PAYNE, W. B. & LEVINGER, J. S. (1956). *Phys. Rev.* **101**, 1020–1026.
PRATT, R. H., KISSEL, L. & BERGSTROM, P. M. JR (1994). *X-ray Anomalous (Resonance) Scattering: Theory and Experiment*, edited by K. FISCHER, G. MATERLIK & C. SPARKS. Amsterdam: Elsevier/North Holland.
RAMASESHAN, F., RAMESH, T. G. & RANGANATH, G. S. (1975). *Anomalous Scattering*, edited by F. RAMASESHAN & S. C. ABRAHAMS, pp. 139–161. Copenhagen: Munksgaard.
REICHE, F. & THOMAS, W. (1925). *Z. Phys.* **34**, 510–525.
ROY, S. C. (1991). *Phys. Rev. A*, **43**, 6393–6394.
ROY, S. C., KISSEL, L. & PRATT, R. H. (1983). *Phys. Rev. A*, **27**, 285–290.
ROY, S. C., PRATT, R. H. & KISSEL, L. (1993). *Rad. Phys. Chem.* **41**, 725–738.
ROY, S. C., SARKAR, B., KISSEL, L. & PRATT, R. H. (1987). *Nucl. Instrum. Methods A*, **255**, 66–67.
ROY, S. C., SARKAR, B., PRATT, R. H. & KISSEL, L. (1986). *Phys. Rev. A*, **34**, 1178–1187.
RULLHUSEN, P., MÜCKENHEIM, W., SMEND, F., SCHUMACHER, M., BERG, G. P. A., MORK, K. & KISSEL, L. (1981). *Phys. Rev. C*, **23**, 1375–1383.
SAKURAI, J. J. (1967). *Advanced Quantum Mechanics.* Reading: Addison-Wesley.
SALOMAN, E. B., HUBBELL, J. H. & SCOFIELD, J. H. (1988). *At. Data Nucl. Data Tables*, **38**, 1–197.
SCHAUPP, D., SCHUMACHER, M., SMEND, F. & RULLHUSEN, P. (1983). *J. Phys. Chem. Ref. Data*, **12**, 467–512.
SCHUMACHER, M. & STOFFREGEN, A. (1977). *Z. Phys. A*, **283**, 15–19.
SCOFIELD, J. H. (1975). *Atomic Inner-Shell Processes*, edited by B. CRASEMANN, pp. 265–292. New York: Academic Press.
SMEND, F. & CZERWINSKI, H. (1986). *Z. Phys. D*, **1**, 139–140.
SMEND, F., SCHAUPP, D., CZERWINSKI, H., SCHUMACHER, M., MILLHOUSE, A. H. & KISSEL, L. (1987). *Phys. Rev. A*, **36**, 5189–5199.
SMITH, D. Y. (1987). *Phys. Rev. A*, **35**, 3381–3387.
SOMMERFELD, A. (1939). *Atombau and Spektrallinien*, Vol. 2. Braunschweig: Vieweg.
STANGLMEIER, F., LENGELER, B., WEBER, W., GÖBEL, H. & SCHUSTER, M. (1992). *Acta Cryst.* A**48**, 626–639.
THOMAS, W. (1925). *Naturwissesnschaften*, **13**, 627.
WANG, M. S. & PRATT, R. H. (1983). *Phys Rev. A*, **28**, 3115–3116.
ZHOU, B. (1991). PhD thesis, Univ. of Pittsburgh, USA.
ZHOU, B., KISSEL, L. & PRATT, R. H. (1992*a*). *Phys Rev. A*, **45**, 2983–2988.
ZHOU, B., KISSEL, L. & PRATT, R. H. (1992*b*). *Phys. Rev. A*, **45**, 6906–6909.
ZHOU, B., KISSEL, L. & PRATT, R. H. (1992*c*). *Nucl. Instrum Methods B*, **66**, 307–312.
ZHOU, B., PRATT, R. H., ROY, S. C. & KISSEL, L. (1990). *Phys. Scr.* **41**, 495–498.

Acta Cryst. (1995). A**51**, 288–294

Reformulation of the Statistical Theory of Dynamical Diffraction in the Case $E = 0$

BY J. P. GUIGAY

Laboratoire Louis Néel, BP 166, 38042 *Grenoble CEDEX* 9, *France*

AND F. N. CHUKHOVSKII

Institute of Crystallography, Russian Academy of Science, Moscow 117333, *Russia*

(*Received* 8 *March* 1994; *accepted* 21 *September* 1994)

Abstract

The statistical theory of dynamical diffraction, in the case $E = 0$, for which there is no long-range order, is reformulated with rigorous boundary conditions. The presence of a coherent forward-diffracted wave along the boundary of the influence region in the spherical-wave approach is taken into account. The integrated reflectivity is calculated in the case of Laue geometry and is found to be significantly different from the result of the previous formulations if $\chi^2\tau T > 1$ (χ is the reciprocal of the extinction distance, τ is the correlation length of the lattice imperfection and T is the crystal thickness along the incident direction).

1. Introduction

Following Kato (1980), the Bragg diffracted intensity from a randomly distorted crystal contains a coherent part and an incoherent part, which are related to the statistical averages and to the statistical fluctuations of the wave amplitudes, respectively. Further developments and some modifications of the original treatment of Kato (1980) have been proposed by the various authors quoted in the reference list of the present paper, and its applicability to some experimental data has been discussed recently by Schneider, Bouchard, Graf & Nagasawa (1992), Takama (1993) and Takama & Harida (1994).

