Acta Cryst. (1992). A**48**, 121–134

Diffraction by a Randomly Distorted Crystal. II. General Theory

BY PIERRE BECKER* AND MOSTAFA AL HADDAD

Laboratoire de Cristallographie, Associé à l'Université J. Fourier, CNRS, 166X, 38042 Grenoble CEDEX, France

(*Received* 28 *November* 1988; *accepted* 31 *July* 1991)

Abstract

A method is presented to deal with the propagation of X-rays or neutrons in a statistically distorted crystal in the general case where both short-range and long-range order are present: it is the generalization of a theory presented in a previous paper [Becker & Al Haddad (1990). *Acta Cryst.* A**46**, 123–129]. The main difference from Kato's formulation is concerned with the correlation length Γ of the incoherent part of the beams [Kato (1980). *Acta Cryst.* A**36**, 763–769, 770–778; Al Haddad & Becker (1988). *Acta Cryst.* A**44**, 262–270; Becker & Al Haddad (1990)]. The present formulation shows that Γ is variable within the sample under study, and is of the same order of magnitude as the correlation length τ of the phase factor [$\exp(2\pi i\mathbf{h}\cdot\mathbf{u})$ where $\mathbf{u}$ is the distortion field]. This is the main difference from Kato's approach where Γ was considered as $\gg\tau$. A detailed solution of the propagation equations is proposed and is applied to the case of silicon crystals containing a variable amount of oxygen, using measurements by Schneider, Gonçalves, Rollason, Bonse, Lauer & Zulehner [*Nucl. Instrum. Methods Phys. Res.* (1988), B**29**, 661–674] using γ-ray diffraction. The present theory is in fair agreement with the observed intensities, although Kato's original proposition does not work.

I. Introduction

The present authors discussed the statistical basis of dynamical diffraction by a randomly distorted crystal in a previous paper (Becker & Al Haddad, 1989; see also Guigay, 1989). Statistical theory was first introduced by Kato (1976, 1980) who proposed propagation equations for the intensities of the incident and diffracted beams derived from Takagi's equations. The object of the present paper is to improve Kato's approach by using a self-consistent theory.

Crystals are distorted by various causes, such as interstitial atoms *etc.* The distortion field is denoted as $\{\mathbf{u}(\mathbf{r})\}$ indicating that an atom at $\mathbf{r}$ is displaced over a distance $\mathbf{u}$. Let φ be the phase factor

$$\varphi=\exp(2\pi i\mathbf{h}\mathbf{u}). \tag{1}$$

If

$$\langle\varphi\rangle=E \tag{2}$$

the fluctuation $\delta\varphi$ of the phase is defined through the equation

$$\varphi=\langle\varphi\rangle+\delta\varphi. \tag{3}$$

E is the long-range order parameter (static Debye–Waller factor). Phase correlation is introduced by

$$\begin{aligned}\langle\varphi^*(\mathbf{r}+\mathbf{t})\varphi(\mathbf{r})\rangle&=E^2+\langle\delta\varphi^*(\mathbf{r}+\mathbf{t})\delta\varphi(\mathbf{r})\rangle\\&=E^2+(1-E^2)g(\mathbf{t}),\end{aligned} \tag{4}$$

$g(\mathbf{t})$ being the pair-correlation function of the phase factor.

The justification for using ensemble averages was fully discussed in the paper by Becker & Al Haddad (1990), hereafter referred to as (I).

Two correlation lengths are needed:

$$\begin{aligned}\tau&=\int_0^\infty g(t)\,\mathrm{d}t\\ \tau_2&=\int_0^\infty g^2(t)\,\mathrm{d}t.\end{aligned} \tag{5}$$

τ represents the distance over which two optical routes lose their mutual phase coherence. τ_2 has a similar meaning and plays a significant role in the theory.

As discussed in detail by Becker & Al Haddad (1989), if one assumes that the correlation length is smaller than the average distance between centres of scattering (of the order of Λ), one obtains an exponential law for the correlation function:

$$g(t)=\exp(-x/\tau)$$

then, for x, $y>0$

$$\begin{aligned}g(x,y)&\simeq g(x)g(y)\\ g(x+y)&\simeq g(x)g(y).\end{aligned} \tag{6}$$

Takagi's equations are written in the form

$$\begin{aligned}\partial D_0/\partial s_0&=i\chi\varphi D_h\\ \partial D_h/\partial s_h&=i\chi\varphi^* D_0.\end{aligned} \tag{7}$$

* Permanent address: Laboratoire de Minéralogie et Cristallographie, Université Pierre & Marie Curie, Tour 16, 4 Place Jussieu, 75252 Paris Cedex 05, France.

0108-7673/92/020121-14$03.00

D_0 and D_h are the amplitudes for the incident and diffracted beams, propagating in the directions s_0 and s_h respectively (Fig. 1), $\chi = 1/\Lambda$ and Λ is the extinction length:

$$1/\Lambda = (\lambda a C/V)F$$

where λ is the wavelength, $a = 10^{-12}$ cm for neutrons, $a = 0.28 \times 10^{-12}$ cm for X-rays, C is the polarization factor, V is the volume of the unit cell and F is the structure factor.

It will be assumed that the crystal is centrosymmetric and non-absorbing and that its dimension $l \gg \tau$, Λ.

The case of a spherical incident wave (a point source, emitting in direction s_0) of unit intensity will be considered in this paper:

$$D_0^0 = \delta(s_h). \qquad (8)$$

Equations (7) can be transformed into

$$\begin{aligned} D_0(s_0, s_h) &= \delta(s_h) + i\chi \int_0^{s_0} D_h(\xi, s_h)\varphi(\xi, s_h)\,\mathrm{d}\xi \\ D_h(s_0, s_h) &= i\chi \int_0^{s_h} \varphi^*(s_0, \eta) D_0(s_0, \eta)\,\mathrm{d}\eta. \end{aligned} \qquad (9)$$

A previous study (Becker & Al Haddad, 1990) was devoted to the special case where $E = 0$ (no significant long-range order). We wish, in the present paper, to extend the theory to the case where E can take any value between 0 and 1. Notice that $E = 1$ corresponds to the perfect crystal.

It is convenient, following Kato (1980) to decompose D_0 and D_h in the following way:

$$\begin{aligned} D_0 &= \langle D_0 \rangle + \delta D_0 \\ D_h &= \langle D_h \rangle + \delta D_h \end{aligned} \qquad (10)$$

where $\langle D_0 \rangle$ and $\langle D_h \rangle$ are ensemble averages of the wave amplitudes and δD_0, δD_h are amplitude fluctuations. According to (10), the intensities are

$$\begin{aligned} I_0 = \langle |D_0|^2 \rangle &= |\langle D_0 \rangle|^2 + \langle |\delta D_0|^2 \rangle \\ &= \quad I_0^c \quad + \quad I_0^i \\ I_h = \langle |D_h|^2 \rangle &= |\langle D_h \rangle^2 + \langle |\delta D_h|^2 \rangle \\ &= \quad I_h^c \quad + \quad I_h^i. \end{aligned} \qquad (11)$$

Following familiar concepts in optics, $|\langle D_{0,h} \rangle|^2$ is the coherent part of the beam and $\langle |\delta D_{0,h}|^2 \rangle$ the incoherent part.

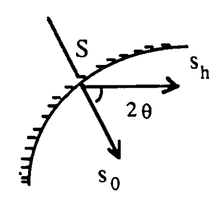


Fig. 1. Incident and diffracted directions.

E plays the role of a static Debye-Waller factor. In analogy with time-dependent fluctuations (vibrations), we expect the coherent intensities to be proportional to E^2, the incoherent ones to be proportional to $(1 - E^2)$. In a perfect crystal, the beams are purely coherent (Al Haddad & Becker, 1990). In a mosaic crystal [$E = 0$, see (I)] the beams are totally incoherent.

II. The coherent wave

Following the discussion by Kato (1980), we start by studying the propagation mechanism for the coherent waves. Taking the ensemble average of (7), we get

$$\begin{aligned} \partial \langle D_0 \rangle / \partial s_0 &= i\chi \langle \varphi D_h \rangle \\ &= i\chi E \langle D_h \rangle + i\chi \langle \delta\varphi D_h \rangle. \end{aligned} \qquad (12)$$

Inserting (9) in (12), we get

$$\begin{aligned} i\chi \langle \delta\varphi D_h \rangle = &-\chi^2 E \int_0^{s_h} \langle \delta\varphi(s_0, s_h) D_0(s_0, \eta) \rangle\,\mathrm{d}\eta \\ &-\chi^2 \int_0^{s_h} \langle D_0(s_0, \eta) \\ &\times \delta\varphi(s_0, s_h)\delta\varphi^*(s_0, \eta) \rangle\,\mathrm{d}\eta. \end{aligned} \qquad (13)$$

The value of $D_0(s_0, \eta)$ is determined through the scattering events at preceding positions (ξ, η) on the optical route: only-nearest neighbour phase correlation is considered, which means that phase coherence is assumed to be lost beyond neighbouring scattering points along an optical route. This approximation allows for neglecting the first term in (13), since $D_0(s_0, \eta)$ is built from scattering events occurring at points that are not nearest neighbours of (s_0, s_h).

We then assume $\langle D_0 \rangle$ and $\langle D_h \rangle$ to have negligible variations in a distance τ, which allows for the following simplification:

$$i\chi \langle \delta\varphi D_h \rangle = -\chi^2 (1 - E^2) \tau \langle D_0 \rangle. \qquad (14)$$

We get the propagation equations for the coherent waves in the form

$$\begin{aligned} \partial \langle D_0 \rangle / \partial s_0 &= i\chi E \langle D_h \rangle - \chi^2 (1 - E^2) \tau \langle D_0 \rangle \\ \partial \langle D_h \rangle / \partial s_h &= i\chi E \langle D_0 \rangle - \chi^2 (1 - E^2) \tau \langle D_h \rangle. \end{aligned} \qquad (15)$$

These statistical equations differ from the original Takagi's equations in two effects. In the first place, χ is replaced by χE: the effective extinction length becomes Λ/E, the structure factor being damped by E. In the second place, an effective absorption factor $[2\chi^2(1 - E^2)\tau]$ occurs for the propagation of the coherent intensities. Energy is lost by the coherent component of the beam during its propagation through the crystal: this lost energy will appear in the form of incoherent beams (Fig. 2). This is the basic mechanism of conversion between coherent and incoherent intensities. At the entrance to the sample, the beam is essentially coherent and through the imperfect phase correlation at various points where scattering occurs, the incoherent beam is gradually created.

