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The Exact Dynamical Wave Fields for a Crystal with a Constant Strain Gradient on the Basis of the Takagi–Taupin Equations

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From the Takagi–Taupin equations for two-beam cases, the exact wave fields for a spherical incident wave are obtained in the Laue case for a crystal having a constant strain gradient. Absorption is taken into account. Both direct and Bragg-reflected waves are essentially expressed in terms of confluent hypergeometric functions. Their characters depend on strain gradient, structure factor and crystal thickness. The wave fields tend to those obtained by Eikonal theory as the strain gradient decreases. For an extremely large strain gradient, the wave fields reduce to those predicted by the kinematical theory.

1. Introduction

The wave-optical theories of crystal diffraction for distorted crystals have been developed along two lines of consideration. One is Eikonal theory (Kato, 1963, 1964*a*; Kambe, 1965, 1968) which is based on the concept of rays. The other theory is purely wave-optical and the master equations were presented by Takagi (1962, 1969) and Taupin (1964). The Eikonal theory is to be derived from the Takagi–Taupin equations by using a kind of W–K–B approximation.

So far, the Takagi–Taupin equations have not been exactly solved for any continuously distorted crystal. On the other hand, the analytical solution within the framework of Eikonal theory is available for the Laue case of a constant strain gradient (Kato, 1964*b*; Kato & Ando, 1966). In this paper, the authors present an analytical solution of the Takagi–Taupin equations for the case mentioned above without any approximation. The solution is represented in a form of Laplace integral, from which two kinds of asymptotic expansions are derived. They are useful for comparison with the kinematical and Eikonal theories. The solution is also represented essentially in terms of confluent hypergeometric functions.

The wave field obtained tends to the results of the kinematical theory as the strain gradient increases, and tends to the solution of the Eikonal theory with decreasing strain gradient unless the crystal is extremely thin. From this analysis, the applicability of Eikonal theory can be elucidated in this particular case.

2. The wave equations and the boundary conditions

We shall consider the crystal wave field in the form

$$D(\mathbf{r}) = D_0(\mathbf{r}) \exp i(\bar{\mathbf{k}}_0 \, . \, \mathbf{r}) + D_g(\mathbf{r}) \exp i(\bar{\mathbf{k}}_g \, . \, \mathbf{r}) \quad (1)$$

where the wave vectors $\bar{\mathbf{k}}_0$ and $\bar{\mathbf{k}}_g$ satisfy the relations

$$\bar{\mathbf{k}}_0^2 = \bar{\mathbf{k}}_g^2 = K^2(1+\chi_0) \quad (2a)$$

and

$$\bar{\mathbf{k}}_g = \bar{\mathbf{k}}_0 + 2\pi\bar{\mathbf{g}} \, , \quad (2b)$$

K and χ_0 being 2π times the wave number in vacuum and the mean polarizability of the crystal respectively. The reciprocal-lattice vector $\bar{\mathbf{g}}$ is referred to a perfect crystal.

When the crystal is distorted by the displacement vector $\mathbf{u}$ at the position $\mathbf{r}$, the spatial variations of the amplitudes D_0 and D_g are described by equations of the Takagi–Taupin type of the form (Kato, 1973)

$$iK^{-1}\frac{\partial D_0}{\partial s_0} + \frac{1}{2}\, C\chi_{-g}D_g \exp\,[2\pi i(\bar{\mathbf{g}} \, . \, \mathbf{u})] = 0 \quad (3a)^*$$

$$iK^{-1}\frac{\partial D_g}{\partial s_g} + \frac{1}{2}\, C\chi_g D_0 \exp\,[-2\pi i(\bar{\mathbf{g}} \, . \, \mathbf{u})] = 0 \quad (3b)^*$$

where (χ_g, χ_{-g}) are the Fourier components of the polarizability χ of the crystal and C is the X-ray polarization factor which is conventionally used. The coordinate variables s_0 and s_g are referred to an oblique coordinate system having the directions of $\bar{\mathbf{k}}_0$ and $\bar{\mathbf{k}}_g$. The assumptions in deriving equations (3) are discussed in the previous papers (Takagi, 1962, 1969; Taupin, 1964; Kato, 1973).

In this paper we shall consider the case in which a spherical wave of the form

$$E(\mathbf{r}) = \frac{E_0}{4\pi|\mathbf{r}-\mathbf{r}_0|} \exp iK|\mathbf{r}-\mathbf{r}_0| \quad (4)$$

falls on a crystal through a sufficiently narrow slit. Under the approximation used in the spherical wave theory, the wave field which is effective in crystal diffraction can be written on the entrance surface $\mathbf{r}_e$ as

$$E(\mathbf{r}_e) = A\delta(s_g) \quad (5)$$

where $\delta(s_g)$ is the Dirac delta function and A is given by

$$A = \frac{E_0}{4\pi \sin 2\theta_B}\left(\frac{2\pi}{KL}\right)^{1/2} \exp i\left(KL + \frac{\pi}{4}\right), \quad L = |\mathbf{r}_0| \quad (6)$$

* Similar but slightly different equations have been proposed by Takagi (1969).

with the origin of the coordinates taken at the entrance point. The justification is given at least for the case of perfect crystals (Saka, Katagawa & Kato, 1973). The mathematical arguments will be reported in a separate paper. In any case, assuming the incident wave (5) on the entrance surface, we shall solve the Takagi–Taupin equations under the boundary conditions

$$D_0(0,s_g)=A\delta(s_g) \quad s_g \geq -\varepsilon \tag{7a}$$

$$D_g(s_0,-\varepsilon)=0 \quad s_0 \geq 0 \tag{7b}$$

on the edges of the Borrmann fan.

