given Bragg reflection we get

$$\bar{\sigma}(\Delta)=\frac{|F_G|^2}{V_{cell}}\frac{6\pi\delta V}{k^2R^2}\int d\alpha\, d\beta\, W'(\alpha,\beta)\,\frac{\sin[RG\cos\theta_B(\Delta-\alpha)]}{RG\cos\theta_B(\Delta-\alpha)}. \tag{76}$$

The integration over β is immediate (giving 1). If we define

$$z\equiv RG\cos\theta_B(\Delta-\alpha), \tag{77}$$

$$W(\alpha)\equiv\int d\beta\, W'(\alpha,\beta), \tag{78}$$

the expression (76) for $\bar{\sigma}(\Delta)$ can be written

$$\bar{\sigma}(\Delta)=\frac{Q}{\pi}\int_{-\infty}^{\infty}dz\, W\left(\Delta-\frac{z}{RG\cos\theta_B}\right)\frac{\sin z}{z}\,dz. \tag{79}$$

Q is defined by Eq. (46). If we assume that W is the Gaussian function as given in Eq. (63), and we make the replacement

$$\sin z/z\rightarrow\exp(-z^2/\pi)\quad\text{(same height and area)}, \tag{80}$$

then

$$\bar{\sigma}(\Delta)=[Q/(2\pi)^{1/2}\eta_{eff}]\exp(-\Delta^2/2\eta_{eff}^2). \tag{81}$$

The effective mosaic spread η_{eff} is related to the actual mosaic spread by

$$\eta_{eff}=\eta\left(\frac{2R^2G^2\cos^2\theta_B\eta^2+\pi}{2R^2G^2\cos^2\theta_B\eta^2}\right)^{1/2}. \tag{82}$$

Although this result has been derived in a somewhat different way, it is the same as Eq. (35) of Zachariasen's paper (Ref. 2). There are two limits of the expression (82): (i) Very small grain size:

$$\eta_{eff}=(\pi/2)^{1/2}(RG\cos\theta_B). \tag{83}$$

(ii) Large mosaic spread η:

$$\eta_{eff}=\eta. \tag{84}$$

Although the derivation of $\bar{\sigma}(\Delta)$ given here (and in Zachariasen's paper) is valid, the use of the formula (81) in the limit (83) in the transport equations (2a) and (2b) is inconsistent with the necessary assumptions. The reason for this is the following: As the grain size R decreases, the natural width of $d\sigma/d\Omega$ increases in all three directions in k space. What one is attempting to do here is to take into account the broadening in one direction, namely perpendicular to **G**, while ignoring the broadening in the direction (in the scattering plane) parallel to **G**. Thus, in the limit where the mosaic grain size is very small, and the width of a Bragg reflection is dominated by this natural linewidth, Eqs. (2a) and (2b) must be rewritten to take this into account. The correct equations are

$$\frac{\partial J_i(x,s,\mathbf{k})}{\partial x}=-J_i(x,s,\mathbf{k})\int d^3\mathbf{k}'\,\bar{\sigma}_t(\mathbf{k}\rightarrow\mathbf{k}')+\int d^3\mathbf{k}'\,\bar{\sigma}(\mathbf{k}'\rightarrow\mathbf{k})J_d(x,s,\mathbf{k}'), \tag{85a}$$

$$\frac{\partial J_d(x,s,\mathbf{k}')}{\partial s}=-J_d(x,s,\mathbf{k}')\int d^3\mathbf{k}\,\bar{\sigma}_t(\mathbf{k}'\rightarrow\mathbf{k})+\int d^3\mathbf{k}\,\bar{\sigma}(\mathbf{k}\rightarrow\mathbf{k}')J_i(x,s,\mathbf{k}), \tag{85b}$$

where

$$\sigma(\mathbf{k}\rightarrow\mathbf{k}')=\frac{1}{\delta V}\int d\alpha\, d\beta\, W'(\alpha,\beta)\,\frac{d^3\sigma}{dk'^3}(\mathbf{k}'-\mathbf{k}-\mathbf{G}). \tag{86}$$

The extraction of J_d from the integral in Eq. (85a) and J_i from the integral in Eq. (85b) is possible only in the case when the domain size is sufficiently large, so that we can assume that the cross section contains the delta function $\delta(\mathbf{k}'-\mathbf{k}-\mathbf{G})$; that is, only when $d\sigma/d\Omega$ is narrow compared to W. It is, of course, possible that after making certain reasonable assumptions about the form of $\bar{\sigma}(\mathbf{k}\rightarrow\mathbf{k}')$ that Eqs. (85a) and (85b) could be solved. We have not pursued this possibility.