The condition of validity of (14) and then (15) is

$$\tau \ll \Lambda/E. \tag{16}$$

The smaller E is, the less stringent this condition is.

It should be noticed that the lengthening of the effective extinction length (Λ/E), which results from the statistical hypotheses introduced earlier, is opposite to what occurs in strained or bent crystals, where the effective extinction length shortens.

To get an explicit solution to (15), boundary conditions have to be discussed for $\langle D_0\rangle$ and $\langle D_h\rangle$. Equations (15) are only valid for s_0, $s_h \gg \tau$. Let us consider a distance ε such that $\tau < \varepsilon \ll s_0, s_h$.

The amplitude of the coherent beam at $(s_0, 0)$ is weakened by scattering events that have taken place before [at positions $(\xi, 0)$ with $\xi < s_0$]. The effective incident amplitude $\langle D_0^0\rangle$ thus obeys the equation

$$\partial\langle D_0^0\rangle/\partial s_0 = -\chi^2(1-E^2)\tau\langle D_0^0\rangle,$$

the solution of which is

$$\langle D_0^0\rangle = \delta(s_h)\exp[-\chi^2(1-E^2)\tau s_0]. \tag{17}$$

$\langle D_h(s_0, \varepsilon)\rangle$ is such that

$$\partial\langle D_h\rangle/\partial s_h = i\chi E\langle D_0^0\rangle$$

so that, after a single scattering,

$$\begin{aligned}\langle D_h(s_0, \varepsilon)\rangle &= i\chi E \exp[-\chi^2(1-E^2)\tau s_0]\int_0^{\varepsilon}\delta(\eta)\,\mathrm{d}\eta\\ &= i\chi E\exp[-\chi^2(1-E^2)\tau s_0].\end{aligned} \tag{18}$$

We can thus adopt the following boundary values for (15):

$$\begin{aligned}&\langle D_0(0, s_h)\rangle = 0\\ &\langle D_h(s_0, 0)\rangle = i\chi E\exp[-\chi^2(1-E^2)\tau s_0]\end{aligned} \tag{19}$$

where only the part of the incident beam which is involved in the scattering is considered. Let us define

$$\mu_e = 2\chi^2(1-E^2)\tau \tag{20}$$

as an effective absorption coefficient. The solution is

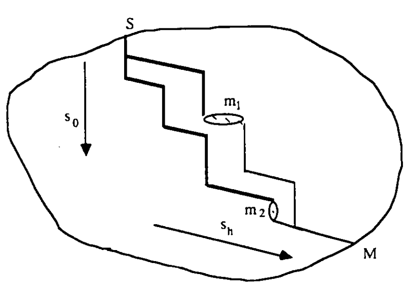


Fig. 2. Two possibilities for decay of coherence by a scattering process at m_1 or m_2.

(Becker, 1977; Kato, 1980)

$$\begin{aligned}\langle D_h\rangle &= i\chi E J_0[2\chi R(s_0 s_h)^{1/2}]\exp[-\tfrac{1}{2}\mu_e(s_0+s_h)]\\ \langle D_0\rangle &= i\chi E(s_0/s_h)^{1/2}\\ &\quad\times J_1[2\chi E(s_0 s_h)^{1/2}]\exp[-\tfrac{1}{2}\mu_e(s_0+s_h)].\end{aligned} \tag{21}$$

J_0 and J_1 are ordinary Bessel functions.

The oscillatory nature of the coherent amplitudes is maintained, Λ being replaced by Λ/E, and the beam being damped by the absorption μ_e.

Finally, we notice that

$$\partial I_0^c/\partial s_0 + \partial I_h^c/\partial s_h = -\mu_e[I_0^c + I_h^c]. \tag{22}$$

This equation gives the rate at which incoherent energy is created from the coherent beam.

III. The incoherent intensity. Kato's approximation

By subtracting $\partial I_0^c/\partial s_0$ and $\partial I_h^c/\partial s_h$ from $\partial I_0/\partial s_0$ and $\partial I_h/\partial s_h$ respectively, one gets the basic propagation equations for the incoherent intensities I_0^i and I_h^i:

$$\begin{aligned}\partial I_0^i/\partial s_0 &= \mu_e I_0^c + i\chi E\{\langle\delta D_0^*\delta D_h\rangle - \langle\delta D_0\delta D_h^*\rangle\}\\ &\quad + i\chi\{\langle D_0^*\delta\varphi D_h\rangle - \langle D_0\delta\varphi^* D_h^*\rangle\}\\ \partial I_h^i/\partial s_h &= \mu_e I_h^c + i\chi E\{\langle\delta D_0\delta D_h^*\rangle - \langle\delta D_0^*\delta D_h\rangle\}\\ &\quad + i\chi\{\langle D_0\delta\varphi^* D_h^*\rangle - \langle D_0^*\delta\varphi D_h\rangle\}.\end{aligned} \tag{23}$$

Insertion of (9) into the brackets allows the introduction of phase and amplitude correlations by a method similar to that used in (I). For instance,

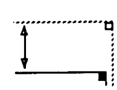
$$\begin{aligned}&i\chi E\langle\delta D_0^*\delta D_h\rangle\\ &\quad = \chi^2 E\int_0^{s_0}\mathrm{d}\xi\langle D_h^*(\xi, s_h)\varphi^*(\xi, s_h)\delta D_h(s_0, s_h)\rangle\\ &\qquad - \chi^2 E\int_0^{s_h}\mathrm{d}\eta\langle\delta D_0^*(s_0, s_h)\varphi^*(s_0, \eta)D_0(s_0, \eta)\rangle.\end{aligned} \tag{24}$$

Limiting the expansion to second order in χ and defining the transverse amplitude correlation functions

$$\begin{aligned}A(s_0, s_h) &= \int_0^{s_h}\mathrm{d}\eta\langle\delta D_0^*(s_0, s_h)\delta D_0(s_0, \eta)\rangle\\ B(s_0, s_h) &= \int_0^{s_0}\mathrm{d}\xi\langle\delta D_h^*(s_0, s_h)\delta D_h(\xi, s_h)\rangle\end{aligned} \tag{25}$$

we get

$$\begin{aligned}&i\chi E\{\langle\delta D_0^*\delta D_h\rangle - \langle\delta D_0\delta D_h^*\rangle\}\\ &\quad = 2\chi^2E^2[B-A]\end{aligned}$$

$= 2\chi^2 \Big\{ -$  $+$  $\Big\}$ (26)

The meaning of the diagrams is the same as in (I), an optical route represented by a full line corresponds to the evolution of D_0 or D_h. An optical route represented by a dotted line corresponds to the evolution of D_0^* or D_h^* where ■: $\langle\varphi\rangle$; □: $\langle\varphi^*\rangle$. For simplicity, we assume A and B to be real. ($\leftrightarrow$) stands for amplitude correlation between the two routes.

Then, we consider a quantity such as

$$\begin{aligned} i\chi\langle D_0^*\delta\varphi D_h\rangle = \chi^2 \int_0^{s_0} & \mathrm{d}\xi\langle D_h^*(\xi, s_h)D_h(s_0, s_h) \\ & \times \delta\varphi(s_0, s_h)\varphi^*(\xi, s_h)\rangle \\ & -\chi^2 \int_0^{s_h} \mathrm{d}\eta\langle D_0^*(s_0, s_h)D_0(s_0, \eta) \\ & \times \delta\varphi(s_0, s_h)\varphi^*(s_0, \eta)\rangle. \end{aligned} \tag{27}$$

If the expansion is again limited to second order in χ:

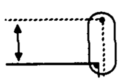
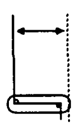
$$\begin{aligned} i\chi\langle D_0^*\delta\varphi D_h\rangle &= \chi^2(1-E^2) \int_0^{s_0} \mathrm{d}\xi\, g(s_0-\xi) \\ & \quad \times\langle D_h^*(\xi, s_h)D_h(s_0, s_h)\rangle \\ & \quad -\chi^2(1-E^2) \int_0^{s_h} \mathrm{d}\eta\, g(s_h-\eta) \\ & \quad \times\langle D_0^*(s_0, s_h)D_0(s_0, \eta)\rangle \\ &= \chi^2(1-E^2)\tau[I_h^c - I_0^c] \\ & \quad +\chi^2(1-E^2)[B'-A'] \end{aligned} \tag{28}$$

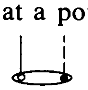
where A' and B' are mixed phase and amplitude correlation functions [already defined in (I)]:

$$\begin{aligned} A'(s_0, s_h) &= \int_0^{s_h} \mathrm{d}\eta\, g(s_h-\eta) \\ & \quad \times\langle \delta D_0^*(s_0, s_h)\delta D_0(s_0, \eta)\rangle \\ B'(s_0, s_h) &= \int_0^{s_0} \mathrm{d}\xi\, g(s_0-\xi) \\ & \quad \times\langle \delta D_h^*(s_0, s_h)\delta D_h(\xi, s_h)\rangle. \end{aligned} \tag{29}$$

Thus, it is possible to write

$$\begin{aligned} & \mu_e I_0^c + i\chi\{\langle D_0^*\delta\varphi D_h\rangle - \langle D_0\delta\varphi^* D_h^*\rangle\} \\ & = \mu_e I_h^c + 2\chi^2(1-E^2)[B'-A'] \end{aligned}$$

$= \mu_e I_h^c + 2\chi^2$  $-2\chi^2$  (30)

• stands for $\delta\varphi$ at a point where scattering occurs, and ○ for $\delta\varphi^*$.  has the meaning of a phase correlation among two routes, as already discussed in (I).

