$F_{\mathbf{h}} > \sigma_1$] and the 'orthogonality' of the Bessel functions to produce a Fourier–Bessel series for the noncentric reflections. Thus, the probability can be expressed as

$$P(I_{\mathbf{h}}) = \sum_{m=1}^{\infty} a_m J_0(j_m I_{\mathbf{h}}^{1/2}/\sigma_1), \qquad (A9)$$

where j_m is the mth zero of J_0. The values of a_m can be determined using the orthogonality condition (Watson, 1942; see also Gradshteyn & Ryzhik, 1965, 6.521)

$$(1/2)\sigma_1^2[J_1(j_m)]^2 \int_0^{\sigma_1} J_0(Fx)J_0(Fj_m/\sigma_1)F\,dF = \delta(j_m - x\sigma_1), \qquad (A10)$$

which gives

$$a_m = \sigma_1^{-2}[J_1(j_m)]^{-2} \prod_{\mu=1}^{N/\varepsilon} J_0(\varepsilon f_\mu j_m/\sigma_1) = \sigma_1^{-2}[J_1(j_m)]^{-2}\exp - (\varepsilon\sigma_2/4\sigma_1^2)j_m^2 \times \sum_{n=0}^{\infty} K_n^1(\varepsilon j_m/2\sigma_1)^{2n}. \qquad (A11)$$

This series expansion is used to calculate the exact distributions for comparison in Figs. 1 and 4. At least 40 terms were used in any expansion.

These series have importance when the central-limit-theorem approximation (*i.e.* the Wilson term) is inapplicable. For example, if the cell is considered to be made up of a small number of fragments with structure factors $|\mathbf{F}_{\mathbf{h}}^{\mu}| = f_\mu(\mathbf{h}, \theta_\mu)$, then the expansions (3) still hold for random fragments but the higher-order terms have more significance. Thus, a joint probability distribution of fragment orientations θ_μ can be calculated.

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Mathematical Structure of the Coherent Wave Field in the Statistical Theory of Dynamical Diffraction

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Abstract

The coherent wave field, which is the ensemble average of the solution of the wave equation, is studied. The approach is similar to that used in the previous theory on extinction [Kato (1976). *Acta Cryst.* A**32**, 453–457, 458–466]. Here it is extended to deal with general cases where the single average and the

second-order correlation of lattice phase factors are mixed. The Laplace transforms of the coherent wave fields are derived first and integro-differential equations (IDEs) are formulated for them. The latter are identical to the previous ones derived directly from the wave equation. A controversial problem of IDEs is explained by the interpretation of IDEs.

1. Introduction

The present author has studied statistical treatments of dynamical diffraction in distorted crystals for more than a decade. The underlying motivations for this work are described in previous papers and summarized in the introduction to the latest one (Kato, 1991); they are not repeated here.

This paper is intended to elucidate the mathematical structure of the coherent wave field (CWF), which is the ensemble average of the wave field. First, we adopt a formal solution of the wave equation of the Takagi–Taupin (T–T) type. Then, the average is taken over the lattice phase factors associated with diffraction (kink) points of every optical path. Correlations higher than second order are neglected. Also, only the case of Laue geometry is considered. Within these limitations, no approximations are assumed.

The method is similar to that used in the theory on extinction (Kato, 1976*a*,*b*, hereinafter referred to as I*a* and I*b*, respectively). In fact, that work is the forerunner of the present theory. It is, here, however, generalized so as to deal with cases in which the average and the second-order correlations of phase factors are mixed. In such general cases, the Laplace transforms of CWFs are first derived and a couple of integro-differential equations (IDEs) are formulated for them.

The present IDE is identical to that obtained in the previous papers (Kato, 1980*a*,*b*, hereinafter referred to as II*a* and II*b*, respectively). Also, the result for the Laplace transforms is similar to that recently obtained by Guigay & Chukhovskii (1992). Their approach, however, is different from the present one.

