Acta Cryst. (1980). A**36**, 171–177

On Extinction.
IV. Integrated Intensities in Secondary-Extinction Theory

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(*Received* 13 *August* 1979; *accepted* 25 *September* 1979)

Abstract

Based on the new energy-transfer equations [Kato (1976). *Acta Cryst.* A**32**, 458–466; Kato (1979). *Acta Cryst.* A**35**, 9–16] the integrated intensities (II) are calculated. Since the energy-transfer equations have physical meanings different from the traditional ones originally given by Darwin [*Philos. Mag.* (1922), **43**, 800–824] and extended by Hamilton [*Acta Cryst.* (1957), **10**, 629–634], the method of calculating (II) must be modified, particularly in the case of a wide incident beam. Since the modified method does not include the angular integration, it is much simpler than the traditional method. Thus, the analytically rigorous expressions of (II) can be obtained for parallel-sided crystals including absorption, and for any diffraction conditions.

1. Introduction

In three previous papers [Kato, 1976*a*,*b*, 1979; (I), (II) and (III) hereafter], the present author developed a statistical dynamical theory of crystal diffraction. With this approach, it became possible to discuss primary and secondary extinction within a single theoretical framework. In fact, a system of energy-transfer equations (ET equations) between the direct (*O*) and Bragg-reflected (*G*) beams could be derived from the fundamental wave equations in distorted crystals above the critical condition $\Lambda \gtrsim \tau$, where Λ is the extinction distance and τ is the correlation length of the lattice phase factors at two different positions. In this paper, the integrated intensities of the *O* and *G* beams are calculated with this limited condition, *i.e.* for the case where secondary extinction is predominant.

The new ET equations are very similar to Hamilton's (1957) and Zachariasen's (1967*a*,*b*) equations. These are also systems of energy-transfer equations in two-dimensional reflection space, and are an extension of Darwin's (1922) classical system of equations. For this reason, in this paper the equations of this type are referred to as D–H–Z or H–Z equations. The new equations are henceforth denoted simply ET equations.

The similarity of ET and D–H–Z equations is merely of mathematical form and the physical meanings are rather different. This was pointed out at the end of paper (II). For the sake of completeness the differences are summarized in § 2. Because of this situation, the traditional method of calculating the integrated intensities must be modified, particularly in the case of a wide incident beam. Fortunately, the modified method is much simpler than the traditional. Thus, at least in the case of parallel-sided crystals, the exact expressions of the integrated intensities of the *O* and *G* beams can be obtained.

In the following, the integrated intensities are discussed separately for two cases in which the incident beam is either sufficiently narrow or wide in space. The calculated integrated intensities in these two cases are identical, as expected. This indicates that the present approach is theoretically satisfactory.

2. The solution for the incident beam of δ-function type

(*a*) *Fundamental equations*

The ET equations can be written in the following forms [equations (II.30*a*,*b*)], including the effect of normal absorption:*

$$\frac{\partial I_o}{\partial s_o} = -\mu_e I_o + \sigma_{-g} I_g, \qquad (1a)$$

$$\frac{\partial I_g}{\partial s_g} = -\mu_e I_g + \sigma_g I_o. \qquad (1b)$$

The coupling constants are given by:

$$\mu_e = \mu_o + 2\,\mathrm{Re}(\kappa_g \kappa_{-g})\tau, \qquad (2a)$$

$$\sigma_g = 2|\kappa_g|^2 \tau, \qquad (2b)$$

* In fact, I_o and I_g are intensities. The energy flow must have the magnitude of the Poynting vector $(c/4\pi)(KI_o, KI_g)$. Since the factor $c/4\pi$, c being the velocity of light, and the angular wave number K are common, they are omitted.

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where μ_o is the linear absorption coefficient, κ_g the reflection strength in amplitude per unit length [equation (I.2)] which is proportional to the structure factor F_g, and τ is the correlation length of the lattice phase factor defined by equation (I.3). For simplicity, the average notation in $\langle I \rangle$ and the suffix in τ_2 are omitted here.

Two points are worth mentioning.

Remark (1): I_o and I_g are not referred to a monodirectional beam specified by an angular parameter θ. If they are so defined, simple relations like equations (1) can no longer be expected. Unless the crystal is very nearly perfect, the beam reflected from a monodirectional beam must be angularly diffuse, so that the beam escapes from the angular channel prespecified by θ. The only way to avoid this complexity is to deal with the angularly integrated intensities from the beginning. As discussed in paper (II), equations (1) were, in fact, derived for the incident beam of δ-type, which is obviously a polydirectional wave.