3. The case of a constant strain gradient

In this section, we shall treat a special case where the displacement has the form

$$2\pi(\bar{\mathbf{g}}\,.\,\mathbf{u})=fs_0s_g\,. \tag{8}$$

Obviously, the force parameter which characterizes diffraction phenomena in the Eikonal theory (Kato, 1964*a*),

$$F=\frac{2\pi}{\sin 2\theta_B}\,\frac{\partial^2}{\partial s_0\partial s_g}\,(\bar{\mathbf{g}}\,.\,\mathbf{u}), \tag{9}$$

turns out to be the constant $f/\sin 2\theta_B$.

Let us introduce the Laplace transform of the wave field D_0

$$V(s_0,p)=\int_{-\varepsilon}^{\infty} D_0(s_0,s_g)\exp(-ps_g)\mathrm{d}s_g\,. \tag{10}$$

By the inversion theorem (Sneddon, 1951), the wave field D_0 can be represented as

$$D_0(s_0,s_g)=\frac{1}{2\pi i}\int_{\gamma-i\infty}^{\gamma+i\infty} V(s_0,p)\exp(ps_g)\mathrm{d}p \tag{11}$$

where γ is a real constant larger than the abscissa of convergence which turns out to be zero after the functional form of $V(s_0,p)$ is determined. Substituting equation (11) into equation (3*a*), one can write the wave field D_g as

$$D_g(s_0,s_g)=-\frac{2i}{KC\chi_{-g}}\exp(-ifs_0s_g)\frac{1}{2\pi i}\times\int_{\gamma+i\infty}^{\gamma+i\infty}\frac{\partial}{\partial s_0}V(s_0,p)\exp(ps_g)\mathrm{d}p\,. \tag{12}$$

By the use of equations (10) and (12), the Laplace transform of equation (3*b*) multiplied by $\exp(ifs_0s_g)$ gives

$$(p-ifs_0)\frac{\partial}{\partial s_0}V(s_0,p)+\frac{1}{4}K^2C^2\chi_g\chi_{-g}V(s_0,p)=\frac{i}{2}KC\chi_{-g}D_g(s_0,-\varepsilon)\,. \tag{13}$$

The right-hand side, however, is zero by virtue of the boundary condition (7*b*). Equation (13), therefore, can be solved easily as

$$V(s_0,p)=V(0,p)\left(\frac{p-ifs_0}{p}\right)^{\sigma} \tag{14}$$

where

$$\sigma=\frac{K^2C^2\chi_g\chi_{-g}}{4if}\,. \tag{15}$$

By the use of the boundary condition (7*a*) and equation (10), the integral constant $V(0,p)$ is determined as

$$V(0,p)=A\int_{-\varepsilon}^{\infty}\delta(s_g)\exp(-ps_g)\mathrm{d}s_g=A\,. \tag{16}$$

The wave field D_0 can be written by substituting the expression (14) together with equation (16) into equation (11)

$$D_0(s_0,s_g)=\frac{A}{2\pi i}\int_{\gamma-i\infty}^{\gamma+i\infty}\left(1-\frac{ifs_0}{p}\right)^{\sigma}\exp(ps_g)\mathrm{d}p\,. \tag{17a}$$

From equations (12) and (13), the wave field D_g can be obtained as

$$D_g(s_0,s_g)=\frac{A}{2\pi}\,\frac{KC\chi_g}{2}\exp(-ifs_0s_g)\times\int_{\gamma-i\infty}^{\gamma+i\infty}\frac{1}{p-ifs_0}\left(1-\frac{ifs_0}{p}\right)^{\sigma}\exp(ps_g)\mathrm{d}p\,. \tag{17b}$$

Since $V(s_0,p)$ has singularities at the origin and a pure imaginary ifs_0 in the p-plane, it turns out that the abscissa of convergence is zero. In addition, when s_g is negative, the contour integral along the path $C(+)$ shown in Fig. 1 is zero. Since the integral path composed of $L(\gamma)$ and $C(+)$ includes no singular point, the integrals in equations (17) become zero, so that $D_g(s_0,s_g)$ always satisfies the boundary condition (7*b*). On the other hand, in the region of interest ($s_g>0$) the contour integral along the path $C(-)$ is zero. Since the integral path $L(\gamma)+C(-)$ encloses singular points, the integral on $L(\gamma)$ may take on some value. The integral path of equations (17) can be modified to any closed curve containing the singular points.

