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The Dynamical Theory of X-ray Bragg Diffraction from a Crystal with a Uniform Strain Gradient. The Green–Riemann Functions

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The dynamical problem of X-ray Bragg diffraction from a thick (semi-infinite) crystal deformed by a uniform strain gradient (USG) is treated on the basis of the Green–Riemann function formalism. The rigorous solution of the problem is formulated by means of the Huygens–Fresnel principle. The exact Green functions are obtained in the form of the Laplace integrals suitable in physical applications. The quasi-classical and the Born (kinematical) asymptotic expansions of the Green functions are constructed as functions of the effective USG parameter B. Special attention is paid to the analysis of the wave-field propagation in a crystal with USG. The spatial harmonics $\mathrm{Re}(q)$ of the diffracted Green function, when $\mathrm{Re}(qB) < 0$, as is shown, propagate within the proper 'waveguides', while the ones with $\mathrm{Re}(qB) > 0$ are damped exponentially in the bulk of the crystal. The Taupin problem of the Bragg dynamical diffraction of the X-ray incident plane wave from a thick crystal, the lattice spacing being a linear function of the coordinate z (along the inward normal to the entrance surface) only is solved exactly in analytical form. In the latter case the waveguide nature of the propagation of the spatial harmonics inside such a crystal, provided that $\mathrm{Re}(qB) < 0$, is clearly revealed.

1. Introduction

Recently (Petrashen', 1973; Chukhovskii, 1974; Petrashen' & Chukhovskii, 1975, 1976; Chukhovskii & Petrashen', 1977; see also Katagawa & Kato, 1974), the dynamical theory of X-ray Laue diffraction by a crystal with a uniform strain gradient (USG) has been developed on the basis of the Green–Riemann function method. It is of special interest to apply a similar approach to the study of X-ray propagation inside such a crystal in the case of Bragg diffraction.

However, for Bragg diffraction there is an additional factor in comparison with the Laue case, namely the X-ray extinction phenomenon: the total internal reflection of the wave field in the regions of a crystal where the exact Bragg condition is fulfilled. This seriously complicates the mathematical treatment of the problem under consideration.

Up to now, there have been no systematic investigations of X-ray Bragg diffraction by imperfect crystals and, in particular, by crystals with USG. Only a few works are known (Bonse, 1962, 1964; Bonse, Kappler & Shill, 1964; Taupin, 1964; Bonse & Graeff, 1973; Baturin, Koval'chuk, Kov'ev & Palapis 1977; Fukuhara & Takano, 1977) in spite of the importance of the topic for studies of the real structure of the crystal regions adjacent to the entrance surface.

Bonse (1964) elaborated the original generalization of the Penning & Polder (1961) theory. He took into account, in principle, the X-ray extinction phenomenon and calculated the energy flow trajectories in a homogeneously bent crystal. Nevertheless, the initial assumptions of the Bonse theory are not quite clear and the mathematical treatment of the problem is very complicated. Petrashen' (1973) first constructed the exact Riemann function in the case of X-ray Bragg diffraction from a crystal deformed by USG [when $\partial^2(\mathbf{hu})/\partial x_i \partial x_j = \text{const.}$ where $\mathbf{u}(\mathbf{r})$ is the elastic displacement field and $\mathbf{h}$ is the diffraction vector]. Unfortunately the Riemann function obtained is an infinite series of confluent hypergeometric functions and all the terms of the series have the same order in powers of the effective USG parameter $4B = \partial^2(\mathbf{hu})/\partial s_0 \partial s_h$ [hereafter, $(s_0 O s_h)$ is the oblique-angled coordinate system with axes Os_0, Os_h along the directions $\mathbf{K}_0$ of the incident beam and $\mathbf{K}_h$ of the diffracted beam in the X-ray scattering plane, respectively].

Thus, the principal point of the solution of the problem in question is to find the Green–Riemann functions suitable for a physical analysis. On the other hand, the main difficulty of the mathematical treatment is caused by the extinction phenomenon mentioned above, since, when the total reflection of the X-rays occurs, the imaginary part of the eikonal function

becomes of the same order as or greater than the real one and, therefore, the quasi-classical description of wave-field propagation fails, as does the eikonal theory.

In the present paper the general properties of Bragg dynamical diffraction of X-rays from a thick crystal with USG are discussed. On the basis of the modified Riemann method the solution of the Takagi equations is formulated in the integral form of the Huygens–Fresnel principle and the Green–Riemann functions are introduced (§§ 2, 3). The Laplace transforms of the exact Green–Riemann functions are found. They enable one to analyse the physical behaviour of the wave field inside the crystal with USG. It is interesting that the spatial harmonics Re(q) of the wave packet corresponding to the Green–Riemann function and describing the wave-field distribution in a crystal from a point source of X-rays propagate within 'waveguides' for Re(qB) < 0, undergoing multiple total reflection, whereas the harmonics Re(qB) > 0 decrease exponentially in the bulk of the crystal. This should also be true for the more complicated distortion fields.

In § 4 the quasi-classical and the Born (kinematical) asymptotes of the Green–Riemann functions are constructed.

The exact analytical solution of the Taupin (1964) problem is obtained. It concerns the Bragg diffraction of an X-ray plane wave incident on a thick crystal deformed so that the lattice spacing is a linear function of the crystal depth only (§ 5). In this case the waveguide nature of the propagation of each spatial harmonic Re(q) with Re(qB) < 0 is clearly revealed.

