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A Green Function for Takagi's Equation of High-Energy-Electron Diffraction

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An integral formulation of Takagi's equation of high-energy-electron diffraction from an imperfect crystal is presented. In particular, the Green function that represents diffraction from a mathematical point defect is investigated. In the two-beam case this function consists of two parts: (i) a singular part propagating down the characteristics, and (ii) a finite oscillatory part propagating between the characteristics. It is conjectured that this general behavior remains true in the n-beam case.

1. Introduction

A system of differential equations first derived by Takagi (1962, 1969) in a phenomenological manner has been shown by Lewis, Hammond & Villagrana (1975) to be the rigorous consequence of certain well defined approximations concerning the localization of intensity in the diffraction pattern. In the latter work they derived an equivalent integral equation for this system, and hence were able to construct a perturbation solution (Born series). However, this integral equation incorporated all of the effects of the potential into a single term, and as a consequence of this, the zeroth-order scattering amplitude for a diffracted wave was the amplitude at the entrance surface of the crystal. Because this amplitude bears little relation to the amplitude a few hundred ångstroms from the entrance surface of the crystal, the convergence of this series would be very slow. This is especially true for a thick crystal, even if the potential is nearly perfectly periodic. As a result, this expansion would not be useful for a practical computation of diffraction unless the crystal was extremely thin ($\ll$ extinction distance).

It is the purpose of this work to remedy this defect of the expansion by splitting the potential into two parts: (i) a perfectly periodic part which describes the underlying reference crystal, and (ii) the remainder which describes the deviation from periodicity due to imperfections in the crystal. One then arrives at a new integral equation whose zeroth-order solution will be the solution to the perfect-crystal equations for a crystal of arbitrary thickness; thus, the convergence of the associated Born series would be expected to be very fast for a nearly perfect crystal. We shall pay particular attention to the Green function that is used to construct this integral equation; and as an illustrative example, we will construct an explicit Green function for two-beam diffraction from a mathematical point defect.

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2. General considerations

We will consider Laue diffraction from a two-dimensional crystal, where x and z are coordinates parallel and perpendicular to the entrance surface of the crystal. (This coordinate restriction does not alter the physics and the generalization to three dimensions is straightforward.) The system of equations we are considering is

$$\hat{L}D(x,z)\equiv\left(I\frac{\partial}{\partial z}+B\frac{\partial}{\partial x}\right)D(x,z)=iA(x,z)D(x,z) \quad (1)$$

where $D(x,z)$ is a column vector containing the amplitudes $d_g(x,z)$ of the transmitted and diffracted waves, $\mathbf{g}$ is a reciprocal-lattice vector, I is the identity matrix, and B is a diagonal matrix containing the tangents of the Bragg angles θ_g of the diffracted beams. Hence, the operator $\hat{L}$ is a diagonal matrix of directional derivatives along the various diffraction directions. The matrix $A(x,z)$ contains the scattering potential of the crystal in the following way: let $V(\mathbf{r})$ be the potential which appears in the Schrödinger equation: then we have the unique expansion $V(\mathbf{r})=\sum_g \tilde{v}_g(\mathbf{r})\exp(i\mathbf{g}\cdot\mathbf{r})$, where the Fourier transform of each $\tilde{v}_g(\mathbf{r})$ has support in the first Brillouin zone. The $A(x,z)$ matrix has elements

$$A_{gh}=\begin{cases}(k_0^2-k_g^2)/2k_{gz} & \text{for } g=h\\ [\hbar^2\tilde{v}_{g-h}(x,z)]/4mk_{gz} & \text{for } g\neq h\,,\end{cases} \quad (2)$$

where $\mathbf{k}_0$ is the wavevector of the transmitted beam, $\mathbf{k}_g\equiv\mathbf{k}_0+\mathbf{g}$, and m is the relativistically corrected electron mass. Equation (1) is an approximation to the Schrödinger equation, derived under the assumption that the Fourier transforms of the components of $D(x,z)$ are well localized in the first Brillouin zone.