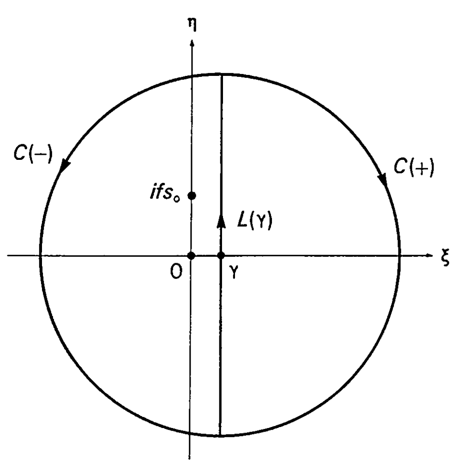

Fig. 1. The integral paths in the plane of $p=\xi+i\eta$.

If the variable p is replaced by $it+\gamma$, the expressions (17) can be written also in the form of the Fourier integrals

$$D_0(s_0,s_g)=\frac{A}{2\pi}\exp(\gamma s_g)\int_{-\infty}^{\infty}\left(1-\frac{fs_0}{t-i\gamma}\right)^{\sigma}\times\exp(its_g)\mathrm{d}t \quad (18a)$$

$$D_g(s_0,s_g)=\frac{A}{2\pi}\frac{KC\chi_g}{2}\exp(\gamma s_g-ifs_0s_g)\times\int_{-\infty}^{\infty}\frac{1}{t-i\gamma-fs_0}\left(1-\frac{fs_0}{t-i\gamma}\right)^{\sigma}\exp(its_g)\mathrm{d}t \quad (18b)$$

where γ is an arbitrary positive number.

For performing the integrals in equations (17), the following Laurent series are useful

$$\left(1-\frac{ifs_0}{p}\right)^{\sigma}=1+\sum_{n=0}^{\infty}\frac{\Gamma(n+1-\sigma)}{\Gamma(-\sigma)\,(n+1)!}\left(\frac{ifs_0}{p}\right)^{n+1} \quad (19a)$$

$$\frac{1}{p-ifs_0}\left(1-\frac{ifs_0}{p}\right)^{\sigma}=\sum_{n=0}^{\infty}\frac{\Gamma(n+1-\sigma)}{\Gamma(1-\sigma)n!}\frac{(ifs_0)^n}{p^{n+1}}\,. \quad (19b)$$

In the integration in equation (17a), the first term of the right-hand side of equation (19a) gives us $A\delta(s_g)$ which is the non-diffracted part of the crystal wave. Since we are interested in the region $s_g>0$, this term is omitted in the following discussion, unless specifically stated. Actually, the series (19) converge uniformly on a circle with the centre at the origin, provided that the radius is larger than $|fs_0|$. If the relations are recalled

$$\frac{1}{2\pi i}\int_{\gamma-i\infty}^{\gamma+i\infty}\frac{\exp(ps_g)}{p^{n+1}}\mathrm{d}p=\frac{1}{2\pi i}\oint\frac{\exp(ps_g)}{p^{n+1}}\mathrm{d}p=\frac{s_g^{\,n}}{n!}\;(s_g>0)\,, \quad (20)$$

the wave fields D_0 and D_g are represented by

$$D_0(s_0,s_g)=iAfs_0\sum_{n=0}^{\infty}\frac{\Gamma(n+1-\sigma)}{\Gamma(-\sigma)n!(n+1)!}(ifs_0s_g)^n \quad (21a)$$

$$=-\frac{1}{4}AK^2C^2\chi_g\chi_{-g}s_0F(1-\sigma,2,ifs_0s_g) \quad (21b)^*$$

$$D_g(s_0,s_g)=\frac{i}{2}AKC\chi_g\exp(-ifs_0s_g)\times\sum_{n=0}^{\infty}\frac{\Gamma(n+1-\sigma)}{\Gamma(1-\sigma)\,(n!)^2}(ifs_0s_g)^n \quad (22a)$$

$$=\frac{i}{2}AKC\chi_gF(\sigma,1,-ifs_0s_g) \quad (22b)^*$$

where $F(a,c,z)$ is a confluent hypergeometric function.*

As a special case, it is worth considering the perfect crystal where f tends to zero, namely $|\sigma|$ tends to infinity. Recalling the relation

$$\lim_{f\to0}\frac{\Gamma(n+1-\sigma)}{\Gamma(1-\sigma)}(if)^n=\lim_{f\to0}(n-\sigma)(n-1-\sigma)\ldots(1-\sigma)(if)^n=(-1)^n\left(\frac{K^2C^2\chi_g\chi_{-g}}{4}\right)^n, \quad (23)$$