If we consider the conversion from coherent beams into incoherent intensity, we observe that incoherence can only occur through a partial phase coupling between two different paths. From (30) we see that the first diagram is compatible with such a process. But this is not true for the second diagram where the correlation involves two points on the same optical path. The diagrams show clearly the intensity at (s_0, s_h) as resulting from the coupled amplitudes at preceding positions, along either the incident or diffracted direction.

Summing up the previous results, one gets

$$\begin{aligned} \partial I_0^i/\partial s_0 &= \mu_e I_h^c + 2\chi^2 E^2[B-A] \\ & \quad +2\chi^2(1-E^2)[B'-A'] \\ \partial I_h^i/\partial s_0 &= \mu_e I_0^c + 2\chi^2 E^2[A-B] \\ & \quad +2\chi^2(1-E^2)[A'-B'] \end{aligned} \tag{31}$$

I_h^c is the source for I_0^i, I_0^c the source for I_h^i. We also observe that (31) reduces to (I.20) when $E\to 0$.

Kato's approximation

Kato (1980) made the following intuitive simplification. He defined Γ as the common width of $\langle\delta D_0^*(s_0, s_h)\delta D_0(s_0, \eta)\rangle$ and $\langle\delta D_h^*(s_0, s_h)\sigma D_h(\xi, s_h)\rangle$. Γ was supposed to be constant. Assuming $\Gamma \ll s_0, s_h$, one gets

$$\begin{aligned} A(s_0, s_h) &\simeq \Gamma I_0^i(s_0, s_h) \\ B(s_0, s_h) &\simeq \Gamma I_h^i(s_0, s_h). \end{aligned} \tag{32}$$

In principle, Γ should be derivable from the theory, but Kato proposed an order of magnitude:

$$\Gamma \simeq \Lambda/E \tag{33}$$

and thus

$$\Gamma \gg \tau. \tag{34}$$

This last inequality leads to the following simplification for A' and B' [already discussed in (I)].

$$\begin{aligned} A'(s_0, s_h) &\simeq \tau I_0^i(s_0, s_h) \\ B'(s_0, s_h) &\simeq \tau I_h^i(s_0, s_h). \end{aligned} \tag{35}$$

If the effective correlation length τ_e is defined by

$$\tau_e = (1-E^2)\tau + E^2\Gamma \tag{36}$$

the propagation equations reduce to

$$\begin{aligned} \partial I_0^i/\partial s_0 &= \mu_e I_h^c + 2\chi^2\tau_e[I_h^i - I_0^i] \\ \partial I_h^i/\partial s_h &= \mu_e I_0^c + 2\chi^2\tau_e[I_0^i - I_h^i]. \end{aligned} \tag{37}$$

A solution can be obtained to these equations (Kato, 1980). The present authors proposed some improvements to the solution (Al Haddad & Becker, 1988; Guigay, 1989). Though mathematically complex, it is feasible to use such a theory for refining extinction as long as E and τ_e are modelled in terms of their $\mathbf{h}$ dependence (Becker & Al Haddad, 1989).

Despite its simplicity, Kato's approximation seems questionable. This point was the object of (I) in the

Table 1. *Expansion of* X *in terms of independent scattering schemes*; $X = \sum x_i - \sum x_i'$

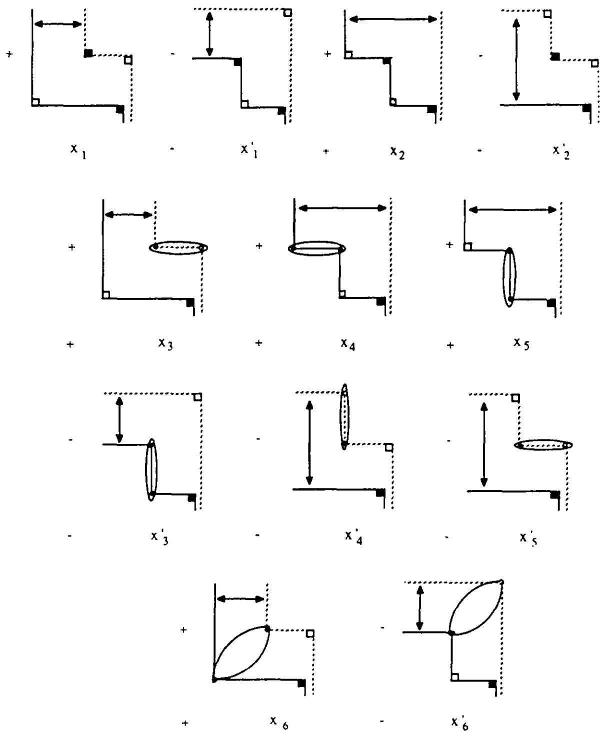
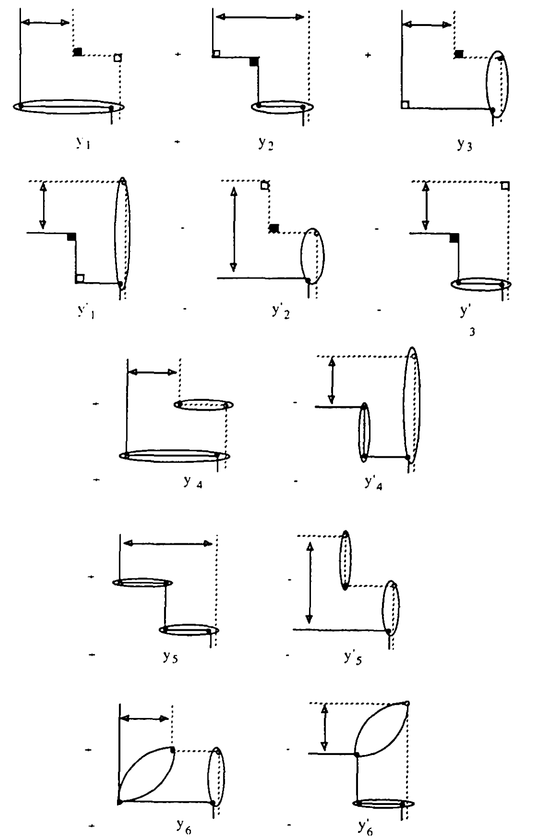


Table 2. *Expansion of* Y *in terms of independent scattering schemes*; $Y = \sum (y_i - y_i')$



case where E is very small. We were able to show that, for $E \ll 1$,

$$A' = \tau_2 I_0^i, \quad B' = \tau_2 I_h^i. \tag{I.32}$$

This modification from (35) means that (33) is not valid for small E. We can infer from (I.32) that, for small E, Γ is of the order of τ.

Furthermore, it is not obvious that Γ should be a constant, or be the same for A and B.

These questions are related to the practical difficulties encountered when trying to use (37) and its solution on experimental data. It is for these reasons that the theory will be extended further, by an expansion of $\partial I_0^i/\partial s_0$ and $\partial I_h^i/\partial s_h$ of equations (23) to fourth order in χ, in order to find self-consistent equations satisfied by A, B, A', B'.

IV. Improved theory for the incoherent intensities

The expansion of $\partial I_0^i/\partial s_0$ (or $\partial I_h^i/\partial s_h$) to fourth order in χ can be represented by the diagrams shown in Tables 1 and 2:

$$\partial I_0^i/\partial s_0 = 2\chi^4[X + Y] + \mu_e I_h^c. \tag{38}$$

Some of these terms represent conversion of coherent into incoherent intensity at points preceding the actual position (s_0, s_h). Since a coherent beam can only become incoherent by phase coupling between two different paths, the only diagrams that correspond to this coherent-incoherent conversion are x_6, x_6', y_6, y_6'.

The detailed calculation of X is presented in the Appendix, the result being the following:

$$E^2[B - A] = \chi^2 X \tag{39}$$

with

$$\begin{aligned}\chi^{-2}\{B-A\} &= 2E^2 \int_0^{s_0} \mathrm{d}\xi \int_0^{s_h} \mathrm{d}\eta\, [A-B](\xi,\eta)\\ &\quad + 2(1-E^2)\tau\left\{\int_0^{s_0} \mathrm{d}\xi\, A(\xi, s_h) - \int_0^{s_h} \mathrm{d}\eta\, B(s_0,\eta)\right\}\\ &\quad + 2(1-E^2)\tau\left\{\int_0^{s_0} \mathrm{d}\eta\, A'(s_0,\eta) - \int_0^{s_0} \mathrm{d}\xi\, B'(\xi, s_h)\right\}\\ &\quad + 2(1-E^2)\tau\left\{\int_0^{s_h} \mathrm{d}\eta\, I_0^c(s_0,\eta) - \int_0^{s_0} \mathrm{d}\xi\, I_h^c(\xi, s_h)\right\}.\end{aligned} \tag{40}$$

A similar calculation for Y (Appendix) leads to

$$(1-E^2)[B' - A'] = \chi^2 Y \tag{41}$$

with

$$\chi^{-2}\{B'-A'\}=2E^2\tau\int_0^{s_0} d\xi\,[A-B](\xi,s_h)$$
$$+2E^2\tau\int_0^{s_h} d\eta\,[A-B](s_0,\eta)$$
$$+2(1-E^2)\tau_2\int_0^{s_0} d\xi\,(A'-B')(\xi,s_h)$$
$$+2(1-E^2)\tau_2\int_0^{s_h} d\eta\,(A'-B')(s_0,\eta)$$
$$+2(1-E^2)\tau\tau_2$$
$$\times\left\{\int_0^{s_h} d\eta\, I_0^c(s_0,\eta)-\int_0^{s_0} d\xi\, I_0^c(\xi,s_h)\right\}. \quad (42)$$

V. Modified propagation equations for the incoherent beams

Equations (31), (40), (42) are in fact very difficult to solve directly. Some simplifications are needed in order to get practical propagation equations for the incoherent intensities.