As a preparation, the present approach and some notation are explained in §2. The rest of this paper is devoted to deriving CWFs step by step and arriving at IDEs. In these sections, the standard mathematics related to the Laplace transform (for example, Sneddon, 1972) are used extensively. In §6, the applicability of the present theory and a few different opinions from Guigay & Chukhovskii on the interpretation of IDEs are discussed.

2. The outline of the theory and a glossary of notation

Only important notations are listed, with a few comments, as details have been given in previous papers. [I*a*, I*b*, II*a*, II*b* and Kato (1991)].

Diffraction amplitudes: κ_g and κ_{-g}, $\kappa^2 = \kappa_g \kappa_{-g}$. (*P*1*a*, *b*)

Lattice phase: $\Phi(\mathbf{r}) = \exp\{-2\pi i[\mathbf{g}\cdot\mathbf{u}(\mathbf{r})]\}$. (*P*2)

The position $\mathbf{r}$ is often denoted by the components (s_o, s_g) in the oblique coordinate axes along the O and G directions.

Static Debye–Waller factor: $E = \langle\Phi(\mathbf{r})\rangle$. (*P*3)

Correlation function: $\langle\Phi\Phi^*(z)\rangle = \langle\Phi^*\Phi(z)\rangle$ (*P*4*a*)

$$= E^2 + (1 - E^2)g(z). \quad (P4b)$$

The distance z represents either z_o or z_g, which are measured along the O and G directions, respectively. $g(z)$ is called the intrinsic correlation function.

Let us consider the formal solution of the wave fields, D_o and D_g. As discussed in I*a*, I*b*, they can be written in the forms

$$D_o(s_o, s_g) = \delta(s_g) + \sum_{r(1,\infty)} \sum_{Rr} (i\kappa_g)\Phi_1(i\kappa_{-g}) \times \Phi_2 \times \ldots \times (i\kappa_{-g})\Phi_{2r}, \quad (P5a)$$

$$D_o(s_o, s_g) = \sum_{r(0,\infty)} \sum_{Rr} (i\kappa_g)\Phi_1(i\kappa_{-g})\Phi_2 \ldots \Phi_{2r}(i\kappa_g)\Phi_{2r+1}, \quad (P5b)$$

where the suffix to Φ indicates the order of kink points in a zig-zag optical path and, symbolically, the position (s_{oi}, s_{gi}) of each kink. The symbol $\sum_{Rr}$ implies the total sum (integration) over possible paths, which are specified by all kink positions $\{s_{oi}, s_{gi}\}$. This is further explained in §§3 and 4.

The first term $\delta(s_g)$ in (*P*5*a*) is the O wave, which experiences no diffraction. The singular form is due to the assumption of a point source located at (0,0) and the characteristic nature of the hyperbolic differential equation of the T–T type.

Next, we consider CWFs, namely $\langle D_o(s_o, s_g)\rangle$ and $\langle D_g(s_o, s_g)\rangle$. Notice that only the lattice phase factors are random variables. If correlations higher than second order are neglected, the sequence of the averaged phase factors consists of E [*cf.* (*P*3)] and $[E^2 + (1 - E^2)g(z)]$ [*cf.* (*P*4*b*)]. However, the first term, E^2, in the latter is nothing other than an independent average of the phase factors at two neighboring kinks. Therefore, after averaging, the sequence consists of E and $(1 - E^2)g(z)$. The situation is illustrated in Fig. 1. The shadowed region is a sequence of $(1 - E^2)g(z)$ and the narrow white column indicates the factor E.

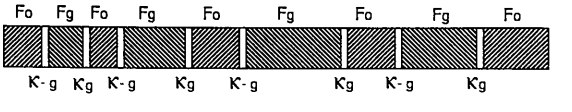

Fig. 1. A schematic diagram of the sequence of the averaged phase factors in the case of $\langle D_o(s_o, s_g)\rangle$ (see §2).