Remark (2): The intensity I_o which satisfies equations (1) does not include the part of the O beam passing through a crystal without the Bragg reflection. This point was not explicitly mentioned in the previous papers. Nevertheless, the statement is obvious because the solution (II.26*a*), or equation (3*a*) in the following, has the form of a power series starting from the term of $|\kappa_g \kappa_{-g}|^2$. Obviously, this term is the sum of the doubly reflected beams.

(*b*) *The solution for the Laue cases*

In this section, in order to make the arguments concrete, we consider the Laue cases.

It is already known from paper (II) that the following expressions satisfy equations (1):

$$I_o(s_o, s_g) = I_e|\kappa_g \kappa_{-g}| \left(\frac{s_o}{s_g}\right)^{1/2} I_1[2\sigma(s_o s_g)^{1/2}] \times \exp[-\mu_e(s_o + s_g)], \quad (3a)$$

$$I_g(s_o, s_g) = I_e|\kappa_g|^2 I_0[2\sigma(s_o s_g)^{1/2}] \times \exp[-\mu_e(s_o + s_g)], \quad (3b)$$

where $I_e = A^2$,

$$\sigma = (\sigma_g \sigma_{-g})^{1/2} = 2\tau|\kappa_g \kappa_{-g}|, \quad (4)$$

and I_0 and I_1 are the modified Bessel functions of the zeroth and first order, respectively. Here, the normal absorption factor $\exp[-\mu_o(s_o + s_g)]$ is multiplied by the expressions (II.26*a*,*b*).

Historically, the following expressions were proposed by Werner, Arrott, King & Kendrick (1966) in the case of $\sigma_g = \sigma_{-g}$ as the solution of H–Z equations:*

$$J_o(s_o, s_g) = I_e\,\delta(s_g)\exp[-\mu_e(s_o + s_g)] + \tau I_o(s_o, s_g), \quad (5a)$$

$$J_g(s_o, s_g) = \tau I_g(s_o, s_g). \quad (5b)$$

These solutions are obtained with the boundary conditions

$$J_o = I_e\,\delta(s_g), \quad J_g = 0 \quad (6a,b)$$

on the entrance surface. The treatments are mathematically correct but are not adequate for the physical requirements of real experiments. As discussed in paper (I), the narrow incident wave of unit amplitude creates the Bragg reflected beam of amount $|\kappa_g|^2$, instead of $\tau|\kappa_g|^2$, along the line $s_g = 0$. Equations (1) hold only for the intensity fields created by $I_e|\kappa_g|^2$ (*Remark* 2). Thus, the present solution (3*b*) differs from solution (5*b*) by the factor τ. We shall discuss this point further in § 4.

(*c*) *Integrated intensity* [*supplement to* § 5 *of paper* (I)]

The intensity fields must be derived from the wave equation by taking an appropriate incident wave. Here, we shall consider the spherical wave

$$D_e = A\,\delta(s_g) \quad (7a)$$

$$= A \sin 2\theta_B\,\delta(x_o), \quad (7b)$$

where x_o is the coordinate perpendicular to the O beam. With these expressions, the total energy (intensity) of incidence is

$$E = \int |D_e|^2\,dx_o$$

$$= I_e(\sin 2\theta_B)^2 \int dx_o \int \exp[i(K_x - K'_x)x_o]\,dK_x\,dK'_x$$

$$= (I_e/2\pi)(\sin 2\theta_B)^2 \int dK_x. \quad (8a)$$

Using the relation $d\theta = dK_x/K$, we obtain the angular density of the incident energy as

$$\frac{\partial E}{\partial \theta} = (I_e/\lambda)(\sin 2\theta_B)^2. \quad (8b)$$

If one redefines the integrated intensity of the G beam per unit density of $\partial E/\partial\theta$,*

$$R^S_g(X'_o) = \lambda(\sin 2\theta_B)^{-2} I_e^{-1} \int I_g(X'_o, X_g)\,dX_g, \quad (9)$$

where I_g is the intensity of the G beam at an observation point X_g (perpendicular coordinate to the G beam) when a point source (7) is put at a position X'_o (perpendicular coordinate to the O beam).