When this is the case these components may be interpreted as the amplitudes of the diffracted waves; physically, this approximation corresponds to negligible diffuse scattering between the observed Bragg maxima and must be questioned in situations where such scattering is not negligible.

In a perfect crystal $A(x,z)$ becomes a constant matrix A_0, since $V(\mathbf{r})$ has a Fourier series. If the boundary conditions are that of a plane wave [*i.e.* $D(x,z=0)=D(0)$] independent of x, then equation (1) has the perfect-crystal solution $D_0(z)=\exp(iA_0z)D(0)$. It should be stressed that there is a solution, independent of x, to equation (1) only when $A(x,z)$ is a constant and $D(0)$ is independent of x. Otherwise, the solution is more complicated and not of this simple form.

As discussed by Lewis, Hammond & Villagrana (1975), the retarded Green function for the operator $\hat{L}$ is given by

$$G^I_{gh}(x-x';\, z-z')=\theta(z-z')\delta_{gh} \times\delta[(x-x')-(z-z')\tan\theta g] \quad (3)$$

where $\theta(z-z')$ is the unit step function, δ_{gh} is the Kronecker delta function, and $\delta[\cdots]$ is the Dirac delta function. This Green function, although of very simple structure due to the simplicity of $\hat{L}$, reveals two important facts about hyperbolic operators such as $\hat{L}$: (i) there exists a region of determinancy for the solution of equation (1) at a point P bounded by the characteristics with the greatest and least slopes passing through P, and (ii) hyperbolic operators tend to propagate singularities down their characteristics.

Now, in order to transform equation (1) into an integral equation of the form described in the introduction, it is only necessary to calculate a new (and more informative) Green function defined by the retarded solution to

$$(\hat{L}-iA_0)\Delta(x-x';\, z-z')=\delta(x-x')\delta(z-z')I\,. \quad (4)$$

Not only does this Green function enable us to apply perturbation theory to the solutions to the perfect crystal equations, but it also solves the problem of scattering by a crystal when the incident wave is not a plane wave but an arbitrary wave.

If the incident wave is a plane wave, we have the integral equation

$$D(x,z)=D_0(z)+\int_0^z\int_{-\infty}^{\infty}\Delta(x-x';\, z-z') \times\delta A(x',z')D(x',z')\mathrm{d}x'\mathrm{d}z' \quad (5)$$

where we have written $A(x,z)=A_0+\delta A(x,z)$. It is obvious that equation (5) is equivalent to equation (1) and now we have $D_0(z)$ as the zeroth-order solution. The Born series to this equation is obtained in the usual way by iterating the integral, beginning with $D_0(z)$.

If the incident wave is not a plane wave, but rather a more general wave, so that $D(x,z=0)$ has explicit dependence on the x coordinate, we have the integral equation

$$D(x,z)=\int_{-\infty}^{\infty}\Delta(x-x';\, z)D(x',z=0)\mathrm{d}x' +\int_0^z\int_{-\infty}^{\infty}\Delta(x-x';\, z-z')\delta A(x',z')D(x',z')\mathrm{d}x'\mathrm{d}z'\,. \quad (6)$$

Hence, even if the crystal is perfect, so that $\delta A=0$, we still have an integration to perform in the first term of equation (6); but we do not have an integral equation.

In summary, Δ describes scattering from a mathematical point defect in an otherwise perfect lattice, while G^I describes scattering from a point in an otherwise empty $[V(\mathbf{r})=0]$ lattice.

3. The calculation of Δ

The non-trivial nature of Δ, in contrast to G^I, is a result of the fact that the matrix operator $\hat{L}$ and the matrix A_0 do not commute as matrix operators in the n-component vector space of the diffracted wave amplitudes. As a consequence of this, Δ is neither a diagonal matrix, nor does it vanish within the triangle of determinancy, as does G^I. Actually to calculate Δ, in coordinate space, for the general n-beam case would be extremely complicated. However, the calculation is quite straightforward in the case of only two beams (a transmitted beam and a single diffracted beam). For completeness, we present the calculation in the Appendix and we will discuss the results of that calculation later in this section.