A more general form of the statistical theory, based on wave equations more rigorous than the Takagi–Taupin equations used below, has also been proposed by Kato (1991), but this new advanced development is not considered in the present paper. The theory based on the Takagi–Taupin equations can itself be reformulated in a more complete form, especially by reconsidering the boundary conditions along the edges of the Borrmann fan. This was first done for the coherent waves (Guigay & Chukhovskii, 1992; see also Kato, 1994). The purpose of the present paper is to continue this reformulation for

ISSN 0108-7673

the incoherent intensities in the particular case for which the static Debye–Waller factor $E=0$. This is to be considered as an intermediate step towards the calculation of the incoherent intensities in the general case for which E has any value between 0 and 1.

In the case where $E=0$, for which there is no long-range order, the correlation length τ of the crystal imperfection can be considered as corresponding to the size of the perfect blocks of the mosaic model in the conventional extinction theory; there would therefore be no primary extinction if τ is supposed to be much smaller than the extinction distance Λ. Consequently, in agreement with the terminology of Becker & Al-Haddad (1990), the $E=0$ case of the statistical theory with $\tau \ll \Lambda$, which is the actual case considered in the present paper, may be considered as the case of pure secondary extinction.

2. Definition of the coherent waves and the incoherent intensities

The dynamical wave propagation in a distorted crystal, for the condition of a single Bragg diffraction, is very conveniently described by the Takagi–Taupin equations

$$\partial G_0/\partial s_0 = i\varphi^*(s_0, s_h)\, G_h(s_0, s_h) \quad (1a)$$

$$\partial G_h/\partial s_h = i\varphi(s_0, s_h)\, G_0(s_0, s_h), \quad (1b)$$

where $G_0(s_0, s_h)$ and $G_h(s_0, s_h)$ are the amplitudes of the wave components along the incident and the Bragg directions; s_0 and s_h are oblique coordinates along these directions. We consider the case of a non-absorbing centrosymmetric crystal. χ is then a real quantity equal to $1/\Lambda$, Λ being the 'extinction distance', which is defined here as $\Lambda = V_c/\lambda F$, V_c being the volume of the unit cell, F the scattering length of this unit cell and λ the radiation wavelength. We shall use $Q = \lambda\chi^2/\sin 2\theta_B$, θ_B being the Bragg angle. $\varphi(s_0, s_h)$ is the 'lattice phase factor' related to the displacement field $\mathbf{u}(s_0, s_h)$ of the distorted crystal lattice and to the diffraction vector $\mathbf{h}$:

$$\varphi(s_0, s_h) = \exp[-i\mathbf{h}\cdot\mathbf{u}(s_0, s_h)].$$

Let us consider an incident beam limited by an infinitely narrow slit located at the origin point O on the entrance surface of the crystal (Fig. 1). In this so-called spherical-wave case, the diffraction process produces a spreading of the total wave in the influence region $s_0, s_h > 0$ (the Borrmann fan). Equations (1a) and (1b) can then be written in integral form as

$$G_0(s_0, s_h) = \delta(s_h) + G_{0d}(s_0, s_h) = \delta(s_h) + i\chi \int_0^{s_0} \mathrm{d}\zeta\, \varphi^*(\zeta, s_h)\, G_h(\zeta, s_h) \quad (2a)$$

$$G_h(s_0, s_h) = i\chi \int_0^{s_h} \mathrm{d}\eta\, \varphi(s_0, \eta)\, G_0(s_0, \eta). \quad (2b)$$

The δ function $\delta(s_h)$ represents the incident wave and $G_{od}(s_0, s_h)$ the wave diffracted into the incident direction $0s_0$. It is convenient to write

$$G_{0d}(s_0, s_h) = i\chi \int_0^{s_0} \mathrm{d}\zeta\, \varphi^*(\zeta, s_h)\, G_h(\zeta, s_h) \quad (3a)$$

$$G_h(s_0, s_h) = i\chi\, \theta(s_h)\, \theta(s_0)\, \varphi(s_0, 0) + i\chi \int_0^{s_h} \mathrm{d}\eta\, \varphi(s_0, \eta)\, G_{0d}(s_0, \eta). \quad (3b)$$

$\theta(s)$ is the step function (equal to 0 for $s < 0$ and equal to 1 for $s > 0$). $\theta(s_h)$ and $\theta(s_0)$ are introduced in the first term of (3b) for the reason that the successive terms of the multiple scattering expansion starting from $G_h^{(1)} = i\chi\theta(s_h)\theta(s_0)\varphi(s_0, 0)$ and obtained by iteration in formulae (3),

$$\begin{aligned} G_h &= G_h^{(1)} + G_h^{(3)} + G_h^{(5)} + \ldots \\ G_{0d} &= G_{0d}^{(2)} + G_{0d}^{(4)} + G_{0d}^{(6)} + \ldots, \end{aligned} \quad (4)$$

are then equal to zero outside the influence region $s_0, s_h > 0$. $G_h^{(1)}$ is the kinematical approximation of the Bragg diffracted wave. On the boundaries of the influence region, we get

$$\begin{aligned} &\text{along } 0s_0: \\ &\quad G_h(s_0, 0) = i\chi\varphi(s_0, 0), \\ &\quad G_{0d}(s_0, 0) = -\chi^2 s_0, \\ &\text{along } 0s_h: \\ &\quad G_{0d}(0, s_h) = 0, \\ &\quad G_h(0, s_h) = i\chi\varphi(0, 0). \end{aligned} \quad (5)$$

Let us also consider the intensity distributions $I_0(s_0, s_h) = G_{0d}^* G_{0d}$ and $I_h(s_0, s_h) = G_h^* G_h$ of the diffracted beams. They are such that

$$\begin{aligned} \partial I_0/\partial s_0 &= -i\chi\varphi\, G_h^* G_{0d} + \text{c.c.} \\ \partial I_h/\partial s_h &= i\chi\varphi\, G_h^* G_{0d} + \text{c.c.} + \chi^2\delta(s_h), \end{aligned} \quad (6)$$

the symbol c.c representing the complex conjugate of the preceding term. We have used the fact that $i\chi\varphi G_{0d}$ is indeed equal to $\partial G_h/\partial s_h$ for s_h not equal to 0; the $\chi^2\delta(s_h)$ term describes the discontinuity of $I_h(s_0, s_h)$ at $s_h = 0$.