For the incident beam which is a homogeneous distribution of the spherical wave source, the integrated intensity must be

$$R_g = \int R^S_g(X'_o)\,dX'_o, \quad (10)$$

* In this paper, the intensity fields of the D–H–Z equations are denoted J_o and J_g.

* In equations (9) and (10), $\int dY$ is assumed to be unity where Y is the perpendicular coordinate to the reflection plane.

where $\partial^2 E/\partial\theta\,\partial X'_o$ is assumed to be unity. The result is identical to that from the plane-wave theory [equation (I.28c); P^p_g]. In fact, the bundle of plane waves assumed for calculating P^p_g satisfies $\partial^2 E/\partial\theta\,\partial X'_o = 1$. The expression (10), therefore, can be universally used independently of the character of the incident beam.

(d) *The integrated intensity for a parallel-sided crystal*

In the Laue case, expression (9) can be written with the use of solution (3b) for I_g in the concrete form

$$R^S_g = \lambda(\sin 2\theta_B)^{-2}|\kappa_g|^2\gamma_g\int_a^b \exp[-\mu_e(s_o+s_g)] \times I_0[2\sigma(s_o s_g)^{1/2}]\,dx, \quad (11)$$

where the coordinates (a,b) and x are shown in Fig. 1, and (γ_o, γ_g) are the direction cosines of the directions of the O and G beams, respectively, with respect to the normal of the crystal.

From the geometrical relations

$$x_o = (x-a)\gamma_o = s_g \sin 2\theta_B, \quad (12a)$$

$$x_g = (b-x)\gamma_g = s_o \sin 2\theta_B, \quad (12b)$$

$$t = s_o\gamma_o + s_g\gamma_g, \quad (12c)$$

we have the width of the intensity field as

$$(b-a) = T\sin 2\theta_B/\gamma_o\gamma_g, \quad (13)$$

where T is the thickness of the crystal.

Introducing the normalized parameter

$$\zeta = [x - \tfrac{1}{2}(a+b)]/\tfrac{1}{2}(b-a) \quad (14)$$

instead of x, and the notations

$$M = \frac{1}{2}\left(\frac{1}{\gamma_o}+\frac{1}{\gamma_g}\right)\mu_e, \quad N = \frac{1}{2}\left(\frac{1}{\gamma_o}-\frac{1}{\gamma_g}\right)\mu_e \quad (15a,b)$$

$$L^2 = \sigma^2/\gamma_o\gamma_g, \quad (15c)$$

equation (11) can be written in the explicit form

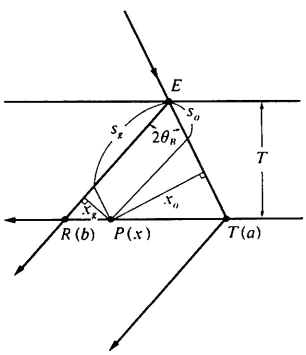

Fig. 1. Geometry of diffraction in the case of a narrow incident beam and a parallel-sided crystal. E: Entrance point. T and R: The region of the intensity field on the exit surface. P: The observation point. (a,b,x) indicate the coordinates of the points (T,R,P), respectively.

$$R^S_g = Q(T/\gamma_o)\exp(-MT)u_g(T), \quad (16a)$$

where

$$u_g(T) = \frac{1}{2}\int_{-1}^{1}\exp(NT\zeta)I_o[LT(1-\zeta^2)^{1/2}]\,d\zeta \quad (16b)$$

$$= \frac{\sinh[(N^2+L^2)^{1/2}T]}{(N^2+L^2)^{1/2}T}. \quad (16c)$$

Here, Q is the well known expression of the integrated intensity per unit volume and the unit intensity of the incident beam in the kinematical theory; *i.e.*

$$Q = (\lambda/\sin 2\theta_B)|\kappa_g|^2 = \left(\frac{e^2}{mc^2}\right)^2 \lambda^3|F_g C|^2/v^2\sin 2\theta_B. \quad (17)$$

Here, C is the polarization factor, v is the volume of the unit cell and the other symbols are the notations of standard usage.