Insertion of (31) into (42) allows one to write

$$B'-A'=\tau_2[I_h^i-I_0^i]+2\chi^2E^2(\tau-\tau_2)$$
$$\times\left\{\int_0^{s_0}[A-B](\xi,s_h)\,d\xi+\int_0^{s_h}[A-B](s_0,\eta)\,d\eta\right\}. \quad (43)$$

Similarly, (40) can be transformed into

$$(B-A)=\hat{L}[A-B]+\tau[I_h^i-I_0^i]$$
$$+2\chi^2(1-E^2)\tau\int_0^{s_0}(A-A')(\xi,s_h)\,d\xi$$
$$+2\chi^2(1-E^2)\tau\int_0^{s_h}(B-B')(s_0,\eta)\,d\eta$$
$$+2\chi^2E^2\tau\int_0^{s_0}(A-B)(\xi,s_h)\,d\xi$$
$$+2\chi^2E^2\tau\int_0^{s_h}(A-B)(s_0,\eta)\,d\eta \quad (44)$$

where $\hat{L}$ stands for the propagator operator:

$$\hat{L}f(s_0,s_h)=2\chi^2E^2\int_0^{s_0}d\xi\int_0^{s_0}d\eta\, f(\xi,\eta). \quad (45)$$

If A and B are assumed to have slow variations on a distance such as Λ, the last two integrals in (44) can be neglected with respect to $\hat{L}[A-B]$.

If $E^2/(1-E^2)$ is larger than 1, its seems also tempting to discard the two integrals involving $(A-A')$ and $(B-B')$.

Under such drastic simplifications, (44) becomes

$$[1+\hat{L}](B-A)=\tau(I_h^iI_0^i). \quad (46)$$

It can be inverted into

$$B-A=\tau[1+\hat{L}]^{-1}(I_h^i-I_0^i). \quad (47)$$

From (31), the two integrals of (43) can also be neglected in the expansion of $\partial I_0/\partial s_0$ or $\partial I_h/\partial s_h$.

Finally, one can write the following propagation equations for the incoherent intensities

$$\begin{aligned}\partial I_0^i/\partial s_0&=2\chi^2(1-E^2)I_h^c+2\chi^2\{E^2\tau[1+\hat{L}]^{-1}\\&\quad+(1-E^2)\tau_2\}(I_h^i-I_0^i)\\ \partial I_h^i/\partial s_h&=2\chi^2(1-E^2)I_0^c+2\chi^2\{E^2\tau[1+\hat{L}]^{-1}\\&\quad+(1-E^2)\tau_2\}(I_0^i-I_h^i).\end{aligned} \quad (48)$$

If we define the operator

$$\hat{\tau}_e=E^2\hat{\Gamma}+(1-E^2)\tau_2, \quad (49)$$

$$\hat{\Gamma}=\tau(1+\hat{L})^{-1}=\tau\sum_0^\infty(-1)^n\hat{L}^n, \quad (50)$$

$$\begin{aligned}\partial I_0^i/\partial s_0&=2\chi^2(1-E^2)I_h^c+2\chi^2\hat{\tau}_e[I_h^i-I_0^i]\\ \partial I_h^i/\partial s_h&=2\chi^2(1-E^2)I_0^c+2\chi^2\hat{\tau}_e[I_0^i-I_h^i].\end{aligned} \quad (51)$$

Equations (51) are a generalization of Kato's equations (37), where $\hat{\Gamma}$ is an operator and not a constant any more. Equation (50) can be written as

$$\hat{\Gamma}=\Gamma_0+\hat{\Gamma}^1 \quad (52)$$

with $\Gamma_0=\tau$ and

$$\hat{\Gamma}^1=\sum_{n=1}^\infty(-1)^n\tau\hat{L}^n. \quad (53)$$

Since the series is alternate, we may expect that Γ_0 is an important part of $\hat{\Gamma}$, Γ' corresponding to fluctuations around Γ_0, and its value is drastically different from (Λ/E), the expression proposed by Kato. The natural boundary values associated with (51) are (generalizing the argument proposed by Kato

$$\begin{aligned}&I_0^i=0\\ &I_h^i(s_0,0)\simeq\chi^2(1-E^2)\\&\qquad\times\exp\{-2\chi^2[E^2\tau+(1-E^2)\tau_2]s_0\}.\end{aligned} \quad (54)$$

It should be noticed that when $E\to0$ equations (51) have the proper limit, identical to the propagation equations derived in (I). Therefore, (51) might have a reasonable behaviour for the whole range of E values.

Remark 1. Equations (51) are difficult to solve directly, since $(I_h^i-I_0^i)$ occur through an integro-differential equation.

The crudest but simplest method of solution consists in neglecting the fluctuating part of $\hat{\Gamma}$ and writing

$$\hat{\Gamma}\simeq\tau. \quad (55)$$

In such a condition, the problem is equivalent to the theory proposed by Kato (1980) and modified by Al Haddad & Becker (1988), with the only modification concerning Γ.

Remark 2. If one wishes to go beyond the approximation (55), we propose the following alternate procedure. Let us go back to (40) for $(B-A)$ and its various contributions (see Appendix for the expressions for the various terms x_i).

$$x_3+x_4=2E^2(1-E^2)\int_0^{s_0}\mathrm{d}\xi\int_0^{s_h}\mathrm{d}\eta\, A(\xi,\eta)g(s_h-\eta).$$

If $\tau \ll s_0$, s_h, we may write

$$x_3+x_4\ll x_1 \qquad x'_3+x'_4\ll x'_1.$$

In any case,

$$(x_3+x_4)/x_2\simeq(1-E^2)\tau/E^2\tau.$$

If this ratio is not large compared to 1, which means that E is not too small, it is legitimate to neglect x_3 and x_4 in addition to x_2 and, similarly, the term (in x_6)

$$\begin{aligned}&2E^2(1-E^2)\tau\int_0^{s_h}\mathrm{d}\eta\, A'(s_0,\eta)\\&\quad\simeq 2E^2(1-E^2)\int_0^{s_0}\mathrm{d}\xi\int_0^{s_h}\mathrm{d}\eta\, g(s_0-\xi)A'(\xi,\eta)\\&\quad< 2E^2(1-E^2)\int_0^{s_0}\mathrm{d}\xi\int_0^{s_h}\mathrm{d}\eta\, g(s_0-\xi)A(\xi,\eta)\\&\quad\ll x_1.\end{aligned}$$

With these simplifications, we get

$$[B-A]=\hat{L}[A-B]+2\chi^2(1-E^2)\tau^2\times\left[\int_0^{s_h}\mathrm{d}\eta\, I_0^c(s_0,\eta)-\int_0^{s_0}\mathrm{d}\xi\, I_h^c(\xi,s_h)\right]. \quad (56)$$

If F is the source function,

$$F=2\chi^2(1-E^2)\tau^2\times\left[\int_0^{s_h}\mathrm{d}\eta\, I_0^c(s_0,\eta)-\int_0^{s_0}\mathrm{d}\xi\, I_h^c(\xi,s_h)\right], \quad (57)$$

uniquely defined by the coherent beam, (56) can be written as

$$[1+\hat{L}](B-A)=F, \quad (58)$$

the solution of which is

$$\begin{aligned}(B-A)&=[1+\hat{L}]^{-1}F\\&=\sum_{n=0}^{\infty}(-1)^n\hat{L}^n(F)\end{aligned}$$

where now $(B-A)$ is calculable from the coherent intensities.

Finally, the propagation equations take the alternative form

$$\begin{aligned}\partial I_0^i/\partial s_0&=2\chi^2(1-E^2)\tau I_h^c+2\chi^2E^2[1+\hat{L}]^{-1}F\\&\quad+2\chi^2(1-E^2)\tau_2[I_h^i-I_0^i]\\\partial I_h^i/\partial s_h&=2\chi^2(1-E^2)\tau I_0^c-2\chi^2E^2[1+\hat{L}]^{-1}F\\&\quad+2\chi^2(1-E^2)\tau_2[I_0^i-I_h^i].\end{aligned} \quad (59)$$

The solution to (59) should not be very different from that to (51).

VI. Solution for the incoherent part of the beam

The purpose of this section is to find the solution to (59), together with the boundary conditions (54). Equations (59) can be written as

$$\begin{aligned}\partial I_0^i/\partial s_0&=\sigma(1-E^2)I_h'^c+\mu_2(I_h^i-I_0^i)\\\partial I_h^i/\partial s_h&=\sigma(1-E^2)I_0'^c+\mu_2(I_0^i-I_h^i)\end{aligned} \quad (60)$$

where

$$\sigma=2\chi^2\tau,\quad \mu_2=2\chi^2(1-E^2)\tau_2, \quad (61)$$

$I_0'^c$ and $I_h'^c$ are the effective sources for the incoherent beams and are given by

$$\begin{aligned}\sigma(1-E^2)I_0'^c&=\sigma(1-E^2)I_0^c-2\chi^2E^2[1+\hat{L}]^{-1}F\\\sigma(1-E^2)I_h'^c&=\sigma(1-E^2)I_h^c+2\chi^2E^2[1+\hat{L}]^{-1}F.\end{aligned} \quad (62)$$

By the use of (45), (56) and (57), (62) can be transformed into

$$\begin{aligned}I_h'^c&=I_0^c-\alpha^2\tau f_1*I_0^c+\alpha^2\tau f_2*I_h^c\\I_0'^c&=I_h^c-\alpha^2\tau f_2*I_h^c+\alpha^2\tau f_1*I_0^c\end{aligned} \quad (63)$$

in which $*$ stands for the convolution product

$$f*g=\int_0^{s_0}\mathrm{d}\xi\int_0^{s_h}\mathrm{d}\eta\, f(\xi,\eta)g(s_0-\xi,s_h-\eta),$$