In the statistical theory, $\varphi(s_0, s_h)$ is a random function characterized by a static Debye–Waller factor E and a correlation function $g(u)$:

$$\begin{aligned} \langle\varphi(s_0, s_h)\rangle &= E \quad (0 \le E \le 1) \\ \langle\varphi^*(s_0 + u, s_h)\varphi(s_0, s_h)\rangle &= \langle\varphi^*(s_0, s_h + u)\,\varphi(s_0, s_h)\rangle \\ &= E^2 + (1 - E^2)\, g(u). \end{aligned} \quad (7)$$

$E = 1$ corresponds to the case of a perfect crystal. $g(u)$ is such that $g(0) = 1$ and $g(\infty) = 0$; correlation lengths τ

and τ_2 are also defined as

$$\tau = \int_0^\infty \mathrm{d}u\ g(u), \quad \tau_2 = \int_0^\infty \mathrm{d}u\ g^2(u). \tag{8}$$

We shall consider the same form of $g(u)$ already used in our previous paper (Guigay & Chukhovskii, 1992),

$$g(u) = \exp(-u/\tau). \tag{9}$$

This is a very convenient model in many analytical calculations; we shall nevertheless write $g(u)$ when this exponential model, which has also been recommended by Becker & Al-Haddad (1989) from a general discussion based on the theory of stochastic processes, is not necessarily assumed.

The coherent diffracted waves are the averaged functions $\langle G_h(s_0, s_h)\rangle$ and $\langle G_{0d}(s_0, s_h)\rangle$. According to Kato (1980), the intensities of the coherent waves represent only a part of the total diffracted intensities which are defined as $I_0 = \langle G^*_{0d}G_{0d}\rangle$ and $I_h = \langle G^*_hG_h\rangle$, the remaining parts being incoherent intensities:

$$\begin{aligned} I^i_0(s_0, s_h) &= \langle G^*_{0d}G_{0d}\rangle - \langle G^*_{0d}\rangle\langle G_{0d}\rangle = \langle G^*_{0d}G_{0d}\rangle - I^c_0 \\ I^i_h(s_0, s_h) &= \langle G^*_hG_h\rangle - \langle G^*_h\rangle\langle G_h\rangle = \langle G^*_hG_h\rangle - I^c_h. \end{aligned} \tag{10}$$

The boundary conditions for the coherent waves and for the incoherent intensities are easily obtained from the general conditions (5):

$$\begin{aligned} &\langle G_h(s_0, 0)\rangle = \langle G_h(0, s_h)\rangle = i\chi E; \\ &\langle G_{0d}(s_0, 0)\rangle = -\chi^2 s_0, \quad \langle G_{0d}(0, s_h)\rangle = 0 \end{aligned} \tag{11}$$

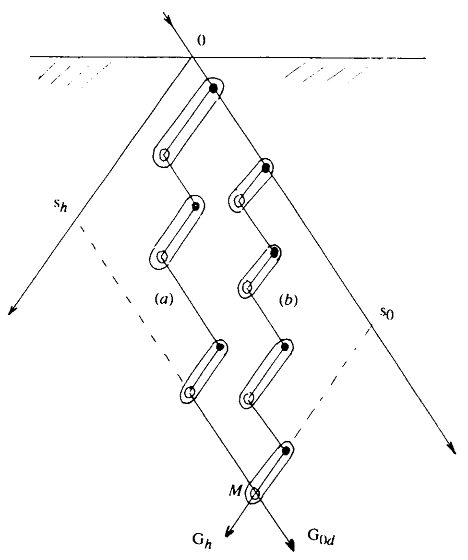

Fig. 1. 'Optical paths' considered as successive pairs of scattering points. Path (*a*) contributes to $\langle G_{0d}\rangle$ given in formula (16) and path (*b*) contributes to $\langle i\chi\varphi^* G_h\rangle$ given in formula (19). An exponential factor $g(\varepsilon_i) = \exp(-\varepsilon_i/\tau)$ is associated with each pair, ε_i being the separation of the points of this pair; this gives a final factor $\exp[-(\varepsilon_1 + \varepsilon_2 + \ldots)/\tau] = \exp(-s_h/\tau)$ independent of the path from the origin point 0 to the observation point $M(s_0, s_h)$.

$$\begin{aligned} &I^i_h(s_0, 0) = I^i_h(0, s_h) = (1 - E^2)\chi^2; \\ &I^i_0(s_0, 0) = I^i_0(0, s_h) = 0. \end{aligned} \tag{12}$$

The coherent waves satisfy integro-differential equations (Polyakov, Chukhovskii & Piskunov, 1991; Guigay & Chukhovskii, 1992; Kato, 1994):

$$\begin{aligned} \partial\langle G_{0d}\rangle/\partial s_0 &= \partial\langle G_0\rangle/\partial s_0 \\ &= i\chi E\langle G_h(s_0, s_h)\rangle - (1 - E^2)\chi^2 \\ &\quad \times \int_0^{s_h} \mathrm{d}\eta\ g(s_h - \eta)\langle G_0(s_0, \eta)\rangle \end{aligned} \tag{13a}$$

$$\begin{aligned} \partial\langle G_h\rangle/\partial s_h &= i\chi E\langle G_0(s_0, s_h)\rangle - (1 - E^2)\chi^2 \\ &\quad \times \int_0^{s_0} \mathrm{d}\zeta\ g(s_0 - \zeta)\langle G_h(\zeta, s_h)\rangle, \end{aligned} \tag{13b}$$

where it must be taken into account that $\langle G_0(s_0, s_h)\rangle = \delta(s_h) + \langle G_{0d}(s_0, s_h)\rangle$.