When the crystal is sufficiently thin, expression (16a) tends to the kinematical expression of the integrated intensity

$$R^K_g = Q(T/\gamma_o), \quad (18)$$

so that the extinction coefficient is given by

$$\eta = R^S_g/R^K_g = \frac{\sinh[(N^2+L^2)^{1/2}T]}{(N^2+L^2)^{1/2}T}\exp(-MT). \quad (19)$$

Similarly, one can calculate the integrated intensity for the direct beam. The formulae corresponding to equations (11) and (16) are as follows:

$$R^S_o = \lambda(\sin 2\theta_B)^{-2}|\kappa_g\kappa_{-g}|\gamma_o\int_a^b\exp[-\mu_e(s_o+s_g)] \times \left(\frac{s_o}{s_g}\right)^{1/2} I_1[2\sigma(s_o s_g)^{1/2}]\,dx \quad (20a)$$

$$= Q_o[T/(\gamma_o\gamma_g)^{1/2}]\exp(-MT)u_o(T), \quad (20b)$$

where

$$Q_o = (\lambda/\sin 2\theta_B)|\kappa_g\kappa_{-g}| \quad (21)$$

and

$$u_o(T) = \frac{1}{2}\int_{-1}^{1}\exp(NT\zeta)\left(\frac{1-\zeta}{1+\zeta}\right)^{1/2} I_1[LT(1-\zeta^2)^{1/2}]\,d\zeta \quad (22a)$$

$$= \left(\frac{1}{LT}\right)\left\{\cosh[(N^2+L^2)^{1/2}T] - \frac{N}{(N^2+L^2)^{1/2}}\sinh[(N^2+L^2)^{1/2}T] - \exp(-NT)\right\}. \quad (22b)$$

The integration of equation (22a) is explained in the Appendix.

In the Bragg case, one can obtain the integrated intensity from similar considerations. For parallel-sided crystals, however, this is not a wise method. The problem will be postponed until § 3(*c*).

3. Integrated intensity for a wide incident beam

(a) Fundamental equations

In practice, we more often encounter the case of a wide incident beam. The penetrating beam without the Bragg reflection then overlaps spatially with the *O* beam in the crystal. The former has to be eliminated from the total *O* beam, because only the latter satisfies the differential equation (1) [*cf. Remark* 2 in § 2(*a*)].

Unlike the traditional treatment, therefore, the fundamental equation must be a system of the inhomogeneous equations:

$$\frac{\partial I_o}{\partial s_o} = -\mu_e I_o + \sigma_{-g} I_g, \qquad (23a)$$

$$\frac{\partial I_g}{\partial s_g} = -\mu_e I_g + \sigma_g I_o + QI_e \exp(-\mu_e \bar{s}_o), \qquad (23b)$$

where $\bar{s}_o$ is the distance between an observation point and the corresponding entrance point. The third term is responsible for the body source of the intensity fields. The expression can be justified by the following argument.

Taking an infinitesimal parallelepiped (Fig. 2), the energy of the Bragg reflected beam created by the incident beam is given by

$$\delta E_g = QI_e \exp[-(\mu_e \bar{s}_o)](\delta s_o\, \delta s_g \sin 2\theta_B), \qquad (24)$$

where ( ) is the infinitesimal volume concerned. The beam travels in the *G* direction with a width of $\delta s_0 \sin 2\theta_B$. Therefore, the increment of *G* beam intensity per unit distance of s_g is

$$\frac{\partial I_g}{\partial s_g} = QI_e \exp[-(\mu_e \bar{s}_o)].$$

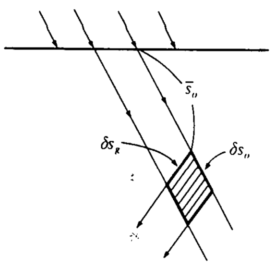

Fig. 2. The explanation of the additional term of equation (23*b*) representing the body source.

(b) The solution for a parallel-sided crystal; the Laue cases

To avoid unnecessary complexity in the mathematics, we shall mainly treat non-absorbing cases in this section. From the consideration of symmetry, I_o and I_g must be independent of *x*. Equations (23) then reduce to

$$\gamma_o \frac{\partial I_o}{\partial t} = -\sigma I_o + \sigma I_g, \qquad (25a)$$

$$\gamma_g \frac{\partial I_g}{\partial t} = -\sigma I_g + \sigma I_o + QI_e \exp[-\sigma(t/\gamma_o)], \qquad (25b)$$

where σ is defined by equation (4), in which $\kappa_g = \kappa^*_{-g}$, and *t* is the coordinate normal to the crystal surface.