For fitting to the boundary condition of the wave, it is postulated that the first shadowed region consists of $(1-E^2)g(z_g)$. That is, only the intrinsic correlation along the G direction is included. Such a region is called an O-type region. Notice that the input and output waves propagate in the O direction. Then, the amplitude factor of the subsequent white column must be $(i\kappa_g)$. Consequently, the next shadowed region is of the G type, consisting of only $(1-E^2)g(z_o)$. Then, the next white column must be characterized by the amplitude factor $(i\kappa_{-g})$. The subsequent region must be of the O type and so on. Obviously, the final region must be of the O type for the O wave and of the G type for the G wave. In summary, the shadowed regions of the O and G types appear alternately and the amplitude factors $(i\kappa_g)$ and $(i\kappa_{-g})$ appear in the same manner, with a fixed order.

If the number of the intrinsic correlation functions includes zero in the shadowed regions, a continuous arrangement of E is automatically taken into account. Moreover, any mixed arrangement of $(i\kappa_g E)$, $(i\kappa_{-g}E)$ and $\kappa^2(1-E^2)g(z)$ that obeys the diffraction condition can be represented with this notation scheme.

In the following sections, we calculate CWFs step by step. Except in the titles and unless ambiguity occurs, the adjective 'averaged' and the notation $\langle\,\rangle$ are omitted.

3. The special averaged wave field, $F_o(\xi,\eta)$ and $F_g(\xi,\eta)$

In Fig. 2, the shadowed regions in Fig. 1 are shown in more detail. The size of the region is specified by ξ and η. The wave fields corresponding to the O- and G-type regions are denoted $F_o(\xi,\eta)$ and $F_g(\xi,\eta)$, respectively. We discuss mainly $F_o(\xi,\eta)$ because the field $F_g(\xi,\eta)$ is obtained in the same way.

According to the present notation scheme, $F_o(\xi,\eta)$ can be expressed in the form

$$F_o(\xi,\eta) = \sum_{k(0,\infty)} S_{\{z_o\}}S_{\{z_g\}}[-\kappa^2(1\text{-}E^2)] \times g(z_{g1})[-\kappa^2(1-E^2)]g(z_{g2}) \times \ldots \times [-\kappa^2(1-E^2)]g(z_{gk}) \qquad (1)$$

where z_{gi} is the distance of ith kink pairs along the G direction. Their distances along the O direction are denoted $\{z_{oi}\}$, which, however, are hidden in this expression.

The notation $S_{\{z_o\}}$ is an integral operator of possible $\{z_{oi}\}$ under the geometrical constraint,

$$\xi = z_{o1} + z_{o2} + \ldots + z_{o,k+1}. \qquad (2a)$$

Similarly, $S_{\{zg\}}$ is an operator of possible $\{z_{gi}\}$ under the constraint

$$\eta = z_{g1} + z_{g2} + \ldots + z_{g,k}. \qquad (2b)$$

They can be written more explicitly as:

$$S_{\{z_o\}} = \int_0^\infty \ldots \int_0^\infty \delta(\xi - \textstyle\sum z_{oi})\mathrm{d}z_{o1}\mathrm{d}z_{o2}\ldots\mathrm{d}z_{o,k+1}, \qquad (3a)$$

$$S_{\{z_g\}} = \int_0^\infty \ldots \int_0^\infty \delta(\eta - \textstyle\sum z_{gi})\mathrm{d}z_{g1}\mathrm{d}z_{g2}\ldots\mathrm{d}z_{g,k}. \qquad (3b)$$

Here, we use the standard expression for the δ function:

$$\delta(x) = (2\pi i)^{-1}\int_{c-i\infty}^{c+i\infty} \exp(px)\mathrm{d}p, \qquad (4)$$

in terms of the inverse Laplace transform, where c is any positive real number and is called the radius of convergence.

If the integrations of (3a) and (3b) are applied to (1), one obtains the result:

$$F_o(\xi,\eta) = (2\pi i)^{-2}\iint \exp(p\xi + q\eta)\mathrm{d}p\mathrm{d}q \times \sum_k (1/p)^{k+1}[-\kappa^2(1-E^2)g(q)]^k$$

where $g(q)$ is the Laplace transform of $g(z_{gi})$, common for all i. The limits of integration are similar to those of (4), but are omitted for convenience. This convention is used frequently hereinafter.