The problem raised here was fully appreciated by Darwin.[1] In his preliminary comments on the problem of reflection from a slab-shaped mosaic crystal he states:

"The whole reflection from a deep crystal results from the reflections in the successive layers. The multiplicity is of a different type from that of [the dynamical theory] because rays are not now coherent. The problem of these multiple reflexions would be exceedingly difficult if it were treated exactly: for each layer will, on account of diffraction, spread out incident parallel rays into a certain range of angles and so will continually change the angle at which they attack successive layers. But, if (as assumed) the crystal is so imperfect that diffraction does not change the direction of the rays to an extent comparable with the scale of variations of the blocks, then it will be legitimate to regard the reflected rays as coming plane parallel off the crystal (at an angle exactly 2θ to the incident beam). In consequence of this it will be possible to replace a highly complicated system of integral equations by differential equations of a simple type."

If the average size of a mosaic grain is sufficiently large for the internally scattered waves to be rescattered within the grain, then the kinematical result for the cross section must be modified to include these multiple scattering effects. It is not correct to describe this process using the transport equations of Sec. II since the incident and diffracted waves are coupled together in a long-range coherent manner. One must solve the Schrödinger equation for the neutron and electron cases, and Maxwell's equations for the x-ray case, subject to the appropriate boundary conditions. The solutions to two problems are well-known: (i) Slab-shaped crystal in reflection (Bragg case) and (ii) a slab in transmission (Laue case). The original development of the dynamical theory of diffraction was carried out by Darwin,[14,15] Laue,[16] Ewald,[17,19] Bethe,[20] Prins,[21] and Lamla.[22] The neutron case has been treated many years ago by Goldberger and Sietz.[23] Reviews and extensions of the theory have been given by James,[24] Zachariasen,[25] and Batteman and Coles.[12] Extensive work on the solution of the dynamical diffraction problem for certain polyhedral-shaped crystals has been carried out by Kato[26,27] and by Saka, Katagawa, and Kato.[28,29]

It is clear that the integrated diffracted current per unit volume leaving a given mosaic grain decreases with increasing size as a result of multiple scattering. Thus, the expression (45) for $\bar{\sigma}$ must be decreased by a certain fraction ϵ_p, called the primary extinction coefficient, namely

$$\bar{\sigma}(\Delta)=\epsilon_p Q W(\Delta). \tag{87}$$

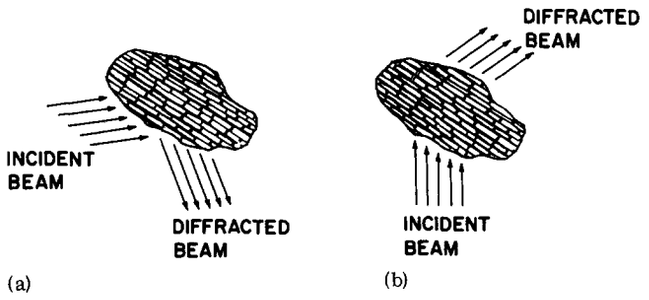

FIG. 5. Schematic diagram of mosaic block orientations used to justify the use of the primary extinction formulas (88) for the Bragg case (a) and the Laue case (b).

The problem is to calculate ϵ_p. It would be important to solve the dynamical diffraction problem for a sphere. This has not been done. In the absence of such a solution, we recommend using the results of the slab problem. This procedure is physically reasonable: Suppose the length of the mosaic grains is somewhat larger than their thickness, and they are oriented as shown schematically in Fig. 5(a). It is then clear that a calculation based on the Bragg-slab geometry would give a fairly accurate estimate of ϵ_p. Alternatively if these grains were oriented as shown in Fig. 5(b), the results of the Laue-slab geometry would be more appropriate. As long as the mosaic grains are numerous and reasonably small, some average of the results of these two configurations should give a result which is approximately correct (and probably functionally nearly exact).