Equation (25) will be solved with the boundary conditions

$$I_o(0) = I_g(0) = 0. \qquad (26a,b)$$

These conditions fit the real experimental conditions in the Laue case ($\gamma_o, \gamma_g > 0$). For the Bragg case ($\gamma_o > 0$, $\gamma_g < 0$), the solution is merely a mathematical tool for finding the real solution. Meanwhile, we shall not bother with the sign of γ_g.

The following treatments are standard in the sense of solving the differential equations. Nevertheless, the mathematical logic will be explained in brief. The detail can be found, for example, in Sneddon (1972).

Taking the Laplace transform of equations (25), and remembering the conditions (26), we obtain the relations between $I_o(p)$ and $I_g(p)$, the Laplace transforms of $I_o(t)$ and $I_g(t)$, as follows:

$$(\gamma_o p + \sigma) I_o(p) = \sigma I_g(p), \qquad (27a)$$

$$(\gamma_g p + \sigma) I_g(p) = \sigma I_o(p) + QI_e \gamma_o/(\gamma_o p + \sigma). \qquad (27b)$$

These can be solved immediately as

$$I_o(p) = (QI_e \sigma/\gamma_o \gamma_g)/(p-a)(p-b)(p-c), \qquad (28a)$$

$$I_g(p) = (QI_e/\gamma_g)/(p-a)(p-b), \qquad (28b)$$

where *a* and *b* are the solutions of the secular equation of equations (27). These and *c* are given in the explicit forms,

$$\left.\begin{matrix} a \\ b \end{matrix}\right\} = -M_0 \pm (N_0^2 + L_0^2)^{1/2} = \left\{\begin{matrix} 0 & (29a) \\ -2M_0 & (29b) \end{matrix}\right.$$

$$c = -\sigma/\gamma_o = -(M_0 + N_0). \qquad (30)$$

Here, M_0, N_0 and L_0 are the special case of *M*, *N* and *L* defined by equations (15), respectively, for non-absorbing cases ($\mu_o = 0$, $\kappa_g = \kappa^*_{-g}$).

The solutions $I_o(t)$ and $I_g(t)$ are given by the inverse Laplace transforms of $I_o(p)$ and $I_g(p)$, namely

$$I_o(t)=\frac{1}{2\pi i}\int_{\gamma-i\infty}^{\gamma+i\infty}(QI_e\,\sigma/\gamma_o\,\gamma_g)\times\frac{e^{pt}}{(p-a)(p-b)(p-c)}\,\mathrm{d}p, \qquad (31a)$$

$$I_g(t)=\frac{1}{2\pi i}\int_{\gamma-i\infty}^{\gamma+i\infty}(QI_e/\gamma_g)\frac{e^{pt}}{(p-a)(p-b)}\,\mathrm{d}p, \qquad (31b)$$

where γ is the radius of convergence; $\gamma>0$ in the present problem. Integrations of this kind can be performed by the standard method of contour integral. The results are

$$I_o(t)=(QI_e\,\sigma/\gamma_o\,\gamma_g)\left[\frac{e^{at}}{(a-b)(a-c)}+\frac{e^{bt}}{(b-a)(b-c)}+\frac{e^{ct}}{(c-a)(c-b)}\right], \qquad (32a)$$

$$I_g(t)=(QI_e/\gamma_g)\left[\frac{e^{at}}{a-b}+\frac{e^{bt}}{b-a}\right]. \qquad (32b)$$

Using equations (29) and (30), we have

$$I_o(t)=(QI_e/\sigma)e^{-M_0t}\left[\cosh M_0t-\frac{N_0}{M_0}\sinh M_0t-e^{-N_0t}\right], \qquad (33a)$$

$$I_g(t)=(QI_e/\gamma_g)e^{-M_0t}\left(\frac{1}{M_0}\right)\sinh M_0t. \qquad (33b)$$

The solution is physical for the Laue cases. In the case of wide beams, the cross sections of the O and G beams are changed by the factor γ_g/γ_o. Therefore, the integrated intensity for the crystal of thickness T is

$$R_{g,L}=I_e^{-1}(\gamma_g/\gamma_o)I_g(T) \qquad (34a)$$

$$=(QT/\gamma_o)e^{-M_0T}\left[\frac{\sinh M_0T}{M_0T}\right]. \qquad (34b)$$

This result is identical to equation (16) since M and $(N^2+L^2)^{1/2}$ reduce to M_0 in non-absorbing cases.