α is defined by

$$\alpha^2=2\chi^2E^2$$

and

$$\begin{aligned}f_1&=\delta(s_0)-\alpha(s_h/s_0)^{1/2}J_1[2\alpha(s_0s_h)^{1/2}]\\f_2&=\delta(s_h)-\alpha(s_0/s_h)^{1/2}J_1[2\alpha(s_0s_h)^{1/2}].\end{aligned} \quad (64)$$

Equations (60) are similar to (37), except for the change of some constants and the replacement of $I_{0,h}^c$ by $I_{0,h}'^c$. Their solution has been discussed by Kato (1980) and Al Haddad & Becker, (1988, § II.4). By the same technique, one gets

$$\begin{aligned}I_h^i&=(1-E^2)[\sigma g_1*I_0'^c+\sigma g_0*I_h'^c+(\chi^2/\mu_2)\\&\quad\times(g_0-\sigma_1g_3)]\\I_0^i&=(1-E^2)[\sigma g_2*I_0'^c+\sigma g_0*I_h'^c+\chi^2g_3]\end{aligned} \quad (65)$$

where

$$g_0 = \mu_2 \mathbb{I}_0[2\mu_2(s_0 s_h)^{1/2}] \exp[-\mu_2(s_0+s_h)]$$
$$g_1 = \{\delta(s_0) + \mu_2(s_h/s_0)^{1/2}\mathbb{I}_1[2\mu_2(s_0 s_h)^{1/2}]\} \times \exp[-\mu_2(s_0+s_h)]$$
$$g_2 = \{\delta(s_h) + \mu_2(s_0/s_h)^{1/2}\mathbb{I}_1[2\mu_2(s_0 s_h)^{1/2}]\} \times \exp[-\mu_2(s_0+s_h)] \tag{66}$$
$$g_3 = \{\exp[-\mu_3(s_0+s_h)]\delta(s_h)\} * g_0 = \sum_{n=0}^{\infty} (-\sigma_1/\mu_2)^n [(s_0 s_h)^{1/2}]^{n+1} \times \mathbb{I}_{n+1}[2\mu_2(s_0 s_h)^{1/2}] \exp[-\mu_2(s_0+s_h)]$$

with

$$\sigma_1 = \mu_3 - \mu_2$$
$$\mu_3 = 2\chi^2[(1-E^2)\tau_2 + E^2\tau],$$

$\mathbb{I}_0$, $\mathbb{I}_1$ are modified Bessel functions.

Remark. If $\sigma_1 = 0$, g_3 becomes

$$g_3 = (s_0/s_h)^{1/2}\mathbb{I}_1[2\mu_2(s_0 s_h)^{1/2}] \exp[-\mu_2(s_0+s_h)].$$

From (63) the incoherent intensities are finally written as

$$I_0^i = (1-E^2)\{\sigma[(1+\alpha^2\tau/\mu_2)g_0 - (\alpha^3\tau/\mu_2)g_0 * f_0] * I_0^c + \sigma[g_2 - \tau\alpha^2 g_2 * f_2 + \tau\alpha^2 g_0 * f_2] * I_h^c + \chi^2 g_3\}$$
$$I_h^i = (1-E^2)\{\sigma[g_1 - \tau\alpha^2 g_1 * f_1 + \tau\alpha^2 g_0 * f_1] * I_0^c + \sigma[(1+\alpha^2\tau/\mu_2)g_0 - (\alpha^3\tau/\mu_2)g_0 * f_0] * I_h^c + (\chi^2/\mu_2)[g_0 - \sigma_1 g_3]\} \tag{67}$$

where

$$f_0 = \alpha J_0[2\alpha(s_0 s_h)^{1/2}].$$

Like Al Haddad & Becker (1988), one finds that I_h^i contains a purely incoherent part:

$$I_h^{i0} = (1-E^2)(\chi^2/\mu_2)[g_0 - \mu_2 g_3]$$

The part

$$I_h^{i,00} = (1-E^2)(\chi^2/\mu_2)g_0 = (1-E^2)\chi^2\mathbb{I}_0[2(s_0 s_h)^{1/2}] \exp[-\mu_2(s_0+s_h)]$$

becomes the leading contribution in the limit $E \to 0$. It corresponds to pure secondary extinction.

The other terms are the 'mixed components' corresponding to the fact that incoherence is generated from the coherent beams, through incomplete phase correlation, at various depths in the sample.

VII. Integrated intensity for a parallel plate in symmetrical transmission geometry

If one writes

$$I_h = I_h^c + I_h^i, \qquad I_h^i = I_h^{i,0} + \sum_{n=1}^{5} I_h^{i,n}, \tag{68}$$

I_h^c and $I_h^{i,0}$ are the purely coherent and incoherent contributions and the five other terms correspond to the mixed contributions

$$I_h^{i,1} = \sigma(1-E^2)g_1 * I_0^c$$
$$I_h^{i,2} = \sigma(1-E^2)[1+(\alpha^2\tau/\mu_2)]g_0 * I_h^c$$
$$I_h^{i,3} = -\tau\alpha^2\sigma(1-E^2)[g_1 * f_1] * I_0^c \tag{69}$$
$$I_h^{i,4} = \sigma(1-E^2)\tau\alpha^2[g_0 * f_0]I_0^c$$
$$I_h^{i,5} = -(\tau\alpha^3/\mu_2)\sigma(1-E^2)[g_0 * f_0]I_h^c.$$

The integrated power (Kato, 1976; Becker, 1977; Al Haddad & Becker, 1988) can be decomposed as

$$P = P^c + P^i$$
$$P^i = P^{i,0} + M \tag{70}$$
$$M = \sum_{l=1}^{5} M_l.$$

M_l is the integrated intensity corresponding to $I_h^{i,l}$.

VII-1. *Calculation of P^c and $P^{i,0}$*

For a parallel plate in symmetrical Laue geometry P^c and $P^{i,0}$ have been calculated by Kato (1980), but τ_e has to be replaced by τ_2. The result is

$$P^c = E^2 QTW(2ET/\Lambda) \exp[-2(1-E^2)\tau T/\Lambda^2]$$
$$P^{i,00} = (\Lambda Q \Lambda / 2\tau_2) \times \sinh[2(1-E^2)(\tau_2/\Lambda)(T/\Lambda)] \times \exp[-2(1-E^2)\tau_2 T/\Lambda^2], \tag{71a}$$

with $T = t/\cos\theta$, t being the thickness of the plate.

$$Q = (\lambda/\sin 2\theta)|\chi|^2$$

and the function $W(x)$ is given by

$$W(x) = \int_0^1 J_0(x\rho)\,d\rho. \tag{72}$$

$P^{i,00}$ is the integrated intensity corresponding to $I_h^{i,00}$.

In order to obtain (71), one first integrates the intensity on the portion of the exit surface excited by the point source S (region ab in Fig. 3), then one sums over the various possible source points S.

In the same way, one can obtain

$$P^{i,0} = P^{i,00} + (1-E^2)[Q\sigma_1/\mu_3(\mu_2-\sigma_1)][\exp(-\mu_3 T) + (\sigma_1/\mu_2)\exp(-\mu_2 T)\sinh(\mu_2 T) - \exp(-\mu_2 T)\cosh(\mu_2 T)]. \tag{71b}$$

VII-2. *Calculation of M*

VII-2(*a*) *Calculation of M_1 and M_2.* $I_h^{i,1}$ and $I_h^{i,2}$ are convolutions between an incoherent and a coherent intensity. The coherent part originates from S and the incoherent phenomenon grows from an intermediate position m' in the sample (Fig. 4).

To calculate M_1 or M_2 one first chooses a point m' at a given depth t' and thus integrates the intensity over the segment (a_1b_1) associated with m'. Then one varies the depth t' of m' (variables x', t' parallel and orthogonal to the plate are used for m', rather than ξ' and η'; notice that $\sin 2\theta\, d\xi'\, d\eta' = dx'\, dt'$). Finally, it is necessary to vary S along the entrance surface. One gets

$$\begin{aligned} M_1 &= \sigma(1-E^2)E^2Q[G_1 * L_0]_T \\ M_2 &= \sigma(1-E^2)E^2 \\ &\quad \times\{1+E^2\tau/[(1-E^2)\tau_2]\}Q[G_0 * L_h]_T \end{aligned} \tag{73}$$

with

$$\begin{aligned} G_i(t) &= \tfrac{1}{2}t \int_{-1}^{1} g_i(\xi, t)\, d\xi, \quad i=0,1,2 \\ E^2\chi^2L_v(t) &= \tfrac{1}{2}t \int_{-1}^{1} I_v^c(\xi, t)\, d\xi, \quad v=0,\ h \end{aligned} \tag{74}$$

Therefore, G_0, G_1, L_0, L_h are given by

$$\begin{aligned} G_0(t) &= \exp(-\mu_2 t)\sinh(\mu_2 t) \\ G_1(t) &= \exp(-\mu_2 t)\cosh(\mu_2 t) \\ L_0(t) &= t[1-W(2\chi Et)]\exp(-\mu_e t) \\ L_h(t) &= tW(2\chi Et)\exp(-\mu_e t), \end{aligned} \tag{75}$$

μ_e being given by (20).