In the limit of small primary extinction ($\epsilon_p \approx 1.0$) and small absorption, the calculations of Zachariasen[25] based on von Laue's[16] dynamical theory for a crystal of thickness t give

$$\epsilon_p = 1 - \frac{1}{3}Q\frac{\sin 2\theta}{\lambda}D^2 + \frac{1}{20}Q^2\frac{\sin^2 2\theta}{\lambda^2}D^4 - \cdots, \quad \text{Laue case}$$

$$= 1 - \frac{1}{3}Q\frac{\sin 2\theta}{\lambda}D^2 + \frac{2}{15}Q^2\frac{\sin^2 2\theta}{\lambda^2}D^4 - \cdots, \quad \text{Bragg case,} \tag{88}$$

where $D = t/\sin\theta$ for the symmetrical Bragg case and $D = t/\cos\theta$ for the symmetrical Laue case. For a mosaic crystal, D should be interpreted as a mean linear dimension of a mosaic grain. The reason why the coefficient of the second term is negative and the coefficient of the third term is positive is clear: The second term results from neutron waves scattered twice, thus contributing to the forward diffracted wave. The third term is positive and results from waves which have been scattered three times, thus increasing the strength of the reflected beam. This sequence of alternating signs must occur in the solution of the dynamical diffraction problem for a crystal of any size or shape. It is also clear that the relevant variable in the expansion is $Q\sin(2\theta)D^2/\lambda$. The numerical coefficients $\frac{1}{3}$, $\frac{1}{20}$, and $\frac{2}{15}$ in Eq. (88) will be geometry dependent. If we accept the suggestion made above, then the average primary extinction coefficient should be taken to be

$$\epsilon_p = 1 - \frac{1}{3}Q\frac{\sin 2\theta_B}{\lambda}D^2 + \frac{11}{120}Q^2\frac{\sin^2 2\theta_B}{\lambda^2}D^4 - \cdots. \tag{89}$$

The attempt to extend the ideas given here to large primary extinction leads to two conceptual difficulties:

(i) If primary extinction is large, the mosaic grain size D may also be large. If D is too large, the assumption of there being numerous randomly placed and oriented grains breaks down, and a calculation of coherent interference effects amongst the grains must be carried out.

(ii) When primary extinction is very large, the neutron current density is highly discontinuous, since a neutron of a given wave vector $\mathbf{k}$ is nearly certain to be Bragg reflected in a given mosaic grain if it is properly oriented. In this limit, the transport equations of Sec. II, which describe J_i and J_d as continuous functions of the coordinates (x, s), break down. To some extent this difficulty can be conceptually circumvented if the mosaic distribution function is broad, and we redefine the current densities J_i and J_d to include neutrons contained in a fairly wide band of angles and wavelengths.

In the limit of a macroscopic perfect crystal, the prescription given by (87) leads to the correct result, since ϵ_p is then very small, and $\bar{\sigma}$ is therefore small. The small $\bar{\sigma}$ expression for ϵ resulting from the definition (58) is then

$$\epsilon = \epsilon_p. \tag{90}$$

This is correct by definition.

V. SUMMARY AND CONCLUSIONS

The purpose of this paper, as mentioned in Sec. I, has been to place the theory of extinction in mosaic crystals on sounder mathematical and physical ground; it has not been to provide yet another simple prescription to correct data for extinction. However, the relatively simple "AB-extinction formulas" of Sec. III, used in conjunction with the suggestion that a correction for weak primary extinction can be obtained from the exact solution of the dynamical diffraction problem in slabs, should prove useful. Anisotropic extinction effects resulting from crystal shape anisotropy can be taken into account using these formulas. Anisotropic extinction resulting from anisotropy in the mosaic structure can be included in the theory in a straightforward manner using the results of Coppens and Hamilton.[30] An attempt has been made to present the calculations in a manner which suggests various extensions and modifications. The formal solutions to the transport equations given in Sec. II should be applied to various simple polyhedral crystals. The solution to the dynamical diffraction problem for a sphere would be very important.

Zachariasen's extinction formulas have now been applied to the results of many experiments (see for example, Zachariasen,[31] Lawrence,[32] Lander and Brun,[33] Killean, Lawrence, and Sharma,[34] Cooper, Rouse and, Willis[35]), and various attempts to extend the calculations have been made (see for example, Cooper and Rouse[36]). It is not our purpose here to attempt to review the entire literature on the subject, or to try to determine whether the formulas are approximately

correct on the basis of a reasonably large amount of experimental data. However, it is fair to say that Zachariasen's extinction correction works well in the limit of small extinction (less than about 20%), that there are angular-dependent effects which lie outside the prescription, and that the correction works for large extinction in some cases, but not in others. In view of the numerous conceptual and mathematical inadequacies of the theory given in Zachariasen's[2,3] papers, why do the recommended corrections work at all?