It is worth mentioning here the conservation of energy. From the fundamental equations (25) and the boundary conditions (26), the relation

$$\gamma_o I_o+\gamma_g I_g=QI_e(\gamma_o/\sigma)\{1-\exp[-(\sigma/\gamma_o)t]\} \qquad (35)$$

must be satisfied everywhere in the crystal. The solutions (33) actually satisfy the relation (35).

(c) *The Bragg case* ($\gamma_o>0$, $\gamma_g<0$)

In this case, the solutions (33) are hypothetical. In fact, $I_g(T)$ is negative! However, the correct solution with the boundary conditions

$$I_o(0)=0,\quad I_g(T)=0 \qquad (36a,b)$$

can be constructed by adding a suitable solution which satisfies the homogeneous part of equations (25).

For this purpose, we shall calculate the solutions of the homogeneous equations with the boundary conditions

$$I_o^H(0)=0,\quad I_g^H(T)=1. \qquad (37a,b)$$

The superscript H indicates the homogeneous equation. If one obtains the solutions

$$I_o^H(T)=\mathbf{R},\quad I_g^H(0)=\mathbf{T}, \qquad (38a,b)$$

one can write the correct solutions of the inhomogeneous equation for the Bragg cases,

$$I_o^B(T)=I_o(T)-\mathbf{R}.I_g(T), \qquad (39a)$$

$$I_g^B(0)=-\mathbf{T}.I_g(T), \qquad (39b)$$

where $I_o(T)$ and $I_g(T)$ are the solutions [equations (33)] at $t=T$. In fact, $\mathbf{T}$ has the physical meaning of transmissivity of the G beam and $\mathbf{R}$ is reflectivity from the G to the O beam.

The general solutions of the homogeneous equations have the forms

$$I_o^H(t)=A\exp(at)+B\exp(bt), \qquad (40a)$$

$$I_g^H(t)=C_aA\exp(at)+C_bB\exp(bt), \qquad (40b)$$

where C_a and C_b are the intensity ratios (I_g^H/I_o^H) for the characteristic solutions $\exp(at)$ and $\exp(bt)$, respectively. They are determined by the secular equations as

$$C_a=(\gamma_o a+\sigma)/\sigma=1, \qquad (41a)$$

$$C_b=(\gamma_o b+\sigma)/\sigma=(N_0-M_0)/(N_0+M_0). \qquad (41b)$$

Using these relations and the boundary conditions (37a,b), we finally have

$$\mathbf{T}=M_0[M_0\cosh M_0T+N_0\sinh M_0T]^{-1}\exp(M_0T), \qquad (42a)$$

$$\mathbf{R}=(N_0+M_0)\sinh M_0T[M_0\cosh M_0T+N_0\sinh M_0T]^{-1}. \qquad (42b)$$

Inserting these and the solutions (33) at $t=T$ into equations (39), we have

$$I_o^B(T)=(QI_e/\sigma)\exp(-M_0T)\times\frac{M_0}{N_0\sinh M_0T+M_0\cosh M_0T}-e^{-N_0T}, \qquad (43a)$$

$$I_g^B(0)=(QI_e/|\gamma_g|)\sinh M_0T[N_0\sinh M_0T+M_0\cosh M_0T]^{-1}. \qquad (43b)$$

The integrated intensity is given by

$$R_{g,B} = (QT/\gamma_o)\,\frac{\sinh M_0 T}{T}\,[N_0 \sinh M_0 T + M_0 \cosh M_0 T]^{-1}. \quad (44)$$

When the thickness T is sufficiently large,

$$R_{g,B}(T \to \infty) = Q/\sigma. \quad (45)$$

On the other hand, if the crystal is thin enough, $R_{g,B}$ $(T \to 0)$ tends to the kinematical result, as it should.