VII-2(b) *Calculation of* M_3, M_4 *and* M_5. We employ a method similar to that used for M_1 and M_2, but now $I_h^{i,3}$, $I_h^{i,4}$, $I_h^{i,5}$ are convolution products between three functions. One needs two points m' and m'' (Fig. 5). First one fixes m' and m'' and integrates the intensity over a_1b_1. Then one varies m' along $a'b'$, m'' being fixed. One then varies m'' along $a''b''$, and finally S along the entrance surface. One finds

$$\begin{aligned} M_3 &= -\tau\alpha^2\sigma E^2(1-E^2)Q[G_1 * F_1 * L_0]_T \\ M_4 &= \tau\alpha^2\sigma E^2(1-E^2)Q[G_0 * F_1 * L_0]_T \\ M_5 &= -(\tau\alpha^3\sigma/\mu_2)E^2(1-E^2)Q[G_0 * F_0 * L_h]_T \end{aligned} \tag{76}$$

with

$$F_i(t) = (t/2)\int_{-1}^{1} f_i(\xi, t)\, d\xi, \quad i=0,1,2,$$

i.e.

$$F_0 = \sin(\alpha t), \quad F_1 = \cos(\alpha t). \tag{77}$$

VII-2(c) *Final expressions for M.* It is possible, using notations that generalize Kato's (1980) expressions, to write

$$\begin{aligned} M &= \tfrac{1}{2}\sigma(1-E^2)E^2Q[4\mu_2 a(Z_1+\tfrac{1}{2}Z_2)+\gamma Z_3 \\ &\quad - a(2\mu_2 n_1+\alpha n_2)+(1+2\mu_2 a)n_3+n_4] \end{aligned} \tag{78}$$

where

$$\begin{aligned} a &= 2\tau\alpha^2/(\alpha^2+4\mu_2^2), \\ \gamma &= [a(\alpha^2-4\mu_2^2)-2\tau\alpha^2]/2\mu_2-2 \end{aligned} \tag{79}$$

and

$$\begin{aligned} Z_1 &= F_1 * L_h \\ &= (\alpha^2+\mu_e^2)^{-1}(\{\exp(-\mu_e t)\, J_0(2\chi Et) \\ &\quad * [\mu_e\cos(\alpha t)+\alpha\sin(\alpha t)]\}_T \\ &\quad - \mu_e T\exp(-\mu_e t)\, W(2\chi ET)) \\ Z_2 &= F_0 * L_h \\ &= (\alpha^2+\mu_e^2)^{-1}(\{\exp(-\mu_e t)\, J_0(2\chi Et) \\ &\quad * [\mu_e\sin(\alpha t)-\alpha\cos(\alpha t)]\}_T \\ &\quad + \alpha T\exp(-\mu_e t)\, W(2\chi ET)) \\ Z_3 &= \exp(-2\mu_2 t) * L_h \\ &= [T/(2\mu_2-\mu_e)][\exp(-\mu_e T)\, W(2\chi ET) \\ &\quad - \exp(-2\mu_2 T)\, W_e((2\mu_2-\mu_e)T, 2\chi ET)] \end{aligned}$$

$$W_e(x, y) = \int_0^1 \exp(-x\rho)J_0(y\rho)\, d\rho \tag{80}$$

$$\begin{aligned} Z_4 &= \int_0^T L_h(t)\, dt \\ &= (T/\mu_e)[W_e(-\mu_e T, 2\chi ET) \\ &\quad - \exp(-\mu_e T)\, W(2\chi ET)] \\ n_1 &= \{[t\exp(-\mu_e t)] * F_1\}_T \\ &= (\alpha^2+\mu_e^2)^{-2}[(\mu_e^2-\alpha^2)\cos(\alpha T) \\ &\quad + 2\mu_e\alpha\sin(\alpha T) - (\mu_e^2-\alpha^2)\exp(-\mu_e T) \\ &\quad - \mu_e T(\mu_e^2+\alpha^2)\exp(-\mu_e T)] \\ n_2 &= \{[t\exp(-\mu_e t)] * F_0\}_T \\ &= (\alpha^2+\mu_e^2)^{-2}[(\mu_e^2-\alpha^2)\sin(\alpha T) \\ &\quad - 2\mu_e\alpha\cos(\alpha T) + 2\mu_e\alpha\exp(-\mu_e T) \\ &\quad + \alpha T(\mu_e^2+\alpha^2)\exp(-\mu_e T)] \\ n_3 &= \{\exp(-2\mu_2 t) * [t\exp(-u_e t)]\}_T \\ &= (2\mu_2-\mu_e)^{-2}\{\exp(-\mu_2 T) \\ &\quad + [(2\mu_2-\mu_e)T-1]\exp(-\mu_e T)\} \\ n_4 &= 1/\mu_e^2 - (T/\mu_e + 1/\mu_e^2)\exp(-\mu_e T); \end{aligned}$$

if $\tau_2 = \frac{1}{2}\tau$,

$$n_3 = \tfrac{1}{2}T^2\exp(-\mu_e T).$$

We present two examples in Fig. 6, corresponding to $E = 0.99$ and $\tau/\Lambda = 0.1$ for Fig. 6(a), $E = 0.9$ and $\tau/\Lambda = 0.1$ for Fig. 6(b). $Z = YA$ is plotted as a function of A, where $A = T/\Lambda$ and Y is the extinction factor, equal to (P/P_{kin}). Z^c corresponds to the coherent contribution, Z^c ($E=1$) to the pure dynamical

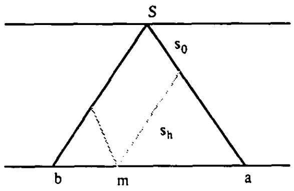


Fig. 3. Geometry for the integration of P^c or $P^{i,0}$.

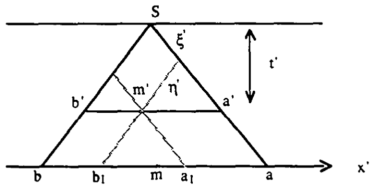


Fig. 4. Geometry for the calculation of m_1 and m_2.

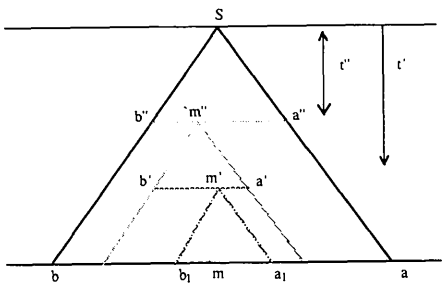


Fig. 5. Geometry for calculating m_3, m_4, m_5.

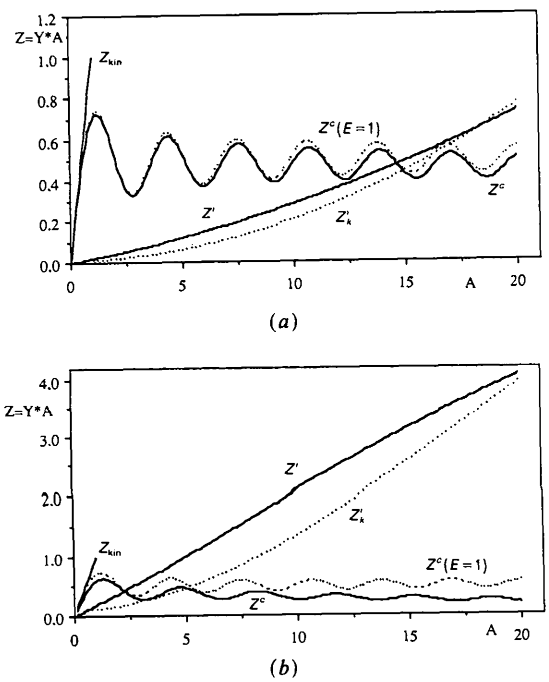


Fig. 6. $Z=P/Q\Lambda$ plotted as a function of $A=T/\Lambda$. (a) $E=0.99$, $\tau/\Lambda=0.1$; (b) $E=0.9$, $\tau/\Lambda=0.1$. Z^c: coherent term (——); Z^i: incoherent term (——); $Z^c(E=1)$: purely dynamical theory (·· ·· ··); Z^i_k: incoherent term with Kato's equations (········). Notice that $Z_{\rm kin}=A$.

result (perfect crystal), Z^i is the incoherent part resulting from the present theory, Z^i_k the incoherent part using Kato's equations (37) with the solution of Al Haddad & Becker (1988). The deviation from dynamical theory is apparent. But the most important observation is the significant modification of the incoherent part, which is larger with the present theory than using Kato's equations [Z^i_k is already significantly larger than Kato's original result (Al Haddad & Becker, 1988)].

It is observed that, even with strong values for E, the incoherent contribution dominates when the thickness is large enough. For $E=0.9$, the crossing occurs for $T/\Lambda=2.5$.

This fact is striking and mixed terms are important in the crossing region, which shows the importance of using this new theory which does not introduce an arbitrary separation between secondary and primary extinction.

VIII. Application: annealed Czochralski-grown silicon with a high degree of perfection

Schneider, Gonçalves, Rollason *et al.* (1988) have studied by γ-ray diffraction (wavelength of the incident radiation $\lambda=0.00392$ Å) various crystals of silicon grown by the Czochralski method. They used disc shape samples 10 cm in diameter and 1 cm thick. The oxygen content varied approximately between 10 and 30 atoms in 10^6. Various samples were prepared, corresponding to different annealing temperature and duration. For each sample, the integrated diffracted power was measured in Laue geometry as a function of the effective thickness (which was varied by rotating the crystal around the reciprocal-lattice vector **h** under study).

An example is given in Fig. 7, corresponding to **h** = 220. *Pendellösung* oscillations are well resolved, indicating the high degree of perfection of the sample. However, dynamical theory is unable to reproduce the observed intensities, which are about 50% higher than predicted for a perfect crystal.

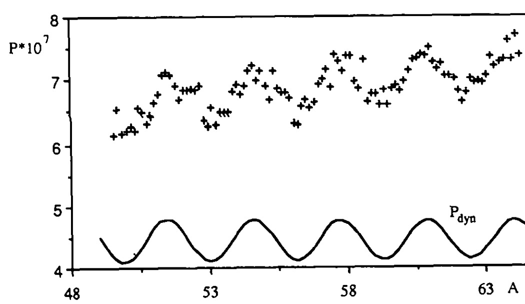


Fig. 7. Integrated diffracted power for the 220 reflexion of a silicon crystal (from Schneider, Gonçalves, Rollason *et al.*, 1988) containing 7.7×10^{17} oxygen atoms cm^{-3}, after annealing for 70 h at 1043 K. Solid line: the result obtained by dynamical theory (perfect crystal).