The sum $\sum_k$ is just a geometrical series, so that we have

$$F_o(\xi,\eta) = (2\pi i)^{-2}\iint \exp(p\xi + q\eta)\mathrm{d}p\mathrm{d}q \times [p + (1-E^2)\kappa^2 g(q)]^{-1}. \qquad (5a)$$

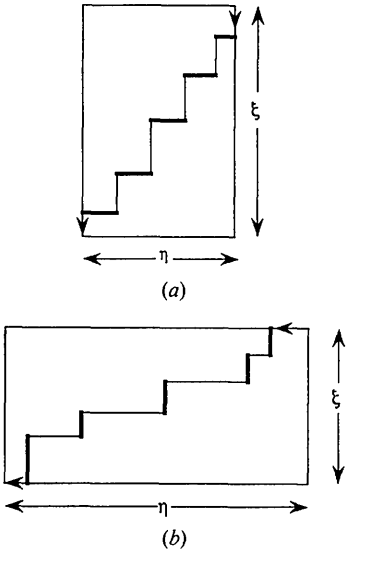

Fig. 2. The optical path and the intrinsic correlations (thick lines) in the special fields. (a) $F_o(\xi,\eta)$, (b) $F_g(\xi,\eta)$.

This implies that the Laplace transform of $F_o(\xi,\eta)$ is

$$F_o(p,q) = [p + (1 - E^2)\kappa^2 g(q)]^{-1}. \quad (5b)$$

In the same manner, one can calculate the special field $F_g(\xi,\eta)$. The result is

$$F_g(\xi,\eta) = (2\pi i)^{-2}\iint \exp(p\xi + q\eta)F_g(p,q)\mathrm{d}p\mathrm{d}q, \quad (6a)$$

where

$$F_g(p,q) = [q + (1 - E^2)\kappa^2 g(p)]^{-1}. \quad (6b)$$

Obviously, $F_g(p,q)$ can be derived from $F_o(p,q)$ and *vice versa*, simply by exchanging the variables p and q.

4. The CWFs, $\langle D_o(s_o,s_g)\rangle$ and $\langle D_g(s_o,s_g)\rangle$ in general cases

As explained in §2, the CWF can be constructed by taking an alternate product of F_o and F_g and inserting the amplitude factor $(i\kappa_g E)$ between F_o and F_g and, also, the factor $(i\kappa_{-g}E)$ between F_g and F_o. The wave field $D_o(s_o,s_g)$ is the total sum of such a product. Therefore, one obtains the expression

$$D_o(s_o,s_g) = \sum_{f(0,\infty)} S_{\{\xi,\eta\}}F_o(\xi_1,\eta_1)(i\kappa_g E)F_g(\xi_2,\eta_2) \times (i\kappa_{-g}E)\times\ldots\times(i\kappa_{-g}E)F_o(\xi_{2f+1},\eta_{2f+1}), \quad (7)$$

where $S_{\{\xi_i\eta_i\}}$ implies multiple integration with respect to possible variables of $\{\xi_i,\eta_i\}$ under the constraints

$$s_o = \xi_1 + \xi_2 + \ldots + \xi_{2f+1}, \quad (8a)$$

$$s_g = \eta_1 + \eta_2 + \ldots + \eta_{2f+1}. \quad (8b)$$

By using a similar technique to that used for obtaining (5), we have

$$D_o(s_o,s_g) = (2\pi i)^{-2}\iint \exp(ps_o + qs_g)\mathrm{d}p\mathrm{d}q \times \sum_f (-\kappa^2E^2)^f[F_o(p,q)F_g(p,q)]^f F_o(p,q) \quad (9a)$$

$$= (2\pi\mathrm{i})^{-2}\iint \exp(ps_o + qs_g)\mathrm{d}p\mathrm{d}qF_o(p,q) \times [1 + \kappa^2E^2F_o(p,q)F_g(p,q)]^{-1}. \quad (9b)$$