one obtains

$$D_0(s_0,s_g)=-\frac{1}{4}AK^2C^2\chi_g\chi_{-g}s_0\sum_{n=0}^{\infty}\frac{(-1)^n}{n!(n+1)!}\times\left(\frac{K^2C^2\chi_g\chi_{-g}}{4}s_0s_g\right)^n=-\frac{1}{2}AK|C|\sqrt{\chi_g\chi_{-g}}\times\sqrt{\frac{s_0}{s_g}}J_1(K|C|\sqrt{\chi_g\chi_{-g}}\sqrt{s_0s_g}) \quad (24a)$$

$$D_g(s_0,s_g)=\frac{i}{2}AKC\chi_g\sum_{n=0}^{\infty}\frac{(-1)^n}{(n!)^2}\left(\frac{K^2C^2\chi_g\chi_{-g}}{4}s_0s_g\right)^n=\frac{i}{2}AKC\chi_gJ_0(K|C|\sqrt{\chi_g\chi_{-g}}\sqrt{s_0s_g})\,. \quad (24b)$$

The results are nothing else but those obtained from the spherical wave theory for perfect crystals (Kato, 1968).

4. Comparison with the Eikonal theory

It is clear that the Eikonal theory is valid only when the observation point is sufficiently far from the edge of the Borrman fan, even when the crystal is perfect. In addition, the theory is expected to be correct in the case of small strain gradient. For this reason, we shall obtain the asymptotic expansion of equations (17) by the method of steepest descent for the cases of large $|s_0s_g|^{1/2}$ and $|\sigma|$.

Equations (17) have the form

$$D_h=\int_{\gamma-i\infty}^{\gamma+i\infty}A_h(p)\exp[\varphi(p)]\mathrm{d}p\quad(h=0\text{ and }g) \quad (25)$$

where the amplitude $A_h(p)$ and the phase $\varphi(p)$ are given by

$$A_0(p)=\frac{A}{2\pi i} \quad (26a)$$

* Here, the notation

$$F(a,c,z)=\frac{\Gamma(c)}{\Gamma(a)}\sum_{n=0}^{\infty}\frac{\Gamma(n+a)}{\Gamma(n+c)}\frac{z^n}{n!}$$

and Kummer's first formula

$$F(a,c,z)=e^zF(c-a,c,-z)$$

are employed. The function F is identical to ${}_1F_1$ in the textbook of Jeffereys & Jeffereys (1956).

$$A_g(p) = \frac{A}{2\pi} \frac{KC\chi_g}{2} \exp(-ifs_0 s_g) \frac{1}{p - ifs_0} \tag{26b}$$

$$\varphi(p) = ps_g + \sigma \log\left(1 - \frac{ifs_0}{p}\right). \tag{27}$$

According to the standard procedure, the first term of the expansion is given by

$$D_h \simeq \sum_j A_h(p^j)\left[-\frac{2\pi}{\varphi''(p^j)}\right]^{1/2} \exp \varphi(p^j). \tag{28}$$

The summation is taken over the relevant saddle points specified by (j), which are determined by the relation

$$\varphi'(p) = s_g + \frac{1}{4} \frac{K^2C^2\chi_g\chi_{-g}}{p^2 - ifs_0 p} s_0 = 0. \tag{29}$$

The saddle points are given by

$$p^{(j)} = \frac{i}{2}\{fs_0 \pm \sqrt{(fs_0)^2 + K^2C^2\chi_g\chi_{-g}(s_0/s_g)}\} \tag{30}$$

where the upper and the lower signs correspond to $(j) = 1$ and 2 respectively. The function $\varphi''(p)$ is given by

$$\varphi''(p) = -\frac{1}{4} K^2C^2\chi_g\chi_{-g} \frac{2p - ifs_0}{(p^2 - ifs_0 p)^2} s_0. \tag{31}$$

Hence, substituting from (30) into (28), one obtains

$$D_0 \simeq A_0^{(1)}\exp\left[i\left(\varphi_A + \frac{\pi}{4}\right)\right] + A_0^{(2)}\exp\left[-i\left(\varphi_A + \frac{\pi}{4}\right)\right] \tag{32a}$$

$$D_g \simeq A_g^{(1)}\exp\left[i\left(\varphi_A - \frac{\pi}{4}\right)\right] + A_g^{(2)}\exp\left[-i\left(\varphi_A - \frac{\pi}{4}\right)\right] \tag{32b}$$

where $A_0^{(j)}$, $A_g^{(j)}$ and φ_A are given by

$$A_0^{(1)} = A_0^{(2)} = \frac{A}{\sqrt{2\pi}} \frac{K|C|\sqrt{\chi_g\chi_{-g}}}{2} \exp\left(\frac{i}{2} fs_0 s_g\right) \times \sqrt{\frac{s_0}{s_g}}\left(\frac{1}{N^2} + \frac{1}{S^2}\right)^{-1/4} \frac{1}{N} \tag{33a}$$