(1) In order to explain the deviation from $P_{\rm dyn}$, Schneider, Gonçalves, Rollason *et al.* (1988) have proposed the following model: one represents the diffracted power as the sum of the perfect-crystal contribution (which does not depend on the thickness for values larger than Λ) and a term that corresponds to the scattering around the SiO_2 precipitates formed during annealing at 1043K. Therefore, they write

$$\begin{aligned} P &= P_{\rm dyn} + CA \\ P_{\rm dyn} &= P^c(E=1). \end{aligned} \tag{81}$$

This assumption has been recently confirmed by neutron small-angle scattering experiments by Messoloras, Schneider, Stewart & Zulehner (1988). The constant C is determined by the best fit to the observations.

The validity of the model is estimated *via* the reliability factor R and the goodness of fit, GOF (see Schneider, Gonçalves & Graf, 1988):

$$R = 100\left[\chi_R^2 \Big/ \sum_{i=1}^{N} (P_i^{\rm obs}/\sigma_i^{\rm obs})^2\right]^{1/2} \tag{V-99}$$

and

$$\mathrm{GOF} = \chi_R^2/(N-p)$$

where

$$\chi_R^2 = \sum_{i=1}^{N} |P_i^{\rm obs} - P_i^{\rm th}/\sigma_i^{\rm obs}|^2. \tag{V-100}$$

N is the number of observations, $P_i^{\rm obs}$ is the observed integrated reflecting power, $P_i^{\rm th}$ is the calculated integrated reflecting power, $\sigma_i^{\rm obs}$ is the standard deviation of $P_i^{\rm obs}$ and P is the number of parameters. For the example shown in Fig. 7, the result is

$$C = 4.28(2)\times 10^{-9}$$
$$R = 2.3\%, \qquad \mathrm{GOF} = 2.4.$$

Fig. 8 shows that (81) leads to a fair representation of the experimental results.

(2) The same data can be analysed using the theory developed in the present paper [as well as Kato's (1980) original approach].

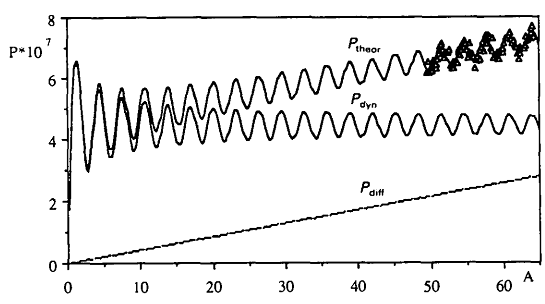


Fig. 8. Diffracted power for the 220 reflexion of Si (from Schneider, Conçalves, Rollason *et al.*, 1988). Same sample as Fig. 7. $P_{\rm theor}$ corresponds to equation (81), with $C = 4.28\times 10^{-9}$. $P_{\rm diff} = CA$.

A fit of the equations obtained in § III of this paper to the experimental data provides the following values:

$$E = 0.998, \qquad \tau/\Lambda = 0.01,$$
$$R = 2.19\% \qquad \mathrm{GOF} = 2.3.$$

Thus, τ/Λ is very small, and we may look at the limit of the theory when $\tau/\Lambda \to 0$, in which case M becomes very small. One obtains the asymptotic formula

$$\begin{aligned} P &\simeq P^c + P^{i,0} \\ &= QT[E^2W(2ET/\Lambda) + (1-E^2)]. \end{aligned} \tag{82}$$

The results are shown in Fig. 9 and are similar to those of Schneider, Conçalves, Rollason *et al.* (1988).

Finally, it is important to compare the present theory with Kato's original expression. In this particular case where $\tau/\Lambda \to 0$, $\tau_e \to E\Lambda$ and differs greatly from zero. The diffracted power is given by

$$\begin{aligned} P_K = Q[&TE^2W(2ET/\Lambda) + [(1-E^2)/2W] \\ &\times \sinh(2EA)\exp(-2EA)]. \end{aligned} \tag{83}$$

The best fit without constraint to the experimental data leads to $E = 0.367$, with $R = 5\%$ and GOF = 11.8. The results is shown in Fig. 10.

This approximation turns out to be poor. The reason for its failure is obviously the approximation made by Kato concerning the parameter τ_e.

If no limitations are made on τ/Λ or E, one can find several values of these parameters for which R

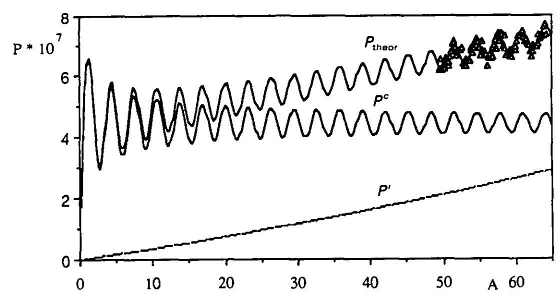


Fig. 9. Diffracted power for the 220 reflexion of Si (from the present theory). $E = 0.998$, $\tau/\Lambda = 0.01$.

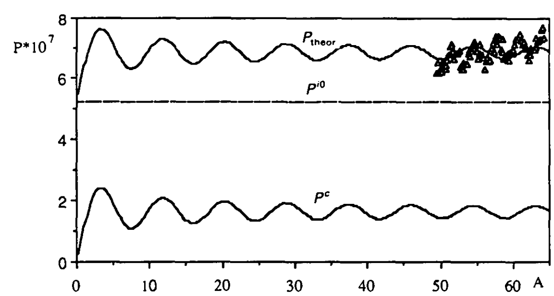


Fig. 10. Application of Kato's theory to the 220 reflexion of Si.

and GOF are nearly constant (Schneider, private communication).

IX. Concluding remarks

In this paper, we have presented a formulation of Kato's statistical dynamical theory which bypasses the approximations (33)-(35) concerning correlation lengths. It turns out that the hypothesis of a constant effective correlation length τ_e must be abandoned.

The resulting modifications to the theory are rather complex and lead to the definition of a variable correlation length, fluctuating around the value $\Gamma \simeq \tau$, which is defined by operation (52): this is an important alteration from Kato's original theory. The propagation equations for the incoherent intensities can be solved in the new scheme. We have presented the explicit solution for the diffracted integrated power for a parallel plate, with the application to annealed Si crystals (Czochralski grown). The present application is limited to crystals with a high degree of perfection (E close to 1). The theory needs to be tested on many examples, but a physical check of the significance of the parameters is only possible for highly perfect crystals. In a following paper we will present a solution that can be used in refinement procedures for finite crystals.

APPENDIX

A. Calculation of X

(a) We can write

$$x_1 = E^4 \int_0^{s_h} \mathrm{d}\eta \int_0^{s_0} \mathrm{d}\xi' \int_0^{s_h} \mathrm{d}\eta' \langle \delta D_0^*(\xi', \eta')\delta D_0(s_0, \eta)\rangle$$

$$= E^4 \int_0^{s_h} \mathrm{d}\eta \int_0^{s_0} \mathrm{d}\xi' \int_0^{s_h} \mathrm{d}\eta' \langle \delta D_0^*(\xi', \eta')\delta D_0(\xi', \eta)\rangle$$

owing to the long correlation of the amplitudes in the longitudinal direction [see Appendix of (I)]. Integration over η' leads to

$$x_1 = 2E^4 \int_0^{s_0} \mathrm{d}\xi \int_0^{s_h} \mathrm{d}\eta\, A(\xi, \eta) \qquad (A1)$$

and similiarly

$$x_1' = 2E^4 \int_0^{s_0} \mathrm{d}\xi \int_0^{s_h} \mathrm{d}\eta\, B(\xi, \eta).$$

Let us now consider the term x_2:

$$x_2 = E^4 \int_0^{s_h} \mathrm{d}\eta \int_0^{\eta} \mathrm{d}\eta' \int_0^{s_0} \mathrm{d}\xi' \times \langle \delta D_0^*(\xi', s_h)\delta D_0(\xi', \eta')\rangle$$

$$= E^4 \int_0^{s_0} \mathrm{d}\xi' \int_0^{s_h} \mathrm{d}\eta' [s_h - \eta'] \times \langle \delta D_0^*(\xi', s_h)\delta D_0(\xi', \eta')\rangle.$$

x_2 is thus of the order of $\int_0^{s_0} \Gamma A(\xi', s_h)\,\mathrm{d}\xi'$ where Γ is the width of the amplitude correlation function. If A is assumed to have small variations over the distance Γ, and if $\Gamma \ll s_h$, x_2 turns out to be very small compared to x_1. It is thus legitimate to neglect x_2 and x_2'.

(b) $$x_3 = E^2(1-E^2) \int_0^{s_h} \mathrm{d}\eta \int_0^{s_0} \mathrm{d}\xi' \int_0^{s_h} \mathrm{d}\eta'\, g(s_h - \eta) \times \langle \delta D_0^*(\xi', \eta')\delta D_0(\xi', \eta)\rangle$$

If we use the approximation (Becker & Al Haddad, 1989)

$$g(s_h - \eta') \simeq g(s_h - \eta) g(\eta - \eta') \quad \text{if } \eta < \eta',$$

$$x_3 = E^2(1-E^2)\tau \int_0^{s_0} \mathrm{d}\xi\, [A(\xi, s_h) + A'(\xi, s_h)]. \qquad (A2)$$

By a similar derivation,

$$x_4 = E^2(1-E^2) \int_0^{s_h} \mathrm{d}\eta \int_0^{\eta} \mathrm{d}\eta' \int_0^{s_0} \mathrm{d}\xi'\, g(\eta - \eta') \times \langle \delta D_0^*(\xi', s_h)\delta D_0(\xi', \eta')\rangle$$

$$\simeq E^2(1-E^2)\tau \int_0^{s_0} \mathrm{d}\xi\, [A(\xi, s_h) - A'(\xi, s_h)] \qquad (A3)$$

[exact if we assume $g(\xi) = \exp(-\xi/\tau)$].