2. Formulation of problem. The diffracted wave field on the entrance surface of a semi-infinite crystal

Inside a crystal oriented, as whole, near the single regular Bragg position with the diffracted vector **h**, the X-ray wave field is formed of a coherent superposition of the transmitted wave $\mathscr{E}_0 \exp(i\mathbf{K}_0\mathbf{r})$ and the diffracted wave $\mathscr{E}_h \exp(i\mathbf{K}_h\mathbf{r})$ and is governed by the Takagi set of equations (see, *e.g.*, Petrashen', 1973; Chukhovskii & Petrashen', 1977):

$$\begin{cases} i\dfrac{\partial \mathscr{E}_0}{\partial s_0} + K\dfrac{\chi_0}{2\gamma_0}\mathscr{E}_0 + \sigma_{-h}\exp(i\mathbf{hu})\,\mathscr{E}_h = 0 \\ i\dfrac{\partial \mathscr{E}_h}{\partial s_h} + K\dfrac{\chi_0 - \alpha}{2|\gamma_h|}\mathscr{E}_h + \sigma_h\exp(-i\mathbf{hu})\,\mathscr{E}_0 = 0, \end{cases} \quad (K = |\mathbf{K}_0|). \tag{2.1}$$

The relationship between the oblique-angled coordinate system (s_0Os_h) and the Cartesian one (xOz), with the x axis along the crystal surface, is given by

$$\begin{cases} x = a_0 s_0 - a_h s_h \\ z = s_0 - s_h \end{cases} \qquad a_{0,h} = (1 - \gamma_{0,h}^2)^{1/2}/\gamma_{0,h}, \tag{2.2}$$

where γ_0 and γ_h are the direction cosines, $\gamma_{0,h} = \cos \angle(\mathbf{s}_{0,h}, \mathbf{n})$ and $\mathbf{n}$ is the inward normal to the entrance surface of the crystal (Fig. 1); $\alpha = (K_h^2 - K^2)/K^2$ is the angular deviation from the exact Bragg position. The complex dynamical coefficients are equal to

$$\begin{aligned} \sigma_{-h} &= \frac{\mathscr{C}K\chi_{-h}}{2\gamma_0} \equiv \frac{\pi}{\Lambda}\frac{1}{|\beta|^{1/2}}\left(\frac{\chi_{-h}}{\chi_h}\right)^{1/2}(1 + ik) \\ \sigma_h &= \frac{\mathscr{C}K\chi_h}{2|\gamma_h|} \equiv \frac{\pi}{\Lambda}|\beta|^{1/2}\left(\frac{\chi_h}{\chi_{-h}}\right)^{1/2}(1 + ik) \end{aligned} \tag{2.3}$$

(for details of the form of the equations and the notations used to see, *e.g.*, Petrashen' & Chukhovskii, 1975; we note that $k < 0$ by definition and usually $|k| \ll 1$).

The mixed boundary conditions of the problem under consideration in the approximation of a semi-infinite crystal are defined as follows

$$\mathscr{E}_0(x,z)|_{z=0} = \mathscr{E}_0^{(\mathrm{inc})}(x), \quad \mathscr{E}_{|h}(x,z)|_{z=\infty} = 0. \tag{2.4}$$

Here $\mathscr{E}_0^{(\mathrm{inc})}(x)$ is the amplitude of the X-ray incident wave on the entrance surface. The second condition of (2.4) for the diffracted wave amplitude is sufficient in the case of a thick absorbing crystal, when $\mu_0 L \gg 1$, where L is the crystal thickness, μ_0 is the X-ray absorption coefficient. This is usually realized in an X-ray Bragg diffraction experiment.

The projection of the displacement field $\mathbf{u}(\mathbf{r})$ on the diffraction vector **h** for a crystal with USG is the quadratic function in coordinates,

$$\mathbf{hu}(\mathbf{r}) = 2(As_0^2 + 2Bs_0s_h + Cs_h^2). \tag{2.5}$$

For simplicity, in (2.5) terms linear in the coordinates are omitted, since their inclusion only leads to renormalization of the Bragg angle.

According to the modified Riemann method (Petrashen', 1973) the solution of the boundary-value

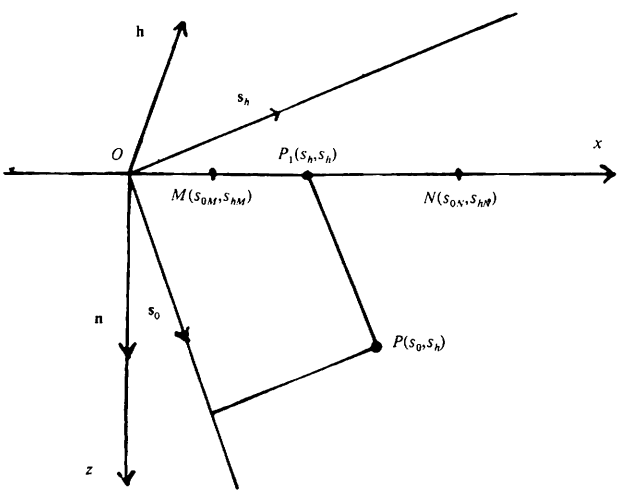

Fig. 1. The X-ray Bragg diffraction geometry and the coordinates used.

problem, (2.1), (2.4), (2.5), for the diffracted wave on the entrance surface can be written in the form

$$E_h(s_0,s_h)|_{s_0=s_h} = i\sigma_h \int_{OP_1} ds'_h E_0(s'_h, s'_h) \exp[-4iBs'^2_h + 4iBs_h(s'_h - s_h)] \tilde{R}_B(s'_h - s_h; s'_h - s_h), \quad (2.6)$$

where

$$E_h(s_0,s_h) = \mathscr{E}_h(s_0,s_h) \exp\left[-iK\left(\frac{\chi_0}{2\gamma_0}s_0 + \frac{\chi_0-\alpha}{2|\gamma_h|}s_h\right) + 2iAs_o^2\right] \quad (2.6^*)$$

$$E_0(s_0,s_h) = \mathscr{E}_0(s_0,s_h) \exp\left[-iK\left(\frac{\chi_0}{2\gamma_0}s_0 + \frac{\chi_0-\alpha}{2|\gamma_h|}s_h\right) - 2iCs_h^2\right].$$

Here the Riemann function $\tilde{R}_B(\xi'_0; \xi'_h)$ is determined from the hyperbolic equation ($\xi'_0 = s'_0 - s_0$, $\xi'_h = s'_h - s_h$; $\xi_0 = -\xi'_0$, $\xi_h = -\xi'_h$)

$$\frac{\partial^2 \tilde{R}_B}{\partial\xi'_0\partial\xi'_h} - 4iB\xi'_h\frac{\partial\tilde{R}_B}{\partial\xi'_h} + (\sigma_h\sigma_{-h} - 4iB)\tilde{R}_B = 0 \quad (2.7)$$

with the following mixed boundary conditions:

(i) on the characteristic $\xi'_0 = 0$

$$\tilde{R}_B|_{\xi'_0=0} = 1; \quad (2.8a)$$

(ii) on the entrance surface

$$\left(\frac{\partial\tilde{R}_B}{\partial\xi'_0} - 4iB\xi'_h\tilde{R}_B\right)\Big|_{\xi'_0=\xi'_h} = 0. \quad (2.8b)$$

Thus, the boundary-value problem (2.1), (2.4), (2.5), reduces to finding the Riemann function.