$$A_g^{(j)} = i\frac{A}{\sqrt{2\pi}} \frac{KC\chi_g}{2} \exp\left(-\frac{i}{2} fs_0 s_g\right)\left(\frac{1}{N^2} + \frac{1}{S^2}\right)^{-1/4} \times \left(\sqrt{\frac{1}{N^2} + \frac{1}{S^2}} \pm \frac{1}{S}\right) \tag{33b}$$

$$\varphi_A = \frac{1}{2}\sqrt{K^2C^2\chi_g\chi_{-g}s_0 s_g + (fs_0 s_g)^2} + \frac{K^2C^2\chi_g\chi_{-g}}{2f} \sinh^{-1}\frac{f}{K|C|}\sqrt{\frac{s_0 s_g}{\chi_g\chi_{-g}}} \tag{34}$$

with the notations

$$N = K|C|\sqrt{\chi_g\chi_{-g}}\sqrt{s_0 s_g} \tag{35a}$$

$$S = K^2C^2\chi_g\chi_{-g}/f. \tag{35b}$$

The details of calculation are explained in the Appendix. The results are identical with those obtained previously from the Eikonal theory [see equations (44a, b) and (39a) of Kato's 1964b paper]. The imaginary part of Eikonal [equation (40a) of the above paper] is automatically included in equation (34). Incidentally, equation (29) gives a relation between s_0 and s_g for the wave specified by p. The relation is identical with the expression for the trajectory of a hyperbolic form derived from Fermat's principle.

The second terms of the asymptotic expansion are discussed in the Appendix. The results are summarized as follows:

The second term of O wave;

$$A_0^{(1)}B_h^{(1)} \exp\left[i\left(\varphi_A + \frac{\pi}{4}\right)\right] + A_0^{(2)}B_0^{(2)} \exp\left[-i\left(\varphi_A + \frac{\pi}{4}\right)\right]; \tag{36a}$$

The second term of the G wave;

$$A_g^{(1)}B_g^{(1)} \exp\left[i\left(\varphi_A - \frac{\pi}{4}\right)\right] - A_g^{(2)}B_g^{(2)} \exp\left[-i\left(\varphi_A - \frac{\pi}{4}\right)\right] \tag{36b}$$

where $B_0^{(j)}$ and $B_g^{(j)}$ are given by

$$B_0^{(j)} = \pm i\left(\frac{1}{N^2} + \frac{1}{S^2}\right)^{-3/2}\left(\frac{3}{8N^4} + \frac{1}{2N^2S^2} + \frac{1}{3S^4}\right) \tag{37a}$$

$$B_g^{(j)} = \mp i\left(\frac{1}{N^2} + \frac{1}{S^2}\right)^{-3/2}\left(\frac{1}{8N^4} \mp \frac{1}{2N^2S}\sqrt{\frac{1}{N^2} + \frac{1}{S^2}} - \frac{1}{3S^4}\right). \tag{37b}$$

5. The approximated formula for a large $|fs_0 s_g|$

In the present case, the following asymptotic expansions of the confluent hypergeometric functions are available, (Jeffereys & Jeffereys, 1956):

$$\begin{aligned} F(1-\sigma, 2, ifs_0 s_g) &= \frac{\exp(ifs_0 s_g)}{\Gamma(1-\sigma)}(ifs_0 s_g)^{-\sigma-1}\left\{1 + \frac{\sigma(1+\sigma)}{ifs_0 s_g} + \frac{\sigma(1+\sigma)(1+\sigma)(2+\sigma)}{(ifs_0 s_g)^2} + \ldots\right\} \\ &+ \frac{1}{\Gamma(1+\sigma)}(-ifs_0 s_g)^{\sigma-1}\left\{1 - \frac{(-\sigma)(1-\sigma)}{ifs_0 s_g} + \frac{(-\sigma)(1-\sigma)(1-\sigma)(2-\sigma)}{2(ifs_0 s_g)^2} - \ldots\right\} \end{aligned} \tag{38a}$$

$$F(\sigma, 1, -ifs_0 s_g) = \frac{1}{\Gamma(1-\sigma)}(ifs_0 s_g)^{-\sigma}\left\{1 + \frac{\sigma^2}{ifs_0 s_g} + \frac{\sigma^2(1+\sigma)^2}{2(ifs_0 s_g)^2} + \ldots\right\}$$

$$+\frac{\exp(-ifs_0s_g)}{\Gamma(\sigma)}(-ifs_0s_g)^{\sigma-1}\left\{1-\frac{(1-\sigma)^2}{ifs_0s_g}\right.$$
$$\left.+\frac{(1-\sigma)^2(2-\sigma)^2}{2(ifs_0s_g)^2}-\ldots\right\}. \qquad (38b)$$

As $|f|$ increases, equation (38*a*) tends to zero. The O wave (17*a*), therefore, includes only the non-diffracted part

$$D_0(s_0,s_g)=A\delta(s_g). \qquad (39a)$$

Since equation (38*b*) tends to unity, one obtains from equation (22*b*)

$$D_g(s_0,s_g)=\frac{i}{2}AKC\chi_g\,. \qquad (39b)$$

This is nothing but the kinematically diffracted wave from the O wave having the form of equation (39*a*). The results are reasonable, although the Takagi–Taupin equation would be invalid in the case of extremely heavy distortions.