For many purposes one does not need any more than the convenient integral expression

$$\Delta(x-x';\, z-z')=\frac{\theta(z-z')}{2\pi} \times\int_{-\infty}^{\infty}\exp[ik(x-x')-i(kB-A_0)(z-z')]\mathrm{d}k\,, \quad (7)$$

which is easily verified to be a solution to equation (4), and the step function reveals that we indeed have the retarded solution. The hyperbolic nature of $\hat{L}$ still requires, as in the case of G^I, that Δ vanish outside the triangle of determinancy. This is also easily verified through equation (7) by noting that the integrand is an entire (matrix) function of k. Now, by looking at the large $|k|$ behavior of the exponential we may effectively ignore the matrix A_0 (and its associated commutation problems); then we note that when we are outside the triangle of determinancy one of the following inequalities holds:

$$\frac{(x-x')}{(z-z')}>\tan\theta_{g(\max)} \quad (8a)$$

$$\frac{(x-x')}{(z-z')}<\tan\theta_{g(\min)}\,, \quad (8b)$$

where $\theta_{g(\max)}$ and $\theta_{g(\min)}$ refer to the characteristics

with the greatest and least slopes passing through the point (x', z'). Since at least one of these inequalities holds, we can complete a contour in either the upper half or the lower half of the k plane without affecting the value of the integral, and so obtain zero due to the analyticity of the integrand. Of course, when we are inside the triangle of determinancy no such choice of contours is possible without changing the value of the integral.

One would also like to verify the unitary nature of Δ as an integral kernel. To be specific, suppose we have a perfect crystal and an arbitrary incident wave, normalized so that

$$\|D(x,z=0)\|^2 \equiv \int_{-\infty}^{\infty} D^+(x,z=0)D(x,z=0)\mathrm{d}x = 1\,; \quad (9)$$

then Δ has an action on this Hilbert space of n-component square integrable functions given by equation (6) when $\delta A = 0$. We then verify that this action is unitary by computing $\|D(x,z)\|^2$, using the integral expression of equation (7), and obtaining one because

$$\int_{-\infty}^{\infty} \Delta^+(x-s;z)\Delta(x-s';z)\mathrm{d}x = \delta(s-s')I\,, \quad (10)$$

which is merely another statement of the unitarity. Once we have verified unitarity, we expect that there is a formal expression for the integral operator as the exponential of a skew-Hermitian operator and, in fact, we have the result

$$D(x,z) = \int_{-\infty}^{\infty} \Delta(x-x';z)D(x',z=0)\mathrm{d}x' = \exp\left[\left(-B\frac{\partial}{\partial x} + iA_0\right)z\right] D(x,z=0)\,. \quad (11)$$

By differentiating $D(x,z)$ and the far right-hand side of equation (11) we recover the perfect-crystal equations. Thus, the skew-Hermitian generator associated with Δ is clearly $(-B\partial/\partial x + iA_0)$ because A_0 is a Hermitian matrix, B is Hermitian, and $-i\partial/\partial x$ is the Hermitian generator of translations. We can see how the integral operator Δ, written in the exponential form above, generates a one-parameter unitary group of transformations on the incident wave. That is to say, if we write $D(x,z) = U(z)D(x,z=0)$; then $U(z_1+z_2) = U(z_1)U(z_2)$.