As a result

$$x_3 + x_4 \simeq 2E^2(1-E^2)\tau \int_0^{s_0} \mathrm{d}\xi\, A(\xi, s_h). \qquad (A4)$$

We consider x_5:

$$x_5 = E^2(1-E^2) \int_0^{s_h} \mathrm{d}\eta \int_0^{\eta} \mathrm{d}\eta' \int_0^{s_0} \mathrm{d}\xi'\, g(s_0 - \xi') \times \langle D_0^*(\xi', s_h)\delta D_0(\xi', \eta')\rangle$$

$$\simeq E^2(1-E^2)\tau \int_0^{s_h} \mathrm{d}\eta' [s_h - \eta'] \times \langle \delta D_0^*(s_0, s_h)\delta D_0(s_0, \eta')\rangle$$

$$\simeq E^2(1-E^2)\tau\Gamma A \ll (x_3 + x_4).$$

We can then neglect x_5 and x_5'. Owing to this latter simplification, all the significant terms contributing to X correspond to an expansion of either A or B. As a consequence, we can write

$$E^2[B - A] = \chi^2 X. \qquad (A5)$$

Obviously,

$$x_3' + x_4' = 2E^2(1-E^2)\tau \int_0^{s_h} \mathrm{d}\eta\, B(s_0, \eta). \qquad (A6)$$

(c) Finally we calculate x_6 and x_6'.

$$x_6 = E^2(1-E^2)\int_0^{s_h} d\eta \int_0^{s_0} d\xi' \int_0^{s_h} d\eta' g(\eta-\eta') \times \langle D_0^*(\xi',\eta')D_0(\xi',\eta)\rangle$$

$$= 2E^2(1-E^2)\tau^2 \int_0^{s_h} I_0^c(s_0-\eta)\, d\eta + 2E^2(1-E^2)\tau \int_0^{s_h} d\eta\, A'(s_0,\eta) \qquad (A7)$$

$$x_6' = 2E^2(1-E^2)\tau^2 \int_0^{s_0} I_h^c(\xi, s_h)\, d\xi + 2E^2(1-E^2)\tau \int_0^{s_0} B'(\xi, s_h)\, d\xi.$$

In ($A7$), the contribution from the coherent source is apparent. We finally see that

$$\chi^{-2}\{B-A\} = 2E^2 \int_0^{s_0} d\xi \int_0^{s_h} d\eta\, [A-B](\xi,\eta) + 2(1-E^2)\tau\left\{\int_0^{s_0} d\xi\, A(\xi,s_h) - \int_0^{s_h} d\eta\, B(s_0,\eta)\right\} + 2(1-E^2)\tau\left\{\int_0^{s_h} d\eta\, A'(s_0,\eta) - \int_0^{s_0} d\xi\, B'(\xi,s_h)\right\} + 2(1-E^2)\tau^2\left\{\int_0^{s_h} d\eta\, I_0^c(\sigma_0,\eta) - \int_0^{s_0} d\xi\, I_h^c(\xi,s_h)\right\}. \qquad (A8)$$

B. Calculation of Y

We shall now proceed in a similar way for Y. From Table 2, it is apparent that all diagrams correspond to an expansion of either A' or B'. Thus:

$$(1-E^2)[B'-A'] = \chi^2 Y. \qquad (A9)$$

y_1 and y_2 can be calculated like x_3 and x_4 and it is shown that

$$y_1 + y_2 = x_3 + x_4 = 2E^2(1-E^2)\tau \int_0^{s_0} d\xi\, A(\xi, s_h).$$

Similarly

$$y_1' + y_2' = x_3' + x_4' = 2E^2(1-E^2)\tau \int_0^{s_h} d\eta\, B(s_0,\eta) \qquad (A10)$$

$$y_3 = 2E^2(1-E^2)\tau \int_0^{s_h} d\eta\, A(s_0,\eta)$$
$$y_3' = 2E^2(1-E^2)\tau \int_0^{s_0} d\xi\, B(\xi, s_h). \qquad (A11)$$

We now calculate y_4 and y_4'.

$$y_4 = (1-E^2)^2 \int_0^{s_0} d\xi' \int_0^{s_h} d\eta \int_0^{s_h} d\eta'\, g(s_h-\eta)g(s_h-\eta') \times \langle \delta D_0^*(\xi',\eta')\delta D_p(\xi',\eta)\rangle.$$

Again we make the assumption

$$g(s_h-\eta') \simeq g(s_h-\eta)g(\eta-\eta') \quad \text{for } \eta > \eta'$$

and we get

$$y_4 = 2(1-E^2)^2\tau_2 \int_0^{s_0} d\xi\, A'(\xi, s_h)$$

and

$$y_4' = 2(1-E^2)^2\tau_2 \int_0^{s_h} d\eta\, B'(s_0,\eta). \qquad (A12)$$

It has been shown in (I) that y_5 and y_5' are negligible. The argument, which was developed for $E \to 0$, holds for any value of E.

(a) We finally consider y_6 and y_6':

$$y_6 = 2(1-E^2)^2\tau\tau_2 \int_0^{s_h} d\eta\, I_0^c(s_0,\eta) + 2(1-E^2)^2\tau_2 \int_0^{s_h} d\eta\, A'(s_0,\eta)$$

$$y_6' = 2(1-E^2)^2\tau\tau_2 \int_0^{s_0} d\xi\, I_h^c(\xi, s_h) + 2(1-E^2)^2\tau_2 \int_0^{s_0} d\xi\, B'(\xi, s_h) \qquad (A13)$$

Equations ($A13$) are obtained in the same way as ($A7$).

(b) We summarize the previous results and obtain

$$\chi^{-2}\{B'-A'\} = 2E^2\tau \int_0^{s_0} d\xi\, [A-B](\xi,s_h) + 2E^2\tau \int_0^{s_h} d\eta\, [A-B](s_0,\eta) + 2(1-E^2)\tau_2 \int_0^{s_0} d\xi\, (A'-B')(\xi,s_h) + 2(1-E^2)\tau_2 \int_0^{s_h} d\eta\, (A'-B')(s_0,\eta) + 2(1-E^2)\tau\tau_2 \times \left\{\int_0^{s_h} d\eta\, I_0^c(s_0,\eta) - \int_0^{s_0} d\xi\, I_h^c(\xi,s_h)\right\}. \qquad (A14)$$

We thank D. Feil for a careful reading of the manuscript and for helpful discussions.

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Modern Equations of Diffractometry. Diffraction Geometry*

BY D. J. THOMAS†

Medical Research Council Laboratory of Molecular Biology, Hills Road, Cambridge CB2 2QH, England

(*Received 26 June 1990; accepted 29 July 1991*)

Abstract

The various geometries of area-detector diffractometers and cameras are best described using a coordinate-free abstract operator notation. Modern methods of geometry, including especially the combined application of vectors and covectors, are used; they confer the simultaneous advantages of simplifying, virtualizing and unifying the analysis, which becomes applicable to all methods and machines. A second, and most valuable, prize arising from this approach, itself a major theme of this paper, is the complete avoidance of computationally expensive and analytically inconvenient trigonometric functions in area diffractometry. The very few occasions when they are unavoidable have already been discussed fully in a previous paper on goniometry. Basic diffraction geometry is presented first, giving all the equations necessary to identify diffraction spots and to calculate a useful generalization of the Lorentz factor. These are a formalized and extended version of those presented to the EEC Cooperative Workshop on Position-Sensitive Detector Software held at LURE in Paris in 1986. Then, various previously unpublished formulae describing beam divergence, dispersion and polarization, crystal mosaicity and angular widths of diffraction spots are presented. Finally, three specific calculations appropriate to the use of an area diffractometer are given, including a calculation of window sizes, a model of the backstop shadow and a method of surveying a diffraction pattern for assessment and prealignment.

1. Introduction – unification through generalization

From the earliest days of crystallographic diffraction studies, the analysis of diffraction geometry has been heavily reliant on the use of trigonometric functions and of radical forms, particularly the square root. This was because, at a time when electronic computers were not available, roots and trigonometric functions could conveniently be read from tables, whereas equivalent vectorial (*i.e.* matrix) calculations would have been intolerably tedious. With the advent of digital computers, particularly in demanding 'real-time' applications, radical and trigonometric calculations became relatively less favourable when compared with component calculations using vectors and matrices, which are the natural variables for 'area detectors'.‡ Although cameras existed and were in common use, they were not perceived as area detectors until their electronic successors appeared. Thus, it did not become apparent until fairly recently that any theory of area diffractometry based on vectorial calculations could exist in contradistinction to that of single-counter diffractometry, where the use of angular variables is entirely natural.

It was not until 1986 at the EEC Cooperative Workshop on Position-Sensitive Detectors in Paris that it became apparent that the simple vectorial equations long used in the Cambridge software package for the Enraf–Nonius FAST system were not, in fact, common knowledge. I was thus encouraged to make them more widely known, and hope that this paper achieves that. At the same workshop, Dr Gérard Bricogne used the term 'virtualized'

* This paper is a sequel to Modern Equations of Diffractometry. Goniometry [*Acta Cryst.* (1990), **A46**, 321–343] in which it is referred to as Thomas (1990*b*). The second author with M. R. Hestenes in the reference list to that paper (p. 342) should have been E. Stiefel.

† Present address: European Molecular Biology Laboratory, Meyerhofstrasse 1, Postfach 10.2209, W-6900 Heidelberg, Germany.

‡ The misnomer 'area detector' (*cf.* 'linear detector', 'single counter') is the accepted name for a 2D imaging detector for recording diffraction patterns. It usually also bears the connotation of a reusable electronically readable device, which in some way justifies the need for a special name; in the present paper the term has a more general meaning and is held to include any detector capable of measuring a 2D image, including film.

0108-7673/92/020134-25$03.00