We shall construct the Riemann function in the form of the contour integral connected with the integral representation of the confluent hypergeometric function (Erdelyi, 1953)

$$\tilde{R}_B(\xi'_0; \xi'_h) = \int dt Q(\xi'_0 - t)\frac{t^\nu \exp(4iB\xi'_h t)}{\Gamma(1+\nu)}, \quad (2.9)$$

where $\nu = i\sigma_h\sigma_{-h}/4B$, $\Gamma(1+\nu)$ is the gamma function; $Q(t)$ is an unknown function, which has, in general, a branching at the point $t = 0$ of the type

$$Q(t)|_{t\to 0} \simeq t^{-1-\nu}.$$

The integration contour in (2.9) is a double loop, which begins at some point on the real axis, t_*, between the values 0 and ξ'_0, goes counter-clockwise round the point $t = \xi'_0$ and round the point $t = 0$, goes back to t_* and goes round the points ξ'_0 and 0 once again but clockwise and ends at the initial point, for which $\arg t_* = \arg(\xi'_0 - t_*) = 0$ by definition (Fig. 2).

Notice that the formal solution (2.9) satisfies (2.7) identically. Expanding the unknown function $Q(t)$ in a Taylor series

$$Q(t) = t^{-1-\nu}\sum_{m=0}^{\infty}\frac{d_m(4iB)^{m/2}}{\Gamma(m+\nu)}t^m$$

and finding the coefficients d_m from the recurrence relations which follow from (2.8), we get the Riemann function in the form of an infinite functional series of the confluent hypergeometric functions

$$\tilde{R}_B(\xi'_0; \xi'_h) = -\sum_{m=0}^{\infty} d_{2m}\frac{(4iB)^m}{(2m)!}{}_1F_1(1+\nu, 2m+1; 4iB\xi'_0\xi'_h).$$

However, the use of this solution for the physical investigation of the problem, (2.1), (2.4), (2.5), encounters significant difficulties. The point is that all the terms of the above series, as is easy to prove, beginning from the second one, are of the same order in powers of the USG parameter B.

In order to overcome this difficulty we write down the expression (2.6) for the amplitude of the diffracted wave as the convolution of the incident wave $E_0(s'_h, s'_h)$ multiplied by $\exp(-2iBs'^2_h)$ and the function which will be called the Green–Riemann function,

$$R_B(s'_0, s'_h; s_0, s_h) = \exp(4iBs_h\xi'_0)\tilde{R}_B(\xi'_0; \xi'_h)\theta(-\xi'_0) \quad (2.10)$$

multiplied by the phase factor

$$\exp[2iB(s_h^2 - s'^2_h)].$$

The boundary conditions for the Green–Riemann function take the form [*cf.* (2.8)]

$$R_B|_{s'_0=s_0} = 1 \quad (2.11a)$$

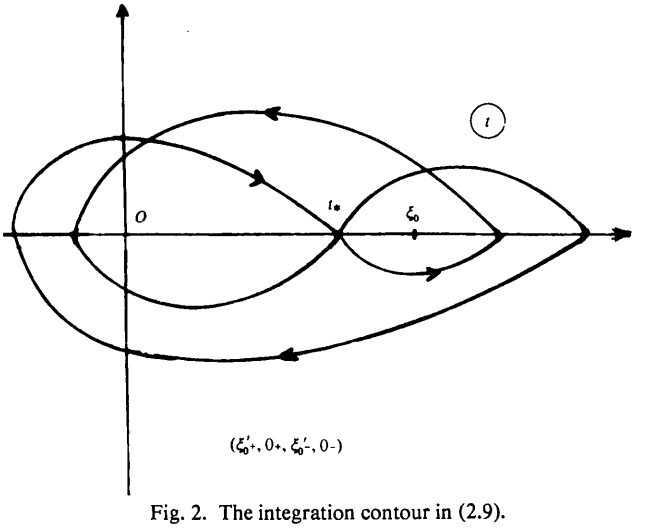

Fig. 2. The integration contour in (2.9).

$$\left(\frac{\partial R_B}{\partial s_0'} - 4iBs_h' R_B\right)\Big|_{s_0'=s_h'} = \delta(s_0 - s_0'). \quad (2.11b)$$

Now, taking into account (2.6) and (2.9)–(2.11) and making use of the direct and reverse Laplace transformations, we get after direct calculation

$$E_h(s_h, s_h) \exp(2iBs_h^2) = i\sigma_h \int_0^\infty ds_h' \exp(-2iBs_h'^2) E_0(s_h', s_h') \times \exp[2iB(s_h^2 - s_h'^2)] R_B(s_h', s_h'; s_h, s_h) \quad (2.12)$$

$$R_B(s_0', s_h'; s_0, s_h) = \exp[2iB\xi_0'(s_h + s_h')] \times \left(\frac{i}{4B}\right)^{1/2} \frac{1}{2\pi i} \int_{\mathrm{Re}\, p_0 - i\infty}^{\mathrm{Re}\, p_0 + i\infty} dp \exp[-p\xi_0' + p(\xi_0' - \xi_h')/2] \times \frac{D_{-1-\nu}\{(i/4B)^{1/2}[p - 2iB(\xi_0' - \xi_h')]\}}{D_{-\nu}\{(i/4B)^{1/2}[p + 2iB(\xi_0' - \xi_h')]\}}, \quad (2.13)$$

where $D_\nu(t)$ is the Weber (parabolic cylinder) function.

Notice that the Green–Riemann function (2.13) has a physical meaning only if $s_0 = s_h$ and describes the diffracted wave field on the entrance surface $z = 0$ in the case of an X-ray point source, *i.e.* $\mathscr{E}_0^{(\mathrm{inc})}(s_h) = \delta(s_h)$, as follows immediately from (2.12).