6. Discussion

1. *Remarks on the general case of constant force parameter F*

The application of the theory so far developed is not limited to the special distortion given by equation (8). In fact, if the displacement vector **u** has the more general form

$$2\pi(\bar{\mathbf{g}}\,.\,\mathbf{u})=fs_0s_g+g(s_0)+h(s_g) \qquad (40)$$

where $g(s_0)$ and $h(s_g)$ are arbitrary functions, one can show that the wave fields

$$\tilde{D}_0=D_0\exp[ih(s_g)-ih(0)] \qquad (41a)$$

$$\tilde{D}_g=D_g\exp[-ig(s_0)-ih(0)] \qquad (41b)$$

satisfy the differential equations (3) for the phase $2\pi(\bar{\mathbf{g}}\,.\,\mathbf{u})$ given by equation (40). The boundary conditions (7) also are automatically satisfied, since the additional phase factor in equation (41*a*) is unity at $s_g=0$, only where D_0 takes a particular value. For this reason, the present results can be used for physically important cases such as the homogeneously bending crystal and the crystals having a homogeneous temperature gradient (Hart, 1966).

2. *Remarks on the boundary conditions*

The boundary conditions (7) yield the correct solution only in the region specified by $s_0>0$ and $s_g>0$, namely in the Borrmann fan. For finding the solution over the whole crystal, it is reasonable to take boundary conditions

$$D_0(0,s_g)=A\delta(s_g) \qquad -\infty<s_g<\infty \qquad (42a)$$

$$D_g(s_0,-\varepsilon)=0 \qquad s_0>0 \qquad (42b)$$

$$D_g(s_0,+\varepsilon)=0 \qquad s_0<0\,. \qquad (42c)$$

The solution under these conditions is obtained by a similar procedure to that described in §3, namely bilateral Laplace transform. Actually, it has the same form as equations (17). In this case, however, the constant γ must be taken either positive or negative according to whether $s_0>0$ or $s_0<0$. The solution takes an appreciable value in the region $s_0s_g>0$ and zero in the region specified by $s_0s_g<0$. In the Laue case the crystal surface always lies in the latter region, so that the solution takes on a value only within the Borrmann fan in the crystal. The waves in the triangular fan specified by $s_0<0$ and $s_g<0$ are regarded as those which would be connected to the wave fields in the real Borrmann fan, if the crystal was hypothetically extended to whole space.

3. *The applicable range of Eikonal theory*

Since the expressions (32) are the asymptotic expansion of the rigorous solution, strictly speaking, the remainders must be calculated for the estimation of errors in taking only the first term, which actually gives the result of Eikonal theory. Here, however, the applicability of the latter theory is examined by putting a critical value Q on the ratio between the main term and the terms of the next approximation in the expansion series, namely on $|B_0^{(j)}|$ for the O wave and $|A_g^{(j)}B_g^{(j)}/A_g^{(1)}|$ or $|A_g^{(j)}B_g^{(j)}/A_2^{(j)}|$ for the G wave, depending on either $|A_g^{(1)}|>|A_g^{(2)}|$ or $|A_g^{(1)}|<|A_g^{(2)}|$. The effect of absorption, $\mathrm{Im}\,(\chi_g\chi_{-g})$, is neglected in the following arguments.

First the case of $f\geq 0(|A_g^{(1)}|\geq|A_g^{(2)}|)$ is discussed; the critical conditions must be

$$|B_g^{(1)}|<Q \quad \text{and} \quad |A_g^{(2)}B_g^{(2)}/A_g^{(1)}|<Q \qquad (43a,b)$$

for the Bragg-reflected waves. On the other hand, the condition for the direct wave is simply

$$|B_0^{(1)}|=|B_0^{(2)}|<Q \qquad (44)$$

because $|A_0^{(1)}|$ is always identical to $|A_0^{(2)}|$. The numerical results of the critical conditions for the case $Q=\frac{1}{30}$ are illustrated in Fig. 2, in which the hatched region is the applicable range of the Eikonal theory. From equations (33) and (37), it is obvious that the critical curve for other figures of Q have the similar form, the change being simply a matter of scaling.

The crystal must be thicker than a certain limit. This situation is not new because even in the perfect crystal Eikonal theory holds only when the wave propagates a certain distance from the entrance point. As another criterion, obviously, the strain gradient must be less than a critical value. The significant thing is that the two critical conditions can be regarded as nearly independent because the applicable range of Eikonal theory for each case of the direct and the Bragg-reflected waves is roughly approximated by a rectangular region in Fig. 2.