The results of the two-beam calculation are easily summarized: Δ consists of two parts, a singular part much resembling G^I and a finite part containing Bessel functions J_0 and J_1. Specifically, we have written the two matrices involved in $(\hat{L} - iA_0)$ in a very general way:

$$B = \begin{pmatrix} B_1 & 0 \\ 0 & B_2 \end{pmatrix} \quad \text{and} \quad A_0 = \begin{pmatrix} A_1 A_3 \\ A_3 A_2 \end{pmatrix}.$$

Of course, $A_1 = 0$, but it is instructive for the n-beam case to leave it as A_1. As might be expected, the entire non-trivial coordinate dependence of Δ appears through the two functions $\mu_1 \equiv (x-x') - B_1(z-z')$ and $\mu_2 \equiv (x-x') - B_2(z-z')$. Of course, we need the characteristic function for the triangle of determinancy which has a vertex at the source point (x', z') in the crystal. We will write this function as $C_\Delta(x', z'; x, z)$, and it will be defined as equal to one when (x,z) is inside the triangle and zero otherwise. (We will often just drop the coordinates and write C_Δ.) Finally, we have the result of the Appendix:

$$\Delta(x-x';\, z-z') = \Delta^{(\mathrm{sing.})}(x-x';\, z-z') + \Delta^{(\mathrm{finite})}(x-x';\, z-z')\,, \quad (12)$$

where

$$\Delta^{(\mathrm{sing.})} = \theta(z-z') \times \begin{pmatrix} \delta[\mu_1]\exp[iA_1(z-z')] & 0 \\ 0 & \delta[\mu_2]\exp[iA_2(z-z')] \end{pmatrix}$$

$$\Delta^{(\mathrm{finite})} = \frac{\theta(z-z')C_\Delta \exp(i\varphi)}{-\Omega} \times \begin{pmatrix} (A_1-A_2)J_0(\xi) - iA_3\left(\frac{-\mu_2}{\mu_1}\right)^{1/2} J_1(\xi) & A_3 J_0(\xi) \\ A_3 J_0(\xi) & (A_2-A_1)J_0(\xi) - iA_3\left(\frac{-\mu_1}{\mu_2}\right)^{1/2} J_1(\xi) \end{pmatrix}$$

$\Omega \equiv B_1 - B_2 = \tan\theta_0 - \tan\theta_g$, $\varphi = (A_1\mu_2 - A_2\mu_1)/\Omega$, and $\xi \equiv -2A_3(-\mu_1\mu_2)^{1/2}/\Omega$. One should notice that when $A_0 = 0$ we recover G^I, and when only $A_3 = 0$ we are left with a diagonal Green function, since then $\hat{L}$ and A_0 would both be diagonal. Also notable is the fact that on the characteristics either $\mu_1 = 0$ or $\mu_2 = 0$, and $\Delta^{(\mathrm{finite})}$ remains bounded there, even though we have factors like $(\mu_2/\mu_1)^{1/2}$, due to the small argument behavior of $J_1(\xi)$.

In the symmetrical Laue case, when we are at the exact Bragg condition (*i.e.* $A_1 = A_2 = 0$, $B_1 = -B_2$) we have the simpler expression

$$\Delta^{(\mathrm{S.L.})} = G^I + \frac{\theta(z-z')C_\Delta A_3}{-\Omega} \begin{pmatrix} -i\tau J_1(\eta) & J_0(\eta) \\ J_0(\eta) & \frac{-i}{\tau} J_1(\eta) \end{pmatrix} \quad (13)$$

where

$$\eta = A_3(z-z')[1 - (\tan\theta/\tan\theta_g)^2]^{1/2}\,,$$

$$\tan\theta = (x-x')/(z-z')$$

and

$$\tau = [(\tan\theta_g - \tan\theta)/(\tan\theta_g + \tan\theta)]^{1/2}\,.$$

The finite part of the Green matrix, in the symmetrical Laue case, has also been derived in the work of Kuriyama & Early (1974). These authors point out that the Bessel functions arose in a calculation by Kato (1968) of X-ray diffraction in a perfect crystal with an incident spherical wave. We see that the argument η is an even function of $\tan\theta$, so that the Bessel functions contribute a function that is symmetrical between the characteristics. The function τ, however, is not