It should be emphasized that the Laplace representation idea of the kind in (2.13) can effectively be used for constructing the asymptotic Green–Riemann functions of interest to us for a wide range of parameters of the problem. This question will be discussed in detail below. Before proceeding to this, however, we shall find the proper Green functions (influence functions) of the problem. They are necessary if one is ever to describe the wave-field propagation inside a crystal.

3. The wave field inside a crystal. The Green functions

According to the Riemann method the amplitude of the diffracted wave at an arbitrary observation point (s_0, s_h) in a crystal is determined by the integral relation

$$E_h(s_0, s_h) = i\sigma_h \int_{OP_1} ds_h' \times \exp(-4iBs_h'^2) E_0(s_h', s_h') R_L(s_h', s_h'; s_0, s_h) + \int_{OP_1} ds_0' E_h(s_0', s_0') \left(\frac{\partial R_L}{\partial s_0'} - 4iBs_h' R_L\right)\Big|_{s_h'=s_0'}, \quad (3.1)$$

where $R_L(s_o', s_h'; s_0, s_h)$ is the Green function in the case of X-ray Laue diffraction and is equal to

$$R_L(s_0', s_h'; s_0, s_h) = \exp[4iBs_h(s_0' - s_0)] \times {}_1F_1[1 + \nu, 1; 4iB(s_0 - s_0')(s_h - s_h')]. \quad (3.2)$$

Then, inserting (2.6) into the second term of the right-hand side of (3.1) and utilizing the equality

$$\frac{\partial R_L}{\partial s_0'} - 4iBs_h' R_L = -4ivB(s_h - s_h') \times \exp[4iBs_h(s_0' - s_0)] \times {}_1F_1[1 + \nu, 2; 4iB(s_0 - s_0')(s_h - s_h')],$$

which follows from (3.2) and from the functional relations for the confluent hypergeometric functions, we write down (3.1) in the form

$$E_h(s_0, s_h) = i\sigma_h \int_{OP_1} ds_h' \exp(-4iBs_h'^2) E_0(s_h', s_h') \times \exp[4iBs_h(s_h' - s_0)] \times {}_1F_1[1 + \nu, 1; 4iB(s_0 - s_h')(s_h - s_h')] + 4\sigma_h \nu B \int_{OP_1} ds_0'(s_h - s_0') \exp[4iBs_h(s_0' - s_0)] \times {}_1F_1[1 + \nu, 2; 4iB(s_0 - s_0')(s_h - s_0')] \times \int_{OP_2(s_0', s_0')} ds_h'' \exp(-4iBs_h''^2) E_0(s_h'', s_h'') \times \exp[4iBs_0'(s_h'' - s_0')] \tilde{R}_B(s_0' - s_h''; s_0' - s_h''). \quad (3.3)$$

Bearing in mind that we are trying to find the Green function, which means that the amplitude of the diffracted wave (3.3) can be expressed as a convolution of the kind

$$E_h(s_0, s_h) = i\sigma_h \int_{-\infty}^{\infty} ds_h' \exp(-4iBs_h'^2) E_0(s_h', s_h') \times G_{B,h0}(s_0, s_h; s_h', s_h'), \quad (3.4)$$

we apply the integral Laplace transformation to both the sides of (3.3)

$$\mathscr{L}[\exp(2iBs_0 t) E_h(s_0, t); p] = i\sigma_h \mathscr{L}\{\exp[-2iBt(t + s_0 - s_h)] E_0(t, t); p\} \times [\mathscr{L}\{\exp[-2iBt(t + s_0 - s_h)] \times {}_1F_1[1 + \nu, 1; 4iBt(t + s_0 - s_h)]; p\} - 4ivB\mathscr{L}\{\exp[-2iBt(t + s_0 - s_h)] \tilde{R}_B(t; t); p\} \times \mathscr{L}\{t \exp[-2iBt(t + s_0 - s_h)] \times {}_1F_1[1 + \nu, 2; 4iBt(t + s_0 - s_h)]; p\}]. \quad (3.5)$$

The Laplace transforms of the expressions including the confluent hypergeometric functions, ${}_1F_1$, in the right-hand side of (3.5) can be calculated exactly (see Appendix 1). Therefore, we have

$$\mathscr{L}\{\exp[-2iBt(t+s_0-s_h)]$$
$$\times {}_1F_1[1+\nu,1;4iBt(t+s_0-s_h)];p\}$$
$$=\exp(-i\pi\nu/2)(i/4B)^{1/2}\exp[p(s_0-s_h)/2]$$
$$\times D_\nu\{(-i/4B)^{1/2}[p+2iB(s_0-s_h)]\}$$
$$\times D_{-1-\nu}\{(i/4B)^{1/2}[p-2iB(s_0-s_h)]\}$$
$$\mathscr{L}\{\exp[-2iBt(t+s_0-s_h)]$$
$$\times t_1F_1[1+\nu,2;4iBt(t+s_0-s_h)];p\}$$
$$=\exp(-i\pi\nu/2)\frac{1}{4B}\exp[p(s_0-s_h)/2]$$
$$\times D_{-1+\nu}\{(-i/4B)^{1/2}[p+2iB(s_0-s_h)]\}$$
$$\times D_{-1-\nu}\{(i/4B)^{1/2}[p-2iB(s_0-s_h)]\}. \tag{3.6}$$

Then, taking into account (2.13), (3.6) and the identity for the Weber functions,

$$D_{-\nu}(t)D_\nu(it)+i\nu D_{-1-\nu}(t)D_{-1+\nu}(it)=\exp(i\pi\nu/2),$$

which is also proved in Appendix 1, one finds from (3.5)

$$\mathscr{L}[\exp(2iBs_0t)E_h(s_0,t);p]=i\sigma_h$$
$$\times\mathscr{L}\{\exp[-2iBt(t+s_0-s_h)]E_0(t,t);p\}$$
$$\times(i/4B)^{1/2}\exp[p(s_0-s_h)/2]$$
$$\times\frac{D_{-1-\nu}\{(i/4B)^{1/2}[p-2iB(s_0-s_h)]\}}{D_{-\nu}\{(i/4B)^{1/2}[p+2iB(s_0-s_h)]\}}, \tag{3.7}$$

where $\mathscr{L}\{\exp[-2iBt(t+s_0-s_h)]E_o(t,t);p\}$ is the Laplace transform of the incident wave $E_0(t,t)$ with a phase shift corresponding to the displacement field (2.5).