According to Kato (1980), the incoherent intensities are supposed to satisfy transfer equations

$$\partial I^i_0/\partial s_0 = 2\chi^2\tau_e(I^i_h - I^i_0) + 2(1 - E^2)\chi^2\tau I^c_h \tag{14a}$$

$$\partial I^i_h/\partial s_h = 2\chi^2\tau_e(I^i_0 - I^i_h) + 2(1 - E^2)\chi^2\tau I^c_0. \tag{14b}$$

Here, τ_e is a new correlation length equal to $(1 - E^2)\tau_2$, according to Al-Haddad & Becker (1988). These equations, in which the coherent intensities are sources of the incoherent intensities, are valid for s_0, $s_h > \tau$, but not for $s_h < \tau$ or $s_0 < \tau$. More rigorous equations valid in the whole influence region, in the special case $E = 0$, have been proposed recently by Chukhovskii & Guigay (1993) and will be more completely discussed in the present paper.

3. The optical theorem in the dynamical theory

In the theory of scattering by a single scattering centre, the total outgoing wave is the coherent sum of the incident plane wave plus the diffracted wave which interfere in the forward direction, giving rise to a negative interference term, which compensates exactly the total diffracted intensity (the intensity of the diffracted wave integrated over the scattering angles).

This so-called 'optical theorem' is also valid in the dynamical theory of Bragg diffraction by a non-absorbing crystal, independently of the crystal deformation. The interference term of the forward-diffracted wave $G_{0d}(s_0, s_h)$ with the incident wave $\delta(s_h)$ is equal to $G_{0d}(s_0, 0) = -\chi^2 s_0$ (Guigay & Chukhovskii, 1992); after leaving the crystal, the total intensity diffracted in the Bragg and in the forward directions is thus equal to $\chi^2 T$, T being the value of s_0 at the exit surface for $s_h = 0$ (T is therefore the path length of the incident beam in the crystal); this corresponds to the kinematical value of the integrated intensity; in other words, the value of the integrated intensity of the Bragg beam only in the well known kinematical approximation is rigorously the sum of the integrated intensities of the Bragg and forward-diffracted beams. This remarkable result is in agreement with the differential relation

$$\partial I_0/\partial s_0 + \partial I_h/\partial s_h = \chi^2 \delta(s_h) \quad (15)$$

obtained from equations (6) for the intensity distributions.

In the statistical theory, we shall calculate the coherent and incoherent parts of the integrated intensities of the Bragg and forward-diffracted beams and we shall therefore pay attention that their total sum is equal to the simple kinematical integrated intensity.

4. The forward coherent diffracted wave in the case $E = 0$

In this case, $\langle G_h(s_0, s_h)\rangle = 0$, but there is, nevertheless, a forward coherent diffracted wave $\langle G_{0d}(s_0, s_h)\rangle$ in the boundary region $s_h < \tau$ (Guigay & Chukhovskii, 1992). We shall prove analytically that, for $E = 0$,

$$\langle G_{0d}(s_0, s_h)\rangle = -\chi(s_0/s_h)^{1/2} J_1[2\chi(s_0 s_h)^{1/2}] \exp(-s_h/\tau). \quad (16)$$

The $E = 0$ case of equations (13a) and (13b), together with the relevant boundary conditions, is

$$\partial\langle G_0\rangle/\partial s_0 = \chi^2 \int_0^{s_h} d\eta\, g(s_h - \eta)\langle G_0(s_0, \eta)\rangle$$

$$\langle G_0(0, s_h)\rangle = \delta(s_h); \quad (17a)$$

$$\partial\langle G_h\rangle/\partial s_h = \chi^2 \int_0^{s_0} d\xi\, g(s_0 - \xi)\langle G_h(\xi, s_h)\rangle$$

$$\langle G_h(s_0, 0)\rangle = 0. \quad (17b)$$

Let us use the Laplace transform of equation (17a) with respect to s_h and the Laplace transform of equation (17b) with respect to s_0. We define:

$$\langle G_0(s_0, p)\rangle = \int_0^\infty ds_h \exp(-p s_h)\langle G_0(s_0, s_h)\rangle$$

$$\langle G_h(p, s_h)\rangle = \int_0^\infty ds_0 \exp(-p s_0)\langle G_h(s_0, s_h)\rangle$$

$$g(p) = \int_0^\infty ds \exp(-ps)\, g(s)$$

(s stands for s_0 or s_h). Equations (17a) and (17b) are transformed into differential equations:

$$\partial\langle G_0(s_0, p)\rangle/\partial s_0 = \chi^2 g(p)\langle G_0(s_0, p)\rangle \quad \langle G_0(0, p)\rangle = 1; \quad (18a)$$

$$\partial\langle G_h(p, s_h)\rangle/\partial s_h = \chi^2 g(p)\langle G_h(p, s_h)\rangle \quad \langle G_h(p, 0)\rangle = 0. \quad (18b)$$

The corresponding solutions are

$$\langle G_0(s_0, p)\rangle = \exp[-\chi^2 s_0 g(p)] \quad \text{and} \quad \langle G_h(p, s_h)\rangle = 0.$$

It is thus verified that $G_h(s_0, s_h) = 0$; $G_0(s_0, s_h)$ can be calculated in the case $g(s) = \exp(-s/\tau)$. We have then

$$\langle G_0(s_0, p)\rangle = \exp[-\chi^2 s_0 (p + 1/\tau)^{-1}]$$

which can be shown to be the Laplace transform with respect to s_h of

$$\langle G_0(s_0, s_h)\rangle = \delta(s_h) - \chi(s_0/s_h)^{1/2} J_1[2\chi(s_0 s_h)^{1/2}] \times \exp(-s_h/\tau).$$