symmetric. For schematic purposes we will distinguish three thicknesses of crystal: (i) $A_3(z-z')\ll 1$ (thickness $\ll$ extinction distance), (ii) $A_3(z-z')\gtrsim 1$ (thickness $\gtrsim$ extinction distance), and (iii) $A_3(z-z')\gg 1$ (thickness $\gg$ extinction distance). In Fig. 1 we have sketched the Bessel functions *vs.* x, at constant $(z-z')$, for the three thicknesses listed above. The zeros in Fig. 1 occur when

$$\tan\theta=[1-(r_{0,1}/v)^2]^{1/2}\tan\theta_g$$

where $v=A_3(z-z')$ and either $J_0(r_0)$ or $J_1(r_1)$ equals zero. Consequently, when v is small there are no roots in the allowable range, and when v is large there are many. From an observational point of view, the best electron-microscope contrast is obtained in case (ii), since the amplitudes remain large there and absorption effects are much reduced from case (iii).

Clearly one would like to know what features of the two-beam case remain in the general n-beam case. For example, is it generally true that Δ may be broken up into a singular and finite part, where the singular part resembles G^I? Apparently the answer to this question is yes, as may be seen by the following argument: Let us write $A_0=A_0^{(d)}+A_0^{(o.d.)}$ where (d) and (o.d.) refer to the diagonal and off-diagonal parts of A_0. Then we write $\Delta=\Delta^{(sing.)}+\Delta^{(finite)}$, where we define

$$(\hat{L}-iA_0^{(d)})\Delta^{(sing.)}(x-x';\,z-z')=\delta(x-x')\delta(z-z')I\,. \quad (14)$$

A solution to this equation is clearly $\Delta^{(sing.)}=\exp[iA_0^{(d)}(z-z')]G^I$. Substitution into equation (4) implies that

$$(\hat{L}-iA_0)\Delta^{(finite)}(x-x';\,z-z')=iA_0^{(o.d.)}\Delta^{(sing.)}(x-x';\,z-z')\,. \quad (15)$$

Since the source term is now only a first-order delta function, it is quite reasonable to expect that $\Delta^{(finite)}$ is indeed finite, although this is not a rigorous conclusion.

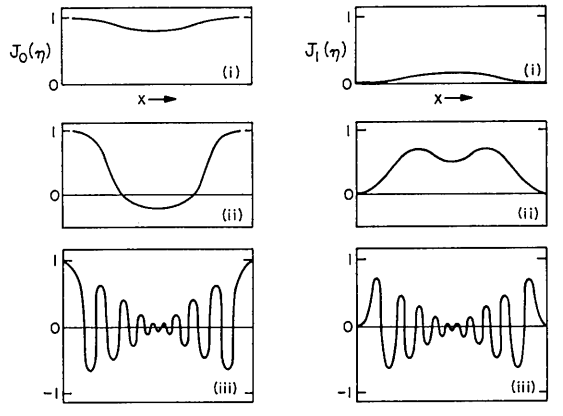

Fig. 1. Sketch of the Bessel functions $J_0(\eta)$ and $J_1(\eta)$ plotted *vs.* x, at constant $(z-z')$ for thickness cases (i), (ii), and (iii). We have rescaled the distance between the characteristics in order to make them all the same.

4. Conclusions

In this paper we have presented an integral formulation of Takagi's (1962, 1969) equation of high-energy-electron diffraction from an imperfect crystal. We have also investigated the Green function that represents scattering from a mathematical point defect in an otherwise perfect crystal, and we have shown that in the two-beam case this function consists of two terms: (i) a propagation of the singularity of the defect down the characteristics and (ii) a finite residual term consisting of oscillatory Bessel functions inside the triangle of determinancy.

We are currently studying the possibility of using the oscillatory part of the Green function to identify real point defects by high-resolution electron microscopy. In particular, we are investigating the validity of describing a point defect as a point scatterer, since the atoms near a point defect are displaced from their positions in a perfect crystal.