Finally, the reverse Laplace transformation turns (3.7) into (3.4) with the Green function

$$G_{B,h0}(s_0,s_h;s_0',s_h')=\exp[-2iB\xi_0(s_h+s_h')]$$
$$\times\left(\frac{i}{4B}\right)^{1/2}\frac{1}{2\pi i}\int_{\mathrm{Re}\,p_0-i\infty}^{\mathrm{Re}\,p_0+i\infty}dp\exp[p\xi_h+p(\xi_0-\xi_h)/2]$$
$$\times\frac{D_{-1-\nu}\{(i/4B)^{1/2}[p-2iB(\xi_0-\xi_h)]\}}{D_{-\nu}\{(i/4B)^{1/2}[p+2iB(\xi_0-\xi_h)]\}}. \tag{3.8}$$

The analogous procedure for finding the Green function describing the transmitted wave is, in principle, feasible. But there is no necessity to carry it out. Consider the second equation of the set (2.1):

$$E_0(s_0,s_h)=-i\sigma_h^{-1}\exp(4iBs_0s_h)\frac{\partial E_h(s_0,s_h)}{\partial s_h},$$

from which follows

$$\exp(-4iBs_0s_h)E_0(s_0,s_h)$$
$$=\int_{-\infty}^{\infty}ds_h'\exp(-4iBs_h'^2)E_0(s_h',s_h')G_{B,00}(s_0,s_h;s_h',s_h'), \tag{3.9}$$

$$G_{B,00}(s_0,s_h;s_0',s_h')=\frac{\partial G_{B,h0}(s_0,s_h;s_0',s_h')}{\partial s_h}. \tag{3.10}$$

Now, keeping in mind (3.8) and the known recurrence relation for the Weber functions, we get

$$G_{B,00}(s_0,s_h;s_0',s_h')=\exp[-2iB\xi_0(s_h+s_h')]$$
$$\times\frac{1}{2\pi i}\int_{\mathrm{Re}\,p_0-i\infty}^{\mathrm{Re}\,p_0+i\infty}dp\exp[p\xi_h+p(\xi_0-\xi_h)/2]$$
$$\times\frac{D_{-\nu}\{(i/4B)^{1/2}[p-2iB(\xi_0-\xi_h)]\}}{D_{-\nu}\{(i/4B)^{1/2}[p+2iB(\xi_0-\xi_h)]\}}. \tag{3.11}$$

Notice that the Green function $G_{B,00}(s_0,s_h;\ s_0',s_h')$ becomes on the entrance surface

$$G_{B,00}(s_h,s_h;s_h',s_h')=\exp[-2iB(s_h^2-s_h'^2)]\,\delta(s_h-s_h')$$

and (3.9) reduces to

$$\mathscr{E}_0(s_h,s_h)=\mathscr{E}_0^{(\mathrm{inc})}(s_h)$$

as it must do.

The formulae (3.4), (3.9), (3.8), and (3.11) constitute the integral formulation of the Huygens–Fresnel principle for the determination of the exact wave field in the case of Bragg dynamical diffraction of X-rays from a crystal with USG and give a complete solution to the problem under consideration.

It is interesting that a comparison of (3.8) with (2.13) makes apparent the symmetry relation between the Green function $G_{B,h0}(s_0,s_h;\ s_0',s_h')$ and the Green–Riemann function $R_B(s_0',s_h';\ s_0,s_h)$. Indeed they are converted to one another by the interchange of the coordinates of the X-ray point source (s_0',s_h') and of the observation point (s_0,s_h). There is no accidental circumstance. Physically this is connected with the fact that the Green–Riemann function (2.13) is nothing but the Green function of the problem for the reciprocal geometry of the Bragg diffraction of X-rays by the crystal with a displacement field $\mathbf{hu}=4Bs_0s_h$ symmetrical with respect to the substitution $s_0\rightleftarrows -s_h$ (see Appendix II for details).

In the case of the 'dynamically transparent' crystal, when the inequality,

$$\frac{1}{2}\left|\frac{k}{B}\right|\frac{\pi^2}{A^2} \ll 1,$$

occurs, it is possible to put Re $\nu = 0$ in (3.8). Then the change of the sign of B is equivalent to the transition to the complex conjugate expression for the Green function. In this case the Friedel law is valid for the integral reflecting power from such a crystal.

In the opposite case of the 'dynamically absorbing' crystal, the Green function (3.8) essentially depends on the sign of B and the Friedel law does not hold.

It should be mentioned that for a crystal deformed by USG with $B = 0$ but $A \neq 0, C \neq 0$ (so-called case of the net plane 'fan') the Green function (3.8) and (3.11) can readily be shown to be equivalent, apart from a phase factor, to the corresponding expressions for a perfect crystal (Chukhovskii, Gabrielyan & Petrashen', 1976). Furthermore, the integral reflecting power is equal to

$$\mathscr{R}_h^I = 4|\sigma_h|^2 \int_{-\infty}^{\infty} \frac{d\alpha}{|q_0 + (q_0^2 - 4\sigma_h\sigma_{-h})^{1/2}|^2},$$

$$q_0 = \frac{K}{2}\left(\frac{\chi_0}{\gamma_0} + \frac{\chi_0 - \alpha}{|\gamma_h|}\right),$$

in this case, *i.e.* to the integral reflecting power of a perfect semi-infinite crystal (see, *e.g.*, Pinsker, 1974).

Now we shall show that the structure of the Green function (3.8) corresponds to waveguide propagation of the wave field in the bulk of a crystal with USG. The consideration below is directly similar to the analysis of the dynamical boundary-value problem of internal sphere oscillations (Smirnov, 1958).

Let us utilize the known functional relation for the Weber functions,

$$D_{-\nu}(t) = \exp(-i\pi\nu)\, D_{-\nu}(-t) + \frac{(2\pi)^{1/2}}{\Gamma(\nu)} \times \exp\left[\frac{i\pi}{2}(1-\nu)\right] D_{-1+\nu}(-it), \qquad (3.12)$$

[the case of $B > 0$ is considered; if $B < 0$ it is necessary to utilize the relation linking the functions $D_{-\nu}(t)$, $D_{-\nu}(-t)$ and $D_{-1+\nu}(it)$].