Relation (16) is thus obtained since $\langle G_0(s_0, s_h)\rangle = \delta(s_h) + \langle G_{0d}(s_0, s_h)\rangle$. This result is also obtained more easily by the geometrical approach illustrated and explained in Fig. 1; simultaneously, we also obtain the useful relation

$$\langle \varphi^* G_h\rangle = i\chi J_0[2\chi(s_0 s_h)^{1/2}] \exp(-s_h/\tau). \quad (19)$$

In (16) and (19), $J_0[2\chi(s_0 s_h)^{1/2}]$ and $J_1[2\chi(s_0 s_h)^{1/2}]$ are Bessel functions; $\langle G_{0d}\rangle$ and $\langle \varphi^* G_h\rangle$ are therefore equal to the perfect-crystal wave functions multiplied by $\exp(-s_h/\tau)$. The coherent forward-diffracted intensity is

$$I_0^c(s_0, s_h) = \chi^2 (s_0/s_h) J_1^2[2\chi(s_0 s_h)^{1/2}] \exp(-2s_h/\tau). \quad (20)$$

It is important for the following discussions to point out that:

$$\begin{aligned}\partial I_0^c/\partial s_0 &= 2\langle G_{0d}\rangle\langle i\chi\varphi^* G_h\rangle \\ &= 2\chi^3 \exp(-2s_h/\tau)\,(s_0/s_h)^{1/2} J_1[2\chi(s_0 s_h)^{1/2}] \\ &\quad \times J_0[2\chi(s_0 s_h)^{1/2}]. \end{aligned} \quad (21)$$

5. More rigorous transfer equations for the incoherent intensity distributions in the case $E = 0$

Using the integral expressions (2a) and (2b) for G_{0d} and G_h, we consider

$$\varphi G_h^* G_{0d} = \chi^2 \varphi(s_0, s_h) \int_0^{s_h} d\eta \int_0^{s_0} d\xi\, \varphi^*(s_0, \eta) \times \varphi^*(\xi, s_h) G_0^*(s_0, \eta) G_h(\xi, s_h). \quad (22)$$

In the statistical theory for $E = 0$, we propose the following approximation:

$$\begin{aligned}&\langle \varphi(s_0, s_h)\varphi^*(s_0, \eta)\varphi^*(\xi, s_h)\, G_{0d}^*(s_0, \eta)\, G_h(\xi, s_h)\rangle \\ &= \langle \varphi(s_0, s_h)\varphi^*(s_0, \eta)\rangle \\ &\quad \times \langle \varphi^*(\xi, s_h)\, G_0^*(s_0, \eta)\, G_h(\xi, s_h)\rangle \\ &\quad + [1 - g^2(s_h)]\langle \varphi(s_0, s_h)\varphi^*(\xi, s_h)\rangle \\ &\quad \times \langle \varphi^*(s_0, \eta)\, G_0^*(s_0, \eta)\, G_h(\xi, s_h)\rangle \\ &= g(s_h - \eta)\langle \varphi^*(\xi, s_h)\, G_0^*(s_0, \eta)\, G_h(\xi, s_h)\rangle \\ &\quad + [1 - g^2(s_h)]\, g(s_0 - \xi) \\ &\quad \times \langle \varphi^*(s_0, \eta)\, G_{0d}^*(s_0, \eta)\, G_h(\xi, s_h)\rangle,\end{aligned}$$

in which $\varphi(s_0, s_h)$ is coupled to $\varphi^*(s_0, \eta)$ or to $\varphi^*(\xi, s_h)$. The factor $[1 - g^2(s_h)]$ is introduced in order to make this approximation valid for $s_h < \tau$ and for $s_h \gg \tau$. Again, using the integral expressions (2*a*) and (2*b*), we get

$$\begin{aligned} i\chi\langle\varphi G_h^* G_{0d}\rangle &= \chi^2 \int_0^{s_h} \mathrm{d}\eta\, g(s_h - \eta) \\ &\quad\times \langle G_0^*(s_0, \eta)\, G_{0d}(s_0, s_h)\rangle \\ &\quad- \chi^2[1 - g^2(s_h)] \int_0^{s_0} \mathrm{d}\xi\, g(s_0 - \xi) \\ &\quad\times \langle G_h(\xi, s_h)\, G_h^*(s_0, s_h)\rangle. \end{aligned} \tag{23}$$

We now propose the new approximations

$$\langle G_h(\xi, s_h)\, G_h^*(s_0, s_h)\rangle + \text{c.c.} = 2g(s_0 - \xi) I_h^i(\xi, s_h)$$
$$\begin{aligned}\langle G_0^*(s_0, \eta)\, G_{0d}(s_0, s_h)\rangle + \text{c.c.} &= 2\langle G_0(s_0, \eta)\rangle\langle G_{0d}(s_0, s_h)\rangle \\ &\quad+ 2g(s_h - \eta) I_0^i(s_0, \eta),\end{aligned}$$

where we use the fact that $\langle G_0\rangle$ and $\langle G_{0d}\rangle$ are real functions. These approximations are consistent with the limiting cases such that $(s_0 - \zeta)$ and $(s_h - \eta)$ are either much larger or smaller than τ; it is also easily verified that the first one is true in the kinematical approximation,

$$\begin{aligned}\langle G_h(\xi, s_h)\, G_h^*(s_0, s_h)\rangle &= \chi^2\langle\varphi(\xi, s_h)\varphi^*(s_0, \eta)\rangle \\ &= \chi^2 g(s_0 - \xi)\end{aligned}$$

and

$$I_h^i = \chi^2.$$

We thus get

$$\begin{aligned} &i\chi\langle G_h^* G_{od}\rangle + \text{c.c.} \\ &= 2\chi^2 \int_0^{s_h} \mathrm{d}\eta\, g^2(s_h - \eta) I_0^i(s_0, \eta) - 2\chi^2[1 - g^2(s_h)] \\ &\times \int_0^{s_0} \mathrm{d}\xi\, g^2(s_0 - \xi) I_h^i(\xi, s_h) + 2\chi^2\langle G_{0d}\rangle \\ &\times \int_0^{s_h} \mathrm{d}\eta\, g(s_h - \eta)\langle G_0(s_0, \eta)\rangle. \end{aligned}$$