APPENDIX

Here we present the derivation of the two-beam Green function under arbitrary diffracting conditions. We shall write Δ in matrix form as

$$\Delta=\begin{pmatrix}\Delta_1\Delta_3\\ \Delta_4\Delta_2\end{pmatrix}.$$

Then, expanding the matrices in equation (4) we obtain the following four differential equations for the components of Δ:

$$\left(\frac{\partial}{\partial z}+B_1\frac{\partial}{\partial x}-iA_1\right)\Delta_1-iA_3\Delta_4=\delta(x-x')\delta(z-z') \quad (A1)$$

$$\left(\frac{\partial}{\partial z}+B_1\frac{\partial}{\partial x}-iA_1\right)\Delta_3-iA_3\Delta_2=0 \quad (A2)$$

$$\left(\frac{\partial}{\partial z}+B_2\frac{\partial}{\partial x}-iA_2\right)\Delta_4-iA_3\Delta_1=0 \quad (A3)$$

$$\left(\frac{\partial}{\partial z}+B_2\frac{\partial}{\partial x}-iA_2\right)\Delta_2-iA_3\Delta_3=\delta(x-x')\delta(z-z')\,. \quad (A4)$$

Making use of this system, we can derive an equation for Δ_4 alone

$$\left[\frac{1}{iA_3}\left(\frac{\partial}{\partial z}+B_1\frac{\partial}{\partial x}-iA_1\right)\left(\frac{\partial}{\partial z}+B_2\frac{\partial}{\partial x}-iA_2\right)-iA_3\right]\Delta_4=\delta(x-x')\delta(z-z')\,. \quad (A5)$$

Similarly we can derive an identical equation for Δ_3; hence $\Delta_3=\Delta_4$ apart from homogeneous solutions, which we discard. Also, once we know Δ_4, equation (A2) and equation (A3) give us the remaining components of Δ.

We can solve equation (A5) with a Laplace transform in z and a Fourier transform in x, yielding the solution

$$\Delta_4(x-x';\, z-z') = \frac{iA_3}{4\pi^2} \times \int_{-\infty}^{\infty}\int_{c-i\infty}^{c+i\infty} \frac{\exp\left[ik(x-x')+s(z-z')\right]}{(s+ikB_1-iA_1)\,(s+ikB_2-iA_2)+A_3^2}\,\mathrm{d}s\mathrm{d}k \tag{A6}$$

where c is to the right of all poles in the complex s plane. It is convenient to rotate this plane by letting $s=iy$ (see Fig. 2) and we obtain

$$\Delta_4(x-x';\, z-z') = \frac{A_3}{4\pi^2 i} \times \int_{-\infty}^{\infty}\int_{-ic-\infty}^{-ic+\infty} \frac{\exp\left[ik(x-x')+iy(z-z')\right]}{[y-y_1(k)]\,[y-y_2(k)]}\,\mathrm{d}y\mathrm{d}k \tag{A7}$$

where $y_{1,2}$ are roots of the quadratic equation

$$y^2+y[(kB_1-A_1)+(kB_2-A_2)] +[(kB_1-A_1)\,(kB_2-A_2)-A_3^2]=0\,, \tag{A8}$$

so that

$$y_1(k)=(-\tfrac{1}{2})[k(B_1+B_2)-(A_1+A_2)]+(\tfrac{1}{2})[d(k)]^{1/2},$$

$$y_2(k)=(-\tfrac{1}{2})[k(B_1+B_2)-(A_1+A_2)]-(\tfrac{1}{2})[d(k)]^{1/2},$$

and

$$d(k)=[k(B_1-B_2)-(A_1-A_2)]^2+4A_3^2\,.$$

The discriminant, $d(k)$, being the sum of two squares (of real quantities), is always positive; hence y_1, and y_2 are real and non-degenerate. If $(z-z')>0$ we complete the y-plane contour to the upper half-plane, thereby collecting the residues of the poles on the real axis. If $(z-z')<0$ we obtain zero; hence

$$\Delta_4(x-x';\, z-z') = -\frac{\theta(z-z')}{\pi i}\,A_3 \times \exp\left[i(A_1+A_2)\,(z-z')/2\right] \times \int_{-\infty}^{\infty} \frac{\sin\left\{(z-z')[\mathrm{d}(k)]^{1/2}/2\right\}}{[\mathrm{d}(k)]^{1/2}} \times \exp\left\{ik[(x-x')-(B_1+B_2)(z-z')/2]\right\}\mathrm{d}k\,. \tag{A9}$$