The wave field $D_g(s_o,s_g)$ can be written in a similar manner to (7). Necessary modifications are only to multiply the right-hand side of (7) by the factor

$$(i\kappa_g E)F_g(\xi_{2f+2},\eta_{2f+2})$$

and to add ξ_{2f+2} and η_{2f+2} to the constraints (8a) and (8b), respectively. Thus, we have

$$D_g(s_o,s_g) = (2\pi i)^{-2}\iint \exp(ps_o + qs_g)\mathrm{d}p\mathrm{d}q \times (i\kappa_g E)F_o(p,q)F_g(p,q) \times [1 + (\kappa E)^2F_o(p,q)F_g(p,q)]^{-1}. \quad (10)$$

In summary, the Laplace transforms of $D_o(s_o,s_g)$ and $D_g(s_o,s_g)$ are given in the forms

$$D_o(p,q) = F_o(p,q)D(p,q), \quad (11a)$$

$$D_g(p,q) = (i\kappa_g E)F_o(p,q)F_g(p,q)D(p,q) = (i\kappa_g E)F_g(p,q)D_o(p,q), \quad (11b)$$

where

$$D(p,q) = [1 + (\kappa E)^2F_o(p,q)F_g(p,q)]^{-1}. \quad (12)$$

5. Relations between $\langle D_o(s_o,s_g)\rangle$ and $\langle D_g(s_o,s_g)\rangle$

First, we consider relations amongst the Laplace transforms. For this purpose, we introduce the notation

$$\overline{D}_o(p,q) = pD_o(p,q), \quad (13a)$$

$$\overline{D}_g(p,q) = qD_g(p,q), \quad (13b)$$

which are the Laplace transforms of $\partial D_o(s_o,s_g)/\partial s_o$ and $\partial D_g(s_o,s_g)/\partial s_g$, respectively, under the boundary conditions $D_o(-\varepsilon,s_g) = 0$ and $D_g(s_o,-\varepsilon) = 0$.

From the identity relation [*cf.* (5b)],

$$[p + (1 - E^2)\kappa^2 g(q)]F_o(p,q) = 1,$$

it follows that

$$pF_o(p,q) = 1 - (1 - E^2)\kappa^2 g(q)F_o(p,q).$$

Therefore, multiplying $D(p,q)$ by this, we have the relation from (11a):

$$\overline{D}_o(p,q) = D(p,q) - (1 - E^2)\kappa^2 g(q)F_o(p,q)D(p,q). \quad (14)$$

On the other hand, it follows from (11b) that

$$(i\kappa_{-g}E)D_g(p,q) = -(\kappa E)^2F_o(p,q)F_g(p,q)D(p,q) = D(p,q) - 1. \quad (15)$$

Combining (14) and (15), we finally have

$$\overline{D}_o(p,q) = (i\kappa_{-g}E)D_g(p,q) - (1 - E^2)\kappa^2 g(q)D_o(p,q) + 1. \quad (16a)$$

The same procedure can be used for obtaining the expression $\overline{D}_g(p,q)$. We start with the identity [*cf.* (6b)]

$$[q + (1 - E^2)\kappa^2 g(p)]F_g(p,q) = 1.$$

After multiplication of $(i\kappa_g E)F_oD$, this gives the relation

$$\overline{D}_g(p,q) = (i\kappa_g E)[1 - (1 - E^2)\kappa^2 g(p)F_g(p,q)] \times F_o(p,q)D(p,q) = (i\kappa_g E)D_o(p,q) - (1 - E^2)\kappa^2 g(p)D_g(p,q). \quad (16b)$$

The inverse Laplace transforms of (16a) and (16b) immediately give the following relations between