We know from (17*a*) that the last integral is equal to $\partial\langle G_0\rangle/\partial s_0 = \partial\langle G_{0d}\rangle/\partial s_0$. Therefore, $I_0^i(s_0, s_h)$ and $I_h^i(s_0, s_h)$ satisfy the following integro-differential equations (Chukhovskii & Guigay, 1993):

$$\begin{aligned}\partial I_0^i/\partial s_0 &= 2\chi^2[1 - g^2(s_h)] \int_0^{s_0} \mathrm{d}\xi g^2(s_0 - \xi) I_h^i(\xi, s_h) \\ &\quad- 2\chi^2 \int_0^{s_h} \mathrm{d}\eta\, g^2(s_h - \eta) I_0^i(s_0, \eta)\end{aligned} \tag{24a}$$

$$\partial I_h^i/\partial s_h = -\partial I_0^i/\delta s_0 + \chi^2\delta(s_h) - \partial I_0^c/\partial s_0. \tag{24b}$$

For $s_h < \tau$, we can use $\partial I_h^i/\partial s_h = \chi^2\delta(s_h) - \partial I_0^c/\partial s_0$. Assuming that $g(s) = \exp(-s/\tau)$, we can obtain (using integration by parts) the following expression for $I_h^i(s_0, s_h)$:

$$\begin{aligned} I_h^i(s_0, s_h) &= \chi^2 - 2\chi^3 \int_0^{s_h} \mathrm{d}\eta \exp(-2\eta/\tau)(s_0/\eta)^{1/2} \\ &\quad\times J_1[2\chi(s_0\eta)^{1/2}] J_0[2\chi(s_0\eta)^{1/2}] \\ &= \chi^2 J_0^2[2\chi(s_0 s_h)^{1/2}] \exp(-2s_h/\tau) + 2(\chi^2/\tau) \\ &\quad\times \int_0^{s_h} \mathrm{d}\eta \exp(-2\eta/\tau) J_0^2[2\chi(s_0\eta)^{1/2}]. \end{aligned} \tag{25}$$

It is interesting to note that we recover the formula for a perfect crystal in the limit $\tau \to \infty$.

If it is assumed that the variations of the functions $I_h^i(\xi, s_h)$ and $I_0^i(s_0, \eta)$ over an interval of width τ for the variables ξ and η can be neglected, the convolution integrals in (24*a*) and (24*b*) can be approximated by $\tau_2 I_h^i(s_0, s_h)$ and $\tau_2 I_0^i(s_0, s_h)$. We thus obtain simpler transfer equations, with $\sigma = 2\chi^2\tau_2$,

$$\partial I_0^i/\partial s_0 = +\sigma[1 - g^2(s_h)] I_h^i(s_0, s_h) - \sigma I_0^i(s_0, s_h) \tag{26a}$$

$$\begin{aligned}\partial I_h^i/\partial s_h &= -\sigma[1 - g^2(s_h)] I_h^i(s_0, s_h) + \sigma I_0^i(s_0, s_h) \\ &\quad+ \chi^2\delta(s_h) - \partial I_0^c/\partial s_0.\end{aligned} \tag{26b}$$

For $s_h \gg \tau$, $g(s_h)$ and $\partial I_0^c/\partial s_0 \to 0$ and $\delta(s_h)$ can be omitted; equations (26*a*) and (26*b*) are then reduced to the $E = 0$ case of (14*a*) and (14*b*) with $\tau_e = \tau_2$.

6. Transfer equations for integrated intensities

Let us consider the case of symmetric Laue diffraction by a crystal of uniform thickness $t \gg \tau$. On the exit surface of the crystal, $s_0 + s_h = T = t/\cos\theta_B$ is a constant equal to the crystal thickness along the incident direction (Fig. 2). The integrated intensity of the Bragg diffracted beam

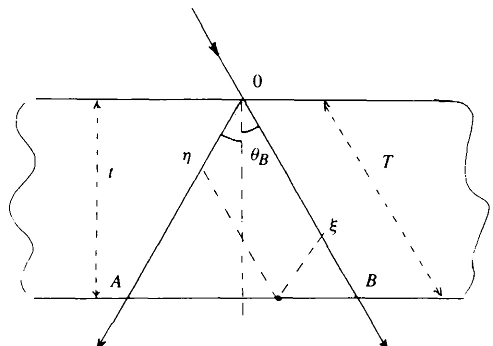

Fig. 2. Geometrical arrangement in the symmetrical Laue case. Because $\xi + \eta = T$ on the exit surface, the integrations along the basis *AB* of the influence region [see formulae (27) and (28)] can be carried out by using ξ or η as the integration variable.

can be written as (ξ and η are used here instead of s_0 and s_h)

$$R_h(T) = Q\chi^{-2}\int_0^T d\xi\, I_h(\xi, T-\xi), \qquad (27)$$

in which $Q\chi^{-2}$ is introduced as a normalization factor (Q has been defined in §2), in order to obtain the well known kinematical result $R_h(T) = QT$ when $I_h(\xi, T-\xi)$ is a constant function equal to χ^2. Similarly, the integrated intensity of the forward-diffracted beam is

$$R_0(T) = Q\chi^{-2}\int_0^T d\eta\, I_0(T-\eta, \eta) \qquad (28)$$

(R_0 and I_0 here stand for R_0^i and I_0^i or for R_0^c and I_0^c); it is indeed convenient, for our calculations below, to use ξ as the integration variable in (27) and η as the integration variable in (28). Following a method used in a previous work on integrated intensities (Guigay, 1989), the integration of (27) or (28) will also be performed on all the other terms of (26*a*) and (26*b*) in order to transform these equations into transfer equations for the integrated intensities:

(*a*) For the integration of $\partial I_0^i/\partial s_0$ and $\partial I_0^c/\partial s_0$, using the fact that $I_0^i(T-\eta, \eta)$ and $I_0^c(T-\eta, \eta)$ are equal to 0 for $\eta = T$, we get from (28) (with the same convention as for R_0 and I_0)

$$dR_0(T)/dT = Q\chi^{-2}\int_0^T d\eta\; \partial I_0(T-\eta, \eta)/\partial T. \qquad (29)$$

(*b*) For the integration of $\partial I_h/\partial s_h$, we must take into account that $\chi^2\delta(s_h)$ is included in our definition of $\partial I_h/\partial s_h$; consequently,

$$dR_h(T)/dT = Q\chi^{-2}\int_0^T d\xi\; \partial I_h(\xi, T-\xi)/\partial T, \qquad (30)$$

without an additional term $I_h(T, 0)$ equal to the value of $I_h(\xi, T-\xi)$ for $\xi = T$, which would be present if $\chi^2\delta(s_h)$ had not been included in $\partial I_h/\partial s_h$; in this case, $\chi^2\delta(s_h)$ would not be present in the left-hand side of (24*b*) and (26*b*); the final results of the calculations are the same if $\chi^2\delta(s_h)$ is included or not included in $\partial I_h/\partial s_h$.

(*c*) The integration of $g^2(s_h)I_h^i(s_o, s_h)$ will be neglected because it leads to a quantity smaller than $Q\tau$, which is itself negligible with respect to R_h^i.

(*d*)

$$Q\chi^{-2}\int_0^T d\eta\; \chi^2\delta(\eta) = Q.$$

We finally obtain transfer equations for the integrated intensities:

$$dR_0^i/dT = \sigma R_h^i(T) - \sigma R_0^i(T) \qquad (31a)$$

$$dR_h^i/dT = \sigma R_0^i(T) - \sigma R_h^i(T) - dR_0^c/dT + Q. \qquad (31b)$$

According to (21) and (29), the derivative of R_0^c is, in the case $g(\eta) = \exp(-\eta/\tau)$,

$$dR_0^c/dT = Q2\chi\int_0^T d\eta \exp(-2\eta/\tau)[(T-\eta)/\eta]^{1/2} \times J_1\{2\chi[(T-\eta)\eta]^{1/2}\}J_0\{2\chi[(T-\eta)\eta]^{1/2}\}.$$

The following approximation is possible because of the exponential term $\exp(-2\eta/\tau)$ and because we suppose $T \gg \tau$; replacing $(T-\eta)$ in this integral by T and setting the upper limit of integration to ∞, we obtain a Laplace-transform integral, which can be calculated exactly:

$$\begin{aligned} dR_0^c/dT &= Q2\chi\int_0^\infty d\eta \exp(-2\eta/\tau)(T/\eta)^{1/2} \\ &\quad \times J_1[2\chi(T\eta)^{1/2}]J_0[2\chi(T\eta)^{1/2}] \\ &= Q - Q\exp(-\sigma T)\mathbf{I}_0(\sigma T), \end{aligned} \qquad (32)$$

where $\sigma = \chi^2\tau$ is the same as in (26*a*) and (26*b*), since here $\tau = 2\tau_2$, $\mathbf{I}_0$ is the modified Bessel function of order 0, such that $\mathbf{I}_0(x) = J_0(ix)$. Equations (31*a*) and (31*b*) can then be written as

$$dR_0^i/dT = \sigma R_h^i(T) - \sigma R_0^i(T) \qquad (33a)$$

$$dR_h^i/dT = \sigma R_o^i(T) - \sigma R_h^i(T) + Q\exp(-\sigma T)\mathbf{I}_0(\sigma T), \qquad (33b)$$

resulting in

$$\begin{aligned} R_h^i + R_0^i &= Q\int_0^T du \exp(-\sigma u)\mathbf{I}_0(\sigma u) \\ &= QT\exp(-\sigma T)[\mathbf{I}_0(\sigma T) + \mathbf{I}_1(\sigma T)] \\ R_h^i - R_0^i &= Q\int_0^T du \exp[-2\sigma(T-u)]\exp(-\sigma u)\mathbf{I}_0(\sigma u) \\ &= QT\exp(-\sigma T)[\mathbf{I}_0(\sigma T) - \mathbf{I}_1(\sigma T)]. \end{aligned}$$

$\mathbf{I}_1$ is the modified Bessel function of order 1 and we have used some classical properties of the modified Bessel functions. The final result is very simple:

$$R_0^i = QT\exp(-\sigma T)\mathbf{I}_1(\sigma T) \qquad (34a)$$

$$R_h^i = QT\exp(-\sigma T)\mathbf{I}_0(\sigma T). \qquad (34b)$$

7. Comparison with the previous formulation for $E = 0$ and concluding remarks

In the previous form of the statistical theory for $E = 0$ by Becker & Al-Haddad (1990), the incoherent intensities $I_h^i(s_0, s_h)$ and $I_0^i(s_0, s_h)$ are obtained as the solutions of the transfer equations

$$\partial I_0^i/\partial s_0 = -\partial I_h^i/\partial s_h = \sigma(I_h^i - I_0^i) \qquad (35)$$

with the 'effective' boundary conditions

$$I_h^i(s_0, 0) = \chi^2 \exp(-\sigma s_0) \quad \text{and} \quad I_0^i(0, s_h) = 0. \tag{36}$$