We now make use of the fact that $B_1\equiv\tan\theta_0$ and $B_2\equiv\tan\theta_g$ to distinguish three regions in coordinate space (see Fig. 3). With the conventions of this coordinate system $B_1<0$ and $B_2>0$, so that: (i) in region I $\gamma-B_1<0$ and $\gamma-B_2<0$, (ii) in region II $\gamma-B_1>0$ and $\gamma-B_2>0$, and (iii) in region III $\gamma-B_1>0$ and $\gamma-B_2<0$ where $\gamma\equiv(x-x')/(z-z')$. Note that the integrand of equation (A9) is an analytic function everywhere in the complex k plane. When we are outside the triangle of determinancy, in regions I, or II, the above inequalities enable us to select either contour Γ_{I} or Γ_{II} (see Fig. 4), so that the integrand falls to zero exponentially along these contours; hence the integral is zero, since the contours enclose a region of analyticity. When we are in region III, the integrand does not fall off exponentially as we move off to infinity in either the upper-half or the lower-half plane, so that no such choice of contours is possible.

In order to render equation (A9) into a more recognizable form, let us write

$$d(k)=(B_1-B_2)^2[(k-k_{0,R})^2+k_{0,I}^2]$$

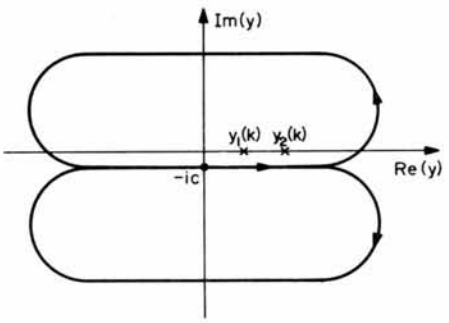

Fig. 2. The complex y plane used for the inversion of the Laplace transform. The poles $y_1(k)$ and $y_2(k)$ are always on the real axis. This transform is easily inverted even in the general n-beam case.

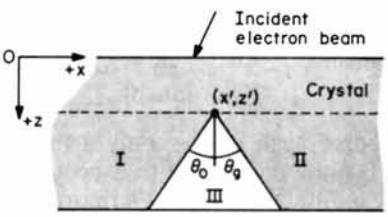

Fig. 3. The coordinate space used for the inversion of the Fourier transform.

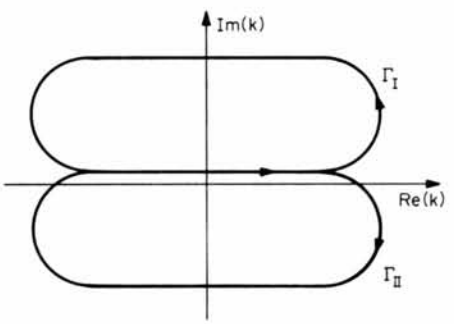

Fig. 4. The complex k plane used for the inversion of the Fourier transform. There are no poles or branch cuts in the k plane and contours Γ_{I} and Γ_{II} may be selected when in region I or region II in coordinate space.

where $k_{0,R}$ and $k_{0,I}$ are the real and imaginary parts of the roots of the equation $d(k)=0$. Then we translate the integral, letting $k'=k-k_{0,R}$. This gives us

$$\Delta_4(x-x';\, z-z') = -\frac{\theta(z-z')A_3C_\Delta}{\pi(B_1-B_2)} \times \exp\{i[(A_1+A_2)(z-z')/2+\alpha k_{0,R}]\} \times \int_{-\infty}^{\infty} \frac{\sin[\beta(k^2+k_{0,I}^2)^{1/2}]}{(k^2+k_{0,I}^2)^{1/2}} \exp(i\alpha k)\mathrm{d}k \qquad \text{(A10)}$$