In the case of $f \le 0$ where $|A_g^{(2)}| \ge |A_g^{(1)}|$ and $|A_0^{(1)}| = |A_0^{(2)}|$, the critical conditions are given by interchanging the indices (1) and (2) in equations (43) and (44). Owing to the symmetrical characters of the expressions (33) and (37), the condition for $f \le 0$ is identical with the criteria for $f \ge 0$.

When the crystal is sufficiently thick, the applicability of the Eikonal theory can be more concretely discussed on the basis of the asymptotic formula (38*b*). Here, only *G* waves are discussed in detail. As $|s_0 s_g|^{1/2}$ is increased, equation (22*b*) tends to the form

$$D_g(s_0, s_g) \simeq \frac{i}{2} AKC\chi_g \frac{(ifs_0 s_g)^{-\sigma}}{\Gamma(1-\sigma)} . \quad (45a)$$

For a small but finite f, therefore, it turns out to be

$$D_g(s_0, s_g) \simeq \frac{i}{2} AKC\chi_g \frac{(ifs_0 s_g)^{-\sigma}}{\sqrt{-2\pi\sigma}(-\sigma)^{-\sigma} e^{\sigma}} . \quad (45b)$$

In fact, this expression is nothing else but the expression of the Eikonal theory, namely equation (32*b*) combined with equations (33*b*) and (34) for a sufficiently large $|s_0 s_g|$. Therefore, the relative error of the intensity in using equation (45*b*) instead of equation (45*a*) which is valid for a large $|fs_0 s_g|$ is

$$E \equiv \frac{|(45b)|^2 - |(45a)|^2}{|(45a)|^2} = \frac{|(-\sigma)^{\sigma} e^{-\sigma}|^2}{2\pi|\sigma|} |\Gamma(1-\sigma)|^2 - 1 . \quad (46a)$$

For non-absorbing cases where σ is a pure imaginary, one can see

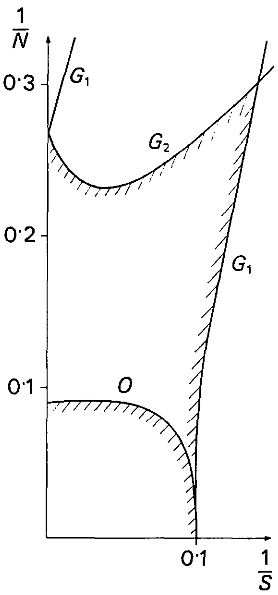

Fig. 2. The applicable ranges of the direct and the Bragg-reflected waves in the Eikonal theory. The curves G_1, G_2 and *O* correspond to equations (43*a*), (43*b*) and (44), respectively for $Q = \frac{1}{30}$. The parameters *N* and *S* are defined by equations (35).

$$E = \frac{1}{\exp\left(\frac{\pi}{2}|S|\right) - 1} . \quad (46b)^*$$

In the case of bent crystals and of the symmetrical Laue, the parameter $|S|$ has the form

$$|S| = \frac{4\pi^2 aR}{\Lambda_0^2} \quad (47)$$

where Λ_0 is the fringe spacing along the net plane in the perfect crystal, a and R are the spacing and the radius of curvature of the net plane respectively. If the value $(4aR/\Lambda_0^2)$ is about unity, the Eikonal theory is practically correct. This form of criterion has been suggested by Kato (1974) based on a naive wave-optical consideration. Taking reasonable figures $\Lambda_0 = 50\mu$m, $a = 1$ Å and $R = 2m$, the relative error is estimated to be 0·7%.

APPENDIX

The mathematical principles in deriving the asymptotic expansion of §4

The method discussed in this Appendix is nothing else but the steepest-descent method. The details can be obtained from Jeffereys & Jeffereys (1956).

Equation (25) can be rewritten in the form

$$D_h = \exp[\varphi(p_0)] \int A_h(p) \exp\left(-\frac{1}{2}\zeta^2\right) \frac{dp}{d\zeta} d\zeta \quad \text{(A-1)}$$

by letting

$$\varphi(p) = \varphi(p_0) - \frac{1}{2}\zeta^2 \quad \text{(A-2)}$$

in the vicinity of each saddle point p_0. In our problem p_0 stands for either one of $p^{(1)}$ and $p^{(2)}$. With the use of the power series

$$A_h(p)\frac{dp}{d\zeta} \equiv L(\zeta) = L(0) + L'(0)\zeta + \frac{1}{2}L''(0)\zeta^2 + \ldots \quad \text{(A-3)}$$

equation (A-1) can be written in the asymptotic expansion

$$D_h \simeq \sqrt{2\pi} \exp[\varphi(p_0)] \left\{ A_h(p_0) \left(\frac{dp}{d\zeta}\right)_{p=p_0} + \frac{1}{2}\left[\frac{d^2}{d\zeta^2}\left(A_h(p)\frac{dp}{d\zeta}\right)\right]_{p=p_0} + \ldots \right\} . \quad \text{(A-4)}$$

* Use the relations

$$|(-\sigma)^{-\sigma}|^2 = \exp(-\pi|\sigma|)$$
$$|\exp(-\sigma)|^2 = 1$$
$$|\Gamma(1-\sigma)|^2 = \pi|\sigma|/\sinh \pi|\sigma|$$

for a pure imaginary value of σ.