It is easy to see that on the integration line $p \equiv \mathrm{Re}\, p_0 - i\mathrm{Re}\, q$ in (3.8)

$$\frac{D_{-1+\nu}\{-i(i/4B)^{1/2}[p + 2iB(\xi_0 - \xi_h)]\}}{D_{-\nu}\{(-i/4B)^{1/2}[p + 2iB(\xi_0 - \xi_h)]\}}$$

does not exceed some number less than unity, provided that the integration variable $-\mathrm{Re}\, q$ is greater than $2B(\xi_0 - \xi_h)$ and the parameter $\mathrm{Re}\, p_0$ is large enough. Taking this into account, the integrand in (3.8) can be expanded as a functional series of the kind

$$G_{B,h0}(s_0, s_h; s_0', s_h') = \exp[-2iB\xi_0(s_h + s_h')]\left(\frac{i}{4B}\right)^{1/2}\frac{1}{2\pi i} \times \left\{ \int_{\mathrm{Re}\, p_0 - i\infty}^{\mathrm{Re}\, p_0 + 2iB(\xi_0 - \xi_h)} dp \exp[p\xi_h + p(\xi_0 - \xi_h)/2] \times \frac{D_{-1-\nu}(p_1)}{D_{-\nu}(p_2)} - \sum_{n=1}^{\infty}\left(-i\frac{(2\pi)^{1/2}}{\Gamma(\nu)}\right)^{n-1} \times \exp\left[i\frac{\pi\nu}{2}(n-1)\right] \times \int_{\mathrm{Re}\, p_0 + 2iB(\xi_0 - \xi_h)}^{\mathrm{Re}\, p_0 + i\infty} dp \exp(p\xi_h + p(\xi_0 - \xi_h)/2) \times \frac{D_{-1-\nu}(-p_1)}{D_{-\nu}(-p_2)}\left[\frac{D_{-1+\nu}(-ip_2)}{D_{-\nu}(-p_2)}\right]^{n-1} \times \left[1 + \frac{(2\pi)^{1/2}}{\Gamma(1+\nu)}\exp\left(i\pi\left\{1 + \frac{\nu}{2}\right\}\right)\frac{D_{\nu}(-ip_1)}{D_{-1-\nu}(-p_1)}\right]\right\},$$

$$p_{1,2} = (i/4B)^{1/2}[p \mp 2iB(\xi_0 - \xi_h)]. \qquad (3.13)$$

The fact of principal importance is that the finite number of terms on the right-hand side of (3.13) differs from zero for an observation point fixed inside a crystal. Indeed, by use of the standard asymptotic representations of the Weber functions, the integral corresponding to nth term of the sum on the right-hand side of (3.13) takes the form

$$\int_{\mathrm{Re}\, p_0 - i\infty}^{\mathrm{Re}\, p_0 + i\infty} dp\, \mathscr{F}_n(p, \xi_h, \xi_0 - \xi_h, \nu),$$

where

$$\mathscr{F}_n(p, \xi_h, \xi_0 - \xi_h, \nu) = \begin{cases} 0 \text{ for } -\mathrm{Re}\, q < 2B(\xi_0 - \xi_h) \\ p_1^{-1-\nu} p_2^{n(2\nu-1)-\nu+1} \times \exp\left(p\xi_h + \dfrac{n-1}{2}p_2^2\right) \times \left[1 + \dfrac{(2\pi)^{1/2}}{\Gamma(\nu)}\exp(i\pi\nu)\, p_1^{2\nu+1} \times \exp(p_1^2/2)\right] \\ \text{for } -\mathrm{Re}\, q > 2B(\xi_0 - \xi_h). \end{cases} \qquad (3.14)$$

From (3.14) it follows immediately that if the condition,

$$\xi_h + \frac{(1-n)}{2}(\xi_0 - \xi_h) + \frac{(1-n)}{2}\frac{(-\mathrm{Re}\, q)}{2B} < 0,$$

is satisfied, the terms of the sum in (3.13) beginning from the number $n = 2 + E[\xi_h/(\xi_0 - \xi_h)]$ vanish. The remaining terms describe the waves undergoing consequently 1, 2, ..., $(n - 1)$-times repeated reflections from the lower wall of the 'waveguide' inside a crystal.

For the physical analysis of the solution (3.4) there is the alternative approach based on the Laplace transforms of the Green functions. It may be fruitful, in particular, to calculate the diffracted wave amplitude (3.4).

With (3.7) in mind, the solution (3.4) can be written in the form of the Laplace integral:

$$E_h(s_0,s_h) = i\sigma_h \exp[-iB(s_0 + s_h)^2 + 2iBs_h^2] \times \left(\frac{i}{4B}\right)^{1/2} \frac{1}{2\pi i} \int_{\mathrm{Re}\, p_0 - i\infty}^{\mathrm{Re}\, p_0 + i\infty} \mathrm{d}p \exp[ps_h + p(s_0 - s_h)/2] \times \mathscr{L}[\exp(-2iBt^2) E_0(t,t); p] \times \frac{D_{-1-\nu}\{(i/4B)^{1/2}[p - 4iB(s_0 - s_h)]\}}{D_{-\nu}[(i/4B)^{1/2} p]}. \tag{3.15}$$

Further, we shall suppose that the poles of $\mathscr{L}[\exp(-2iBt^2) E_0(t,t); p]$ do not coincide with zeros of the Weber function $D_{-\nu}[\sqrt{i/4B}\, p]$. Then we get

$$E_h(s_0,s_h) = i\sigma_h \exp[-iB(s_0 + s_h)^2 + 2iBs_h^2] \times \left\{ E_h^{(*)}(s_0,s_h) + \sum_j \exp[p_j s_h + p_j(s_0 - s_h)/2] \times \mathscr{L}[\exp(-2iBt^2) E_0(t,t); p] \times \frac{D_{-1-\nu}\{(i/4B)^{1/2}[p_j - 4iB(s_0 - s_h)]\}}{\frac{\mathrm{d}}{\mathrm{d}p_j} D_{-\nu}[(i/4B)^{1/2} p_j]} \right\}, \tag{3.16}$$

where the function $E_h^{(*)}(s_0,s_h)$ corresponds to the sum of the residues at the poles of $\mathscr{L}[\exp(-2iBt^2) E_0(t,t); p]$. The series in the terms of the residues of the Laplace transform of the Green function at the points $D_{-\nu}[\sqrt{i/4B}\, p_j] = 0$ represents by itself the superposition of the real oscillations of the wave field in the crystal in question.