It is clear that as long as the extinction is small, a simple linear correction is quite good (see Fig. 4), provided that the coefficient of the linear term is left as an adjustable parameter. Any theory, independent of its complexity, will give this result. In the determination of complex crystal structures there are many adjustable parameters, and any reasonable (for example, linear) form for the extinction coefficient which includes several more parameters will inevitably lead to a better fit. However, a detailed study of the wavelength dependence of the extinction will show the inadequacy of the formulas. It would be very interesting to have a fairly complete set of experimental data on a simple crystal, of known structure, for a wide selection of wavelengths. Neutron data would be preferable to x-ray data, since the scattering lengths are known and are not angular dependent.

APPENDIX

The scattering cross section for a small crystal in the Born approximation is

$$\frac{d\sigma}{d\Omega} = \left(\frac{m_n}{2\pi\hbar^2}\right)^2 \left| \int \exp(-i\mathbf{K}\cdot\mathbf{r})\, V(\mathbf{r}) d^3\mathbf{r} \right|^2, \tag{A1}$$

where $V(\mathbf{r})$ is the interaction potential of the neutron with the crystal, and $\mathbf{K}$ is the scattering vector

$$\mathbf{K} = \mathbf{k}' - \mathbf{k}. \tag{A2}$$

The derivation given here is for nuclear scattering; the generalization of the results to include magnetic scattering is straightforward. $V(\mathbf{r})$ is related to the scattering lengths b_j by,

$$V(\mathbf{r}) = (2\pi\hbar^2/m_n)\sum_j b_j \delta(\mathbf{r}\cdot\mathbf{r}_j). \tag{A3}$$

$V(\mathbf{r})$ can also be expressed in terms of its Fourier components, namely,

$$V(\mathbf{r}) = \sum_{\mathbf{G}} V_{\mathbf{G}} \exp(i\mathbf{G}\cdot\mathbf{r}), \tag{A4}$$

where the $\mathbf{G}$'s are the reciprocal lattice vectors. The Fourier coefficients $V_{\mathbf{G}}$ are related to the structure factors $F_{\mathbf{G}}$ by

$$V_{\mathbf{G}} = \frac{2\pi\hbar^2}{m_n} \frac{1}{V_{\text{cell}}} F_{\mathbf{G}}, \tag{A5}$$

where

$$F_{\mathbf{G}} = \sum_{j=1}^{N} b_j \exp(-i\mathbf{G}\cdot\mathbf{r}_j) \ (N \text{ atoms/unit cell}), \tag{A6}$$

$$V_{\mathbf{G}} = (1/\delta V) \int_{\text{crystal}} V(\mathbf{r}) \exp(-i\mathbf{G}\cdot\mathbf{r})\, d^3\mathbf{r}. \tag{A7}$$

δV is the volume of the small perfect crystal. We now assume that the crystal is spherical, with radius R.

Using Eqs. (A1), (A4), and (A5) it is easy to show that the scattering cross section is

$$\frac{d\sigma}{d\Omega} = (\delta V)^2 \sum_{\mathbf{G}} \frac{|F_{\mathbf{G}}|^2}{V_{\text{cell}}^2} \left(\frac{3\sin(q_{\mathbf{G}}R) - 3q_{\mathbf{G}}R\cos(q_{\mathbf{G}}R)}{(q_{\mathbf{G}}R)^3} \right), \tag{A8}$$

where

$$\mathbf{q}_{\mathbf{G}} = \mathbf{K} - \mathbf{G}. \tag{A9}$$

$d\sigma/d\Omega$ is highly peaked about the points $\mathbf{q}_{\mathbf{G}} = 0$. We will assume that the crystal is oriented close to one of these points, and concentrate on only one term in the sum, namely on one Bragg reflection.

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$$\frac{\partial J_i}{\partial x} = -\bar{\sigma}_t^{(1)} J_i + \bar{\sigma}^{(2)} J_d,$$

$$\frac{\partial J_d}{\partial s} = -\bar{\sigma}^{(2)} J_d + \bar{\sigma}^{(1)} j_i.$$

Note also that Eqs. (2a) and (2b) are valid even if $\bar{\sigma}$ depends upon position. However, it is unlikely that realistic problems could be solved if $\bar{\sigma}$ were not taken to be constant.
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