where $\alpha=(x-x')-(B_1-B_2)(z-z')/2$ and $\beta=[(B_2-B_1)(z-z')/2]>0$. Next, we make two more obvious transformations; first let $k=k_{0,I}\sinh t$ and write out the sine in terms of exponentials. Then, let $\alpha\sinh t+\beta\cosh t=\pm(\beta^2-\alpha^2)^{1/2}\cosh\theta$ where $\beta>0$ and $\beta>\alpha$ and the $+(-)$ goes with the first (second) exponential. These transformations result in the integral in equation (A10) becoming

$$\frac{1}{\pi}\int_{-\infty}^{\infty} \sin[|k_{0,I}|(\beta^2-\alpha^2)^{1/2}\cosh\theta]\mathrm{d}\theta = J_0[|k_{0,I}|(\beta^2-\alpha^2)^{1/2}]\,.$$

We note that $k_{0,R}=(A_1-A_2)/\Omega$ and $k_{0,I}=\pm 2A_3/\Omega$ where $\beta^2-\alpha^2=-(\beta+\alpha)(\alpha-\beta)=-\mu_1\mu_2$; therefore we can write equation (A10) as

$$\Delta_4(x-x';\, z-z') = -\frac{\theta(z-z')C_\Delta A_3}{\Omega}\exp(i\varphi)J_0(\xi)\,. \qquad \text{(A11)}$$

To compute the diagonal elements of Δ, we need the relations:

$$\left(\frac{\partial}{\partial z}+B_1\frac{\partial}{\partial x}\right)\mu_1 = \left(\frac{\partial}{\partial z}+B_2\frac{\partial}{\partial x}\right)\mu_2 = 0$$

and

$$\left(\frac{\partial}{\partial z}+B_1\frac{\partial}{\partial x}\right)\mu_2 = -\left(\frac{\partial}{\partial z}+B_2\frac{\partial}{\partial x}\right)\mu_1 = \Omega\,.$$

Then using these relationships we obtain

$$\Delta_1(x-x';\, z-z') = \theta(z-z')\left\{\delta[\mu_1]\exp[iA_1(z-z')] - \frac{C_\Delta\exp(i\varphi)}{\Omega}\left[(A_1-A_2)J_0(\xi) - iA_3\left(-\frac{\mu_1}{\mu_2}\right)^{1/2}J_1(\xi)\right]\right\} \qquad \text{(A13)}$$

and

$$\Delta_2(x-x';\, z-z') = \theta(z-z')\left\{\delta[\mu_2]\exp[iA(z-z')] - \frac{C_\Delta\exp(i\varphi)}{\Omega}\left[(A_2-A_1)J_0(\xi) - iA_3\left(-\frac{\mu_2}{\mu_1}\right)^{1/2}J_1(\xi)\right]\right\} \qquad \text{(A14)}$$

where the Dirac delta functions are a result of having to differentiate the characteristic functions, while J_1 is a result of having to differentiate J_0.

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The Joint Probability Distribution of Structure Factors: The First-Order Term

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The first-order term of the joint probability distribution of $E_{\mathbf{h}_1+\mathbf{k}},\ldots,E_{\mathbf{h}_m+\mathbf{k}}$, for $\mathbf{h}_1,\ldots,\mathbf{h}_m$ fixed and $\mathbf{k}$ variable, is derived for both space groups $P\bar{1}$ and $P1$. It appears that the first-order term affects the most probable values for the moduli of the structure factors, but that it has no influence on the most probable values for the phases.

Introduction

The main term of the joint probability distribution of an arbitrary number of structure factors has been obtained from the central-limit theorem (Tsoucaris, 1970). From this distribution, formulae for the most probable values of structure factors have been derived (Tsoucaris, 1970; de Rango, Tsoucaris & Zelwer, 1974). The present paper deals with the first-order term of this distribution. To calculate this term use is made of Hauptman's (1971) method for the derivation of the joint probability distributions of two and three