The first term in the braces is easily calculated as follows. By the differentiation of equation (A-2), one gets

$$\frac{\mathrm{d}^2}{\mathrm{d}\zeta^2}\varphi(p)=\varphi''(p)\left(\frac{\mathrm{d}p}{\mathrm{d}\zeta}\right)^2+\varphi'(p)\frac{\mathrm{d}^2p}{\mathrm{d}\zeta^2}=-1\,. \quad \text{(A-5)}$$

Since $\varphi'(p)$ is zero at the point $p=p_0$, one obtains

$$\left(\frac{\mathrm{d}p}{\mathrm{d}\zeta}\right)_{p=p0}=\left[\frac{-1}{\varphi''(p_0)}\right]^{1/2}. \quad \text{(A-6)*}$$

By the use of this relation, the first term of (A-4) gives equation (28).

The second term can be calculated with similar procedures. By the differential manipulation, one gets

$$\frac{\mathrm{d}^2}{\mathrm{d}\zeta^2}\left(A_h(p)\frac{\mathrm{d}p}{\mathrm{d}\zeta}\right)=A_h''(p)\left(\frac{\mathrm{d}p}{\mathrm{d}\zeta}\right)^3$$
$$+3A_h'(p)\frac{\mathrm{d}p}{\mathrm{d}\zeta}\frac{\mathrm{d}^2p}{\mathrm{d}\zeta^2}+A_h(p)\frac{\mathrm{d}^3p}{\mathrm{d}\zeta^3}. \quad \text{(A-7)}$$

The derivatives $(\mathrm{d}^2p/\mathrm{d}\zeta^2)_{p=p0}$ and $(\mathrm{d}^3p/\mathrm{d}\zeta^3)_{p=p0}$ can be successively determined from the relations derived by differentiating equation (A-5). Inserting these into (A-7), one obtains

$$\left[\frac{\mathrm{d}^2}{\mathrm{d}\zeta^2}\left(A(p)\frac{\mathrm{d}p}{\mathrm{d}\zeta}\right)\right]_{p=p0}=\left\{-\left(\frac{1}{\varphi''}\right)^{1/2}\left[-\frac{A_h''}{\varphi''}\right.\right.$$
$$\left.\left.+\frac{A_h'\varphi^{(3)}}{(\varphi'')^2}-\frac{5A_h(\varphi^{(3)})^2}{12(\varphi'')^3}+\frac{A_h\varphi^{(4)}}{4(\varphi'')^2}\right]\right\}_{p=p0} \quad \text{(A-8)}$$

where $\varphi^{(3)}$ and $\varphi^{(4)}$ imply $\mathrm{d}^3\varphi/\mathrm{d}p^3$ and $\mathrm{d}^4\varphi/\mathrm{d}p^4$ respectively. With this result, the second term of (A-4) gives equations (34).

* As to the arguments on the phase angle of the expression $[\;]^{1/2}$, see Jeffereys & Jeffereys (1956), p. 505.

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Largest Likely Values of Residuals

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The value of the residual $R_2=\langle(I_1-I_2)^2\rangle/\langle I_1^2\rangle$, where I_1 and I_2 are the intensities of reflexion for two unrelated structures having the same symmetry and containing the same atoms, has the value $1-\Sigma_4/(2\Sigma^2-\Sigma_4)$ for non-centrosymmetric structures and $\frac{4}{3}-2\Sigma_4/3(\Sigma^2-\Sigma_4)$ for centrosymmetric; the corresponding values for $R_1=\langle(F_1-F_2)^2\rangle/\langle F_1^2\rangle$ are approximately $2(1-\pi/4)\simeq 0{\cdot}43$ and $2(1-2/\pi)\simeq 0{\cdot}73$. More complex expressions are derived for hyper- and sesquisymmetric structures. If a residual with a scaling factor, such as $S_2=\langle(I_1-EI_2)^2\rangle/\langle I_1^2\rangle$, is used, and the scaling factor E is refined by least-squares, the value of E obtained is about $\frac{1}{2}$ or $\frac{1}{3}$, instead of the true value unity.

Introduction

Lenstra (1974) has considered the values of the residual

$$R_2=\frac{\sum(I_1-I_2)^2}{\sum I_1^2} \quad (1)$$

$$=\frac{\langle(I_1-I_2)^2\rangle}{\langle I_1^2\rangle}, \quad (2)$$

where, in Lenstra's application, I_1 represents the intensity of the hkl reflexion from a correct structure and I_2 represents the corresponding intensity calculated for a structure that is incomplete or in some way incorrect. Wilson (1969) had earlier considered the case in which the structures differed only through the misplacement of a single atom, and still earlier (Wilson, 1950) the case where the structures were entirely unrelated, except that they consisted of the same atoms and had the same symmetry; in this first paper the less convenient