$D_o(s_o,s_g)$ and $D_g(s_o,s_g)$:

$$\partial D_o(s_o,s_g)/\partial s_o = (i\kappa_{-g}E)D_g(s_o,s_g) - (1-E^2)\kappa^2 \times \int_0^{s_g} D_o(s_o,s_g-z_g)g(z_g)\mathrm{d}z_g + \delta(s_o)\delta(s_g), \quad (17a)$$

$$\partial D_g(s_o,s_g)/\partial s_g = (i\kappa_g E)D_o(s_o,s_g) - (1-E^2)\kappa^2 \times \int_0^{s_o} D_g(s_o-z_o,s_g)g(z_o)\mathrm{d}z_o. \quad (17b)$$

The integrals in the right-hand sides result from the convolution theorem of the Laplace transform.

Because we are interested in the area of $s_o > \tau$ and $s_g > \tau$ for the fundamental relations of CWFs, the last term of (17*a*) can be dropped. In this sense, (17) is identical (10) of II*a*. This is further discussed in §6.

Until now, everything has been exact. Assuming a gentle variation of $D_o(s_o,s_g)$ and $D_g(s_o,s_g)$ over the correlation length τ of $g(z)$, one can obtain straightforward but approximate formulae

$$\partial D_o(s_o,s_g)/\partial s_o = (i\kappa_{-g}E)D_g(s_o,s_g) - (1-E^2)\kappa^2\tau D_o(s_o,s_g), \quad (18a)$$

$$\partial D_o(s_o,s_g)/\partial s_g = (i\kappa_g E)D_o(s_o,s_g) - (1-E^2)\kappa^2\tau D_g(s_o,s_g). \quad (18b)$$

The approximations used here are equivalent to putting $g(p) = g(q) \simeq g(0) = \tau$ in (16*a*) and (16*b*).

6. Discussion

6.1. *Applicability of the present theory*

(*a*) *Absorbing crystals.* The present results for the Laplace transforms of CWFs [*cf.* (11) and (12)] and IDEs [*cf.* (17) and (18)] may be used for absorbing crystals. As explained in I*a*, the normal absorption is taken into account simply by multiplying the factor

$$\exp[-\mu_o(s_o+s_g)]$$

by the intensities derived from CWFs, where μ_o is the linear absorption coefficient.

The Borrmann absorption can be represented by introducing the imaginary part in the expression κ^2 [equation (*P*1*b*)]. Through the treatments of the Laplace transform,

$$F(s) = \int_0^{\infty} f(t)\exp(-st)\mathrm{d}t,$$

and the inverse transform,

$$f(t) = (2\pi i)^{-1}\int_{c-i\infty}^{c+i\infty} F(s)\exp(st)\mathrm{d}s,$$

only the variable t is assumed to be real – the variable s and the functions $f(t)$ and $F(s)$ are not necessarily real. Therefore, the present results need not be modified for absorbing crystals.

(*b*) *Asymmetric geometry.* As in the spherical-wave theory for perfect crystals, the starting expression (*P*5) is valid for asymmetric cases, provided that the coordinate axes are taken along the *O* and *G* directions. Also, the statistical quantities such as E [*cf.* (*P*3)] and $g(z)$ [*cf.* (*P*4)] are independent of the absolute position at which the average is taken. Therefore, all arguments following §3 remain valid without any modification.

(*c*) *The spatial range over which* (17) *is applicable.* The original equation (1) is correct as a solution of the T–T equation. However, the averaged (17) must be understood as a nonlocal relation over a range where the average is meaningful; *i.e.* $s_o, s_g \simeq > \tau$. It is too stringent to take the limitation $s_o, s_g >> \tau$ as assumed by Guigay & Chukhovskii (1992).

6.2. *The interpretation of* (17)

There is a difference between the IDE of Guigay & Chukhoskii (1992) (G&C) and the old formula proposed by the present author. Their equation [the first of (11)] includes $i\kappa_g\delta(s_g)$ in the present notation but the old formula [(10*a*) in II*a*] has no such term. On the other hand, (17*a*) of the present paper, which we shall call the new formula, includes $\delta(s_o)\delta(s_g)$. These differences are a subtle problem depending on the interpretation of IDEs.