Notice that the situation is of physical interest when the pole (poles) of $\mathscr{L}[\exp(-2iBt^2) E_0(t,t); p]$ coincides (coincide) with the zero (zeros) of the Weber function $D_{-\nu}[\sqrt{i/4B}\, p]$. In this case the 'resonance' spatial harmonics, including the term linear in the coordinate s_h, appear on the right-hand side of (3.16).

4. The asymptotic Green functions

In physical applications the asymptotic Green functions in powers of the eikonal functions are certain to be very effective. In order to obtain adequate asymptotes we shall proceed from the Laplace integral representations of the Green functions (3.8) and (3.11).

In the case of large USG parameter $|B| \gg |\sigma_h \sigma_{-h}|$ the X-ray dynamical scattering is suppressed and the incident wave undergoes kinematical diffraction from the extremely deformed crystal. Then one can utilize the standard asymptotic representations for the Weber functions included in the Laplace transforms of the Green functions (3.8), (3.11). As a result, the latter acquire the form

$$G_{B,h0}(s_0,s_h; s_0',s_h') = \exp[-2iB\xi_0(s_h + s_h')] \left(\frac{i}{\pi 4B}\right)^{1/2} \frac{1}{2\pi i} \times \int_{\mathrm{Re}\, p_0 - i\infty}^{\mathrm{Re}\, p_0 + i\infty} \mathrm{d}p \exp(p\xi_h) p_1^{-1-\nu} p_2^{\nu} \times \left\{ \sum_{k=0}^{\infty} (-1)^k \Gamma(k + \tfrac{1}{2}) \left(\frac{(2\pi)^{1/2}}{p_1}\right)^{2k} + \varepsilon(1 + \nu, \arg p_1) \frac{(2\pi)^{1/2}}{\Gamma(1 + \nu)} \exp(p_1^2/2)\, p_1^{1+2\nu} - \varepsilon(\nu, \arg p_2) \frac{(2\pi)^{1/2}}{\Gamma(\nu)} \exp(p_2^2/2) p_2^{-1+2\nu} \right\} \tag{4.1}$$

$$G_{B,00}(s_0,s_h; s_0',s_h') = \exp[-2iB\xi_0(s_h + s_h')] \times \frac{1}{2\pi i} \int_{\mathrm{Re}\, p_0 - i\infty}^{\mathrm{Re}\, p_0 + i\infty} \mathrm{d}p \exp(p\xi_h) p_1^{-\nu} p_2^{\nu} \times \left\{ 1 + \varepsilon(\nu, \arg p_1) \frac{(2\pi)^{1/2}}{\Gamma(\nu)} \exp(p_1^2/2) p_1^{-1+2\nu} - \varepsilon(\nu, \arg p_2) \frac{(2\pi)^{1/2}}{\Gamma(\nu)} \exp(p_2^2/2) p_2^{-1+2\nu} \right\}. \tag{4.2}$$

The jump coefficients, $\varepsilon(1 + \nu, \arg p)$ and $\varepsilon(\nu, \arg p)$ determine the behaviour of the asymptotic expansions of $D_{-1-\nu}(p)$ and $D_{-\nu}(p)$ in adjacent sectors of $\arg p$ (the Stokes phenomenon) and are determined as

$$\varepsilon(\nu, \arg p) = \begin{cases} 0 & \text{for } |\arg p| < \pi/4 \\ -\exp(-i\pi\nu) & \text{for } \pi/4 \le \arg p < 5\pi/4 \\ -\exp(i\pi\nu) & \text{for } -5\pi/4 < \arg p \le -\pi/4. \end{cases}$$

The asymptotes (4.1), (4.2) describe the Born (kinematical) approximation of the Green functions in accordance with the terminology of the scattering theory. When $|B|$ increases, (4.1) and (4.2) tend to the

'kinematical' Green functions:

$$G_{B,h0}(s_0,s_h;\, s_0',s_h') = \exp[-4iBs_h'(s_0 - s_0')]\,\theta(s_h - s_h') \quad (4.3)$$

$$G_{B,00}(s_0,s_h;\, s_0',s_h') = \exp[-2iB(s_0 - s_0')(s_h + s_h')]\,\delta(s_h - s_h') \quad (4.4)$$

and in the limit of $|B| \to \infty$ the integrated intensity of the diffracted wave is equal to the corresponding (kinematical) value for an ideal-mosaic crystal

$$\mathscr{R}_h^i(|B| \to \infty) = \frac{2\gamma_0|\gamma_h|\,|\sigma_h|^2}{K^2\,\mathrm{Im}\,\chi_0(\gamma_0 + |\gamma_h|)}. \quad (4.5)$$

For the slightly deformed crystal, when $|B| \ll |\sigma_h\sigma_{-h}|$, in order to construct the quasi-classical expansion of the Green function (3.8) we shall use the Darwin method (see, *e.g.*, Miller, 1955). The direct calculations yield the following asymptotic representation of the Weber function of interest to us

$$D_{-1-\nu}(p) = \frac{(2\pi)^{1/4}}{[\Gamma(1+\nu)]^{1/2}} \exp[v_{1+\nu}(p^2)]\,\{\exp[-\theta_{1+\nu}(p)] + \varepsilon(1+\nu, \arg p)\exp[\theta_{1+\nu}(p)]\}$$

$$D_{-\nu}(p) = \frac{(2\pi)^{1/4}}{[\Gamma(\nu)]^{1/2}} \exp[v_{\nu}(p^2)]\,\{\exp[-\theta_{\nu}(p)] + \varepsilon(\nu, \arg p)\exp[\theta_{\nu}(p)]\}. \quad (4.6)$$