These equations are obtained by supposing that $I_h^c = I_0^c = 0$ and $E = 0$ in the more general equations (14*a*) and (14*b*). As pointed out by Kato (1980) and by Becker & Al-Haddad (1990), the boundary condition $I_h^i(s_0, 0) = \chi^2$ cannot be used together with equations (35) because these equations are not valid for $s_h = 0$. The effective conditions (36) are based on the idea that the incident-beam intensity is attenuated in the crystal because of intensity transfer from the incident beam to the diffracted beams. In such an intuitive approach, the interference effect described as the optical theorem in §3 is absent. We can apply the method of the preceding section to (35) and (36) in order to get the following transfer equations for the integrated intensities:

$$\mathrm{d}R_0^i/\mathrm{d}T = \sigma R_h^i(T) - \sigma R_0^i(T) \quad \text{(previous formulation)}$$

$$\mathrm{d}R_h^i/\mathrm{d}T = \sigma R_0^i(T) - \sigma R_h^i(T) + Q\exp(-\sigma T),$$

from which we easily get

$$R_0^i = (Q/2\sigma)[1 - \exp(-\sigma T)]^2 \tag{37a}$$

(previous formulation)

$$R_h^i = (Q/2\sigma)[1 - \exp(-2\sigma T)]. \tag{37b}$$

It is interesting, from the point of view of possible applications, to compare the expressions (34*b*) and (37*b*) for the integrated reflectivity $R_h^i(T)$; they are similar for $\sigma T \ll 1$:

$$R_h^i = QT(1 - \sigma T + \tfrac{3}{4}(\sigma T)^2 + \ldots) \quad \text{(new formulation)}$$

$$R_h^i = QT(1 - \sigma T + \tfrac{2}{3}(\sigma T)^2 + \ldots) \quad \text{(previous formulation)} \tag{38}$$

but a significant difference appears for values of σT around unity. For large values of σT such that $\sigma T \gg 1$, using the well known asymptotic form of the modified Bessel functions, it is found that R_h^i and R_0^i increase as $(T/\tau)^{1/2}$ in our new formulation, instead of reaching a finite limit in the previous one:

$$R_h^i(T), R_0^i(T) \to QT/(2\pi\sigma T)^{1/2} = Q\chi^{-1}(T/2\pi\tau)^{1/2} \quad \text{(new formulation)} \tag{39}$$

$$R_h^i(T), R_0^i(T) \to QT/2\sigma = Q/(2\chi^2\tau) \quad \text{(previous formulation).}$$

The fact that R_h^i and R_0^i have the same limiting behaviour in the new formulation (and in the previous one also) is due to the analytical form of equations (31*a*) and (31*b*): because the 'source' term $(Q - \mathrm{d}R_0^c/\mathrm{d}T)$ tends to zero as σT tends to infinity, $R_h^i(T)$ and $R_0^i(T)$ are then simply equivalent to $[QT - R_0^c(T)]/2$ for $\sigma T \gg 1$.

The fact that R_h^i and R_0^i have a finite limit in the previous formulation is clearly due to the effective boundary conditions (36); because of the factor $\exp(-\sigma s_0)$, intensity transfer from the incident beam to the diffracted beams is effectively limited to a finite depth in the crystal. This is not the case in our new formulation, in which a coherent forward-diffracted wave is present; this is a fundamental difference with respect to the previous formulation because the intensity conservation requirement is then based on the interference effect of the optical theorem. Nevertheless, it is shown in (38) that the previous formulation represents a useful approximation to our new, more complete, formulation if the crystal is not too thick.

The calculation of the incoherent intensities in the general case, E having any value between 0 and 1, will be considered in a future paper on the same basis as in the present paper for $E = 0$. Special attention will be paid to the conditions imposed by the optical theorem which is of general validity. We expect an analytically new behaviour for the integrated reflectivity $R_h^i(T)$, similar to the results obtained in the special case $E = 0$.

Errata to a preceding paper [Guigay & Chukhovskii (1992). *Acta Cryst.* A**48**, 819–826].

In order to avoid confusion, we point out the following corrections to be introduced in the preceding paper concerning the coherent waves:

(1) The term $+i\chi E\delta(s_h)$, corresponding to the discontinuity of $G_h(s_0, s_h)$ at $s_h = 0$, must be added to the right-hand side of the second formula (10), in order to have the same definition of $\partial G_h/\partial s_h$ in equations (10) and (11).

(2) The same term $i\chi E\delta(s_h)$ must be deleted from the first formula (11); this was an error independent of correction (1).

These corrections were taken into account effectively later in this preceding paper; they had therefore no consequences on the calculations.

References

AL-HADDAD, M. & BECKER, P. (1988). *Acta Cryst.* A**44**, 262–270.
BECKER, P. & AL-HADDAD, M. (1989). *Acta Cryst.* A**45**, 333–337.
BECKER, P. & AL-HADDAD, M. (1990). *Acta Cryst.* A**46**, 123–129.
CHUKHOVSKII, F. N. & GUIGAY, J. P. (1993). *J. Phys. D*, **26**, A53–56.
GUIGAY, J. P. (1989). *Acta Cryst.* A**45**, 241–244.
GUIGAY, J. P. & CHUKHOVSKII, F. N. (1992). *Acta Cryst.* A**48**, 819–826.
KATO, N. (1980). *Acta Cryst.* A**36**, 763–769, 770–778.
KATO, N. (1991). *Acta Cryst.* A**47**, 1–11.
KATO, N. (1994). *Acta Cryst.* A**50**, 17–22.
POLYAKOV, A. M., CHUKHOVSKII, F. N. & PISKUNOV, D. I. (1991). *Sov. Phys. JETP*, **72**(2), 330–340.
SCHNEIDER, J. R., BOUCHARD, R., GRAF, H. A. & NAGASAWA, H. (1992). *Acta Cryst.* A**48**, 804–819.
TAKAMA, T. (1993). *Acta Cryst.* A**49**, C384.
TAKAMA, T. & HARIMA, H. (1994). *Acta Cryst.* A**50**, 239–246.