For absorbing crystals, we can calculate the integrated intensities along a similar line of considerations. We then have to start with the fundamental equations (23). In the Laue cases, we obtain the same results as equations (16), (20) and (22). In the Bragg cases, here, the final results of the integrated intensities are presented:

$$R_{o,B} = (Q_o I_e)[T/(\gamma_o\gamma_g)^{1/2}]\exp(-MT) \times (1/LT)[(N^2+L^2)^{1/2} \times \{N\sinh[(N^2+L^2)^{1/2}T] + (N^2+L^2)^{1/2} \times \cosh[(N^2+L^2)^{1/2}T] - \exp(-NT)\}^{-1}], \quad (46a)$$

$$R_{g,B} = (QI_e)(T/\gamma_o)\sinh[(N^2+L^2)^{1/2}T](1/T) \times \{N\sinh[(N^2+L^2)^{1/2}T] + (N^2+L^2)^{1/2}\cosh[(N^2+L^2)^{1/2}T]\}^{-1}. \quad (46b)$$

It is worth noticing that the intensities obtained by the conventional approach are τ times these expressions in the respective cases. This point will be discussed in the following section.

4. Discussion

In the present scheme of secondary-extinction theory, ET equations were derived from the wave equations. Through this task, the applicability and the meanings of ET equations were elucidated. The following points are particularly significant for the use of ET equations.

(i) Applicable range: $\Lambda \gtrsim \tau$. This has been fully discussed in papers (II) and (III).

(ii) *Remarks* (1) and (2) of § 2. Before discussing a few problems related to these remarks, it is worth pointing out that the intensity fields I_o and I_g [equations (3)] are first obtained from the wave equation and then ET equations are justified by noticing that I_o and I_g satisfy them. For this reason, the expressions of I_o and I_g are primary and ET equations are auxiliary. This is not a hens-and-eggs argument.

(a) *The direct beam and the kinematical G beam*

The essence of *Remark* 2 is that ET equations are nothing to do with the direct beam (not wave) which penetrates through the crystal without creating the Bragg reflected beam. This point can be seen in equations (II.8 and 25). The direct beam is undefined there. If, however, we define it formally by taking the term $r = 0$ (non-reflected beam), it has a singular form $(1/a\tau)|A|^2\exp(-\mu_e s_0)$ $[a \to 0]$. By using the reverse of the rule to convert the differential equation to the difference equations adopted throughout papers (I) and (II) [equations (I.11 and 12)], the expression can be interpreted as $(1/\tau)|A|^2\delta(s_g)\exp(-\mu_e s_o)$. Thus, the correct solution is $(1/\tau)$ times the conventional solution assuming $|A|^2\delta(s_g)$ for the incident beam [Werner *et al.*'s (1966) solution; equation (5)]. If we normalize $|A|^2$ on the scale of $\partial E/\partial\theta = 1$, the effective direct beam has the form $(\sin 2\theta_B)^{-2}(\lambda/\tau)\delta(s_g)$. Thus, one can see that only (λ/τ) of the total incident energy contributes to the Bragg reflection. This is very reasonable because (λ/τ) is the angular range of diffraction for a crystal of coherent size τ in the order of magnitude.

Unlike the direct beam, the integrated intensity of kinematical reflection must be independent of the crystal perfection except for the effective absorption factor, which includes dynamical effects. It is, in fact, $|A|^2|\kappa_g|^2\exp(-\mu_e s_o)$ [the term with $r = 1$ in equations (II.8)]. This point will be discussed further in the next section.

(b) *Comparison of the traditional integrated intensity and the present one*

Traditionally, the integrated intensity is given by

$$R_g = \frac{\gamma_g}{\gamma_o}\int J_g(T, \varepsilon)\,d\varepsilon, \quad (47)$$

where ε is the angle of deviation of the incident direction from the maximum reflection [see equation (50) below], and J_g is the solution of D–H–Z equations with the boundary conditions

$$J_o(0) = I_e, \quad J_g(0) = 0 \quad (48a,b)$$

in wide-beam cases. Here, for clarity, we shall discuss this problem referring to the Laue case of a non-absorbing parallel-sided crystal, and the crystal is assumed to be type I according to Zachariasen's (1967) definition. Other cases can be discussed using similar considerations. What then happens is that the intensity J_g is given as follows,

$$J_g(T, \varepsilon) = [\bar{\sigma}(T/\gamma_o) + a_1(T)\bar{\sigma}^2 + \cdots], \quad (49)$$

where $\bar{\sigma}$ is the coupling constant of the D–H–Z equations and is given by [*cf.* equations (I.4 and 5)]

$$\bar{\sigma} = QW(\varepsilon). \quad (50)$$

In these equations $a_1(T)$ is a coefficient depending on the diffraction conditions, and Q is proportional to $|\kappa_g|^2$ [equation (17)] and $W(\varepsilon)$ is the angular spectrum of the mosaic blocks. Inserting (49) into (47), one obtains

$$R_g(T) = Q(T/\gamma_o) + a_1(T)Q^2 \int [W(\varepsilon)]^2 \,\mathrm{d}\varepsilon + \ldots. \quad (51)$$