Fundamental relations like the IDEs considered here are used in two ways. One usage is to represent the *relation* among the wave fields, which must be free from the boundary condition, in particular the incident wave. The other is to represent a *solution* to the original equation, which is usually inhomogeneous because the boundary condition owing to the incident wave is included. The old formula corresponds to the former usage and the new formula corresponds to the latter. This situation is similar to the difference between the Schrödinger equation, $\mathbf{H}\Psi = 0$, and the equation for the Green function, $\mathbf{H}G = \delta(\mathbf{r})$, $\mathbf{H}$ being the same Hamiltonian. With this interpretation, both the new and the old formulae are correct, if one uses them legitimately. Obviously, when equation (10) of II*a* is used, the proper boundary conditions such as equations (33*a*) and (34*a*) in II*a* must be employed.

For the same reason, the physical content of equation (10) of G&C for G_{od} is no different from the old formula of the present author with the boundary conditions mentioned above. Although they stress the importance of the difference, the choice would be merely a matter of convenience.

The following difference seems more serious. The term $i\kappa_g\delta(s_g)$ in equation (11) of G&C is hardly understood. It must be $\delta(s_o)\delta(s_g)$ as in the present new formula. Otherwise, their two-dimensional

Laplace transform [the unity on the right-hand side of the first equation of (14)] cannot be derived straightforwardly. Nevertheless, because their equation (14) is correct for some reason, the author believes that the rest of their calculation is useful. In fact, their equation (15) is the same as the present result [(16*a*) and (16*b*)] if one replaces $\{\tau/(1+\tau p_{o,h})\}$ with the general expression $g(p_{o,h})$ after putting $p_o = p$ and $p_h = q$.

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The Joint Probability Distribution of Any Set of Phases Given Any Set of Diffraction Magnitudes. III. A Function to Maximize

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Abstract

The joint probability distribution method described in paper I [Giacovazzo, Burla & Cascarano (1992). *Acta Cryst.* A**48**, 901–906] and paper II [Burla, Cascarano & Giacovazzo (1992). *Acta Cryst.* A**48**, 906–912] of this series has been considered in order to obtain a function that is maximized by the true crystal structure. The phasing process is carried out by maximizing such a function *via* a modified tangent refinement: this implies the active use of negative triplet and quartet relationships. The major effects provoked in a direct procedure by the use of numerous phase relationships with expected negative cosines are analysed. Practical applications are also described.

Symbols and notation

We adopt the symbols and notation used in papers I and II of the series (Giacovazzo, Burla & Cascarano, 1992; Burla, Cascarano & Giacovazzo, 1992).

Introduction

In papers I and II, the conditional joint probability distribution of n phases given p ($p \geq n$) moduli was studied. The calculations were performed in order to allow for large values of n and p: *i.e.* n should be the number of strong reflections to be phased by a standard direct procedure and p may be the number of measured reflections. Two expressions were derived, both including triplet and quartet invariant contributions: the first formula [see equation (1) of paper II] may be considered as a development of Hauptman's mathematical approach, the second [see equation (2) of paper II] of Giacovazzo's approach. Both expressions were checked to assess their theoretical soundness and practical usefulness for phase solution. The first one was found to present unacceptable features for a well behaved distribution (in agreement with some recent results obtained by Altomare, Burla, Cascarano, Giacovazzo & Guagliardi, 1993). The second distribution function, even though better designed, is not maximized by the correct set of phases (as one would expect for sufficiently large values of n and p). Accordingly, the combined use of triplet and quartet invariants in the tangent procedure proved to be of limited usefulness. This was ascribed to the limited accuracy of the probabilistic estimates of quartet invariants and, therefore, to some insufficiency in the mathematics used by Hauptman and by Giacovazzo. We show in this paper that a modified expression for the distribution (2) of paper II is frequently maximized by the correct solution. As a practical consequence, the