Here, one of the complexities in the traditional theory of secondary extinction arises. Even in the case of parallel-sided crystals, in which $J_g(T, \varepsilon)$ can be solved exactly, we have to know the integrations of multiple powers of the distribution $W(\varepsilon)$. This gives rise to an ambiguity in $R_g(T)$.

In the present theory, the integrated intensity is $(\gamma_g/\gamma_o)I_g$ because ET equations involve the relations of the total intensities of O and G beams [*Remark* 1]. If we write I_g in the form of a power series of the new coupling constant σ, we have

$$R_g(T) = Q[(T/\gamma_o) + b_1(T)\sigma + \ldots]. \quad (52)$$

As discussed above [§ 4(*a*)], the correlation length τ does not appear in the first term. Unnecessary complexity of the angular integration is now swept out.

In § 2, the intensity distributions of the O and G beams were given for sufficiently narrow beams. The integrated intensities were given by a spatial integration over the exit surface. In § 3, the intensity fields excited in the crystal for a sufficiently wide incident beam were calculated for both Laue and Bragg cases. This calculation immediately gives the integrated intensity. In Laue cases, it was confirmed that the results obtained by the two methods were identical. This shows not only a mathematical beauty but also that the present approach to secondary extinction is theoretically firm.

One final point should be mentioned. In all the treatments of this paper, we have started with the fundamental equations (1) for simplicity. As discussed in paper (III), the theory will be improved by multiplying suitable reduction factors R_o and R_g by the correlation length $\tau(=\tau_2)$. This situation also holds for the integrated intensity.

APPENDIX
The explicit form of $u_o(t)$ and $u_g(t)$

The functions are defined by equations (16*b*) and (22*a*), respectively. First, we consider them in the forms of power series:

$$u_g(t) = \tfrac{1}{2} \int_{-1}^{1} \exp(Nt\zeta) \sum_{n=0}^{\infty} \frac{(\frac{1}{2}Lt)^{2n}(1-\zeta^2)^n}{n!\,n!} \,\mathrm{d}\zeta, \quad (A.1a)$$

$$u_o(t) = \tfrac{1}{2} \int_{-1}^{1} \exp(Nt\zeta)(1-\zeta) \times \sum_{n=0}^{\infty} \frac{(\frac{1}{2}Lt)^{2n+1}(1-\zeta^2)^n}{n!(n+1)!} \,\mathrm{d}\zeta. \quad (A.1b)$$

For convenience, we shall define the similar function

$$v = \tfrac{1}{2} \int_{-1}^{1} \exp(Nt\zeta) \sum_{n=0}^{\infty} \frac{(\frac{1}{2}Lt)^{2n+1}(1-\zeta^2)^n}{n!(n+1)!} \,\mathrm{d}\zeta. \quad (A.2)$$

Letting $y = Nt$ and $\rho = (\frac{1}{2}Lt)^2$, from these expressions we have the relations

$$\frac{\partial}{\partial \rho}[\rho^{1/2} v] = u_g \quad (A.3)$$

and

$$u_o = v - \frac{\mathrm{d}v}{\mathrm{d}y}. \quad (A.4)$$

For integrating (*A*.3), the boundary condition

$$v(\rho = 0) = 0 \quad (A.5)$$

is employed. Thus, one can obtain u_o from u_g simply by the integral and differential operations.

The functional form $u_g(t)$ is well known, *i.e.*

$$u_g(t) = \sinh[(N^2 + L^2)^{1/2}t]/[(N^2 + L^2)^{1/2}t]. \quad (A.6)$$

From this expression it turns out that

$$u_o(t) = (1/Lt)\left\{\cosh[(N^2 + L^2)^{1/2}t] - \left[\frac{N}{(N^2 + L^2)^{1/2}}\right] \times \sinh[(N^2 + L^2)^{1/2}t] - \exp(-Nt)\right\}. \quad (A.7)$$

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