Here the functions $\theta_{1+\nu}(p)$, $\theta_\nu(p)$ and $v_{1+\nu}(p^2)$, $v_\nu(p^2)$ have the forms

$$\theta_{1+\nu}(p) = \tfrac{1}{4}p(p^2+4\nu)^{1/2} + (\nu+\tfrac{1}{2})\ln\frac{p+(p^2+4\nu)^{1/2}}{2\nu^{1/2}} + \sum_{s=1}^{\infty}\frac{d_{1+\nu,3s}(p)}{(p^2+4\nu)^{3s/2}}$$

$$\theta_{\nu}(p) = \tfrac{1}{4}p(p^2+4\nu)^{1/2} + (\nu-\tfrac{1}{2})\ln\frac{p+(p^2+4\nu)^{1/2}}{2\nu^{1/2}} + \sum_{s=1}^{\infty}\frac{d_{\nu,3s}(p)}{(p^2+4\nu)^{3s/2}}$$

$$v_{1+\nu}(p^2) = -\tfrac{1}{4}\ln(p^2+4\nu) + \sum_{s=1}^{\infty}\frac{f_{1+\nu,s}(p^2)}{(p^2+4\nu)^{s+1}}$$

$$v_{\nu}(p^2) = -\tfrac{1}{4}\ln(p^2+4\nu) + \sum_{s=1}^{\infty}\frac{f_{\nu,s}(p^2)}{(p^2+4\nu)^{s+1}}. \quad (4.7)$$

The functions $d_{1+\nu,3s}$; $d_{\nu,3s}$; $f_{1+\nu,s}$; $f_{\nu,s}$ can be calculated by use of the Darwin procedure (Miller, 1955). Therefore the first and the second $d_{1+\nu,3}$; $d_{\nu,3s}$; $f_{1+\nu,3s}$; $f_{\nu,3s}$ functions are equal to

$$d_{1+\nu,3} = d_{\nu,3} = -\frac{p}{4\nu}\left(\frac{p^2}{6} - \nu\right),$$

$$d_{1+\nu,6} = -d_{\nu,6} = \tfrac{5}{4}p(p^2+4\nu)^{1/2}$$

$$f_{1+\nu,1} = -f_{\nu,1} = -\tfrac{1}{2}, \quad f_{1+\nu,2} = f_{\nu,2} = \tfrac{5}{4}p^2.$$

As is seen from (4.6)–(4.7) [*cf.* the derivation of (3.13)] the spatial harmonics $\mathrm{Re}\,q$ ($p = \mathrm{Re}\,p_0 - i\,\mathrm{Re}\,q$) of the wave packets (3.8) and (3.11) are sharply differentiated from one another in terms of the physical nature of their propagation inside a crystal. Particularly, harmonics with $\mathrm{Re}(qB) < 0$ move in the proper 'waveguide' channels, whereas those with $\mathrm{Re}(qB) > 0$ are transmitted in the bulk of the crystal, being damped exponentially.

Now, if one neglects n-multiple reflections of spatial harmonics $\mathrm{Re}(qB) < 0$ from the lower walls of the appropriate 'waveguides', beginning from $n = 2$, the Green function (3.8) can be written as

$$G_{B,h0}(s_0,s_h;\, s_0',s_h') = \exp[-2iB\xi_0(s_h + s_h')]\,(\sigma_h\sigma_{-h})^{-1/2} \times \frac{1}{2\pi i}\int_{\mathrm{Re}\,p_0 - i\infty}^{\mathrm{Re}\,p_0 + i\infty} dp\,\exp[p\xi_h + p(\xi_0 - \xi_h)/2] \times \exp[v_{1+\nu}(p_1^2) - v_\nu(p_2^2) - \theta_{1+\nu}(p_1) + \theta_\nu(p_2)] \times \{1 + \varepsilon(1+\nu, \arg p_1)\exp[2\theta_{1+\nu}(p_1)] - \varepsilon(\nu, \arg p_2)\exp[2\theta_\nu(p_2)]\}. \quad (4.8)$$

The investigation of the problem based on the above quasi-classical asymptotic Green function will be carried out in a further paper. The particular case of X-ray Bragg diffraction by the crystal with the 'fan' of the net planes, when $B = 0$ but $A \neq 0$, $C \neq 0$, was treated by Chukhovskii, Gabrielyan & Petrashen' (1976) who took into account the formation of the caustic for the ray trajectories.

Notice that the formula (4.8) can readily be shown to be equivalent to (4.1) for large $|B| \gg |\sigma_h\sigma_{-h}|$. This in turn means that the quasi-classical Green function (4.8) can be used in physical applications for any USG.

5. The exact solution of the Taupin problem

Taupin (1964) treated numerically the problem of X-ray incident plane-wave Bragg diffraction from a thick (semi-infinite) crystal, the lattice spacing being a linear function of the crystal depth, when the displacement field $\mathbf{hu(r)}$ is given by

$$\mathbf{hu(r)} = -2Bz^2. \quad (5.1)$$

The dependence in (5.1) is a particular form of (2.5) when

$$A = -B = C. \quad (5.2)$$

The boundary conditions of the problem become [Taupin, 1964; *cf.* (2.4)]

$$\mathscr{E}_0(x,z)|_{z=0} = 1, \quad \mathscr{E}_h(x,z)|_{z=\infty} = 0. \quad (5.3)$$

The previous study makes it possible to find the exact analytical solution of the Taupin problem. With (5.2) and (5.3) in mind, it is not difficult to see that the

appropriate expressions for the diffracted and transmitted wave amplitudes are nothing but the exact Laplace transforms of the Green functions (3.8) and (3.11),

$$\mathscr{E}_h(s_0,s_h) = i\sigma_h \exp\left[i\frac{K}{4}(s_0 - s_h)\left(\frac{\chi_0}{\gamma_0} - \frac{\chi_0 - \alpha}{|\gamma_h|}\right) + iB(s_0 - s_h)^2 \right] + \left(\frac{i}{4B}\right)^{1/2} \frac{D_{-1-\nu}\{-i(i/4B)^{1/2}[q_0 + 4B(s_0 - s_h)]\}}{D_{-\nu}[-i(i/4B)^{1/2} q_0]} \tag{5.4}$$

$$\mathscr{E}_0(s_0,s_h) = \exp\left[i\frac{K}{4}(s_0 - s_h)\left(\frac{\chi_0}{\gamma_0} - \frac{\chi_0 - \alpha}{|\gamma_h|}\right) - iB(s_0 - s_h)^2 \right] \times \frac{D_{-\nu}\{-i(i/4B)^{1/2}[q_0 + 4B(s_0 - s_h)]\}}{D_{-\nu}[-i(i/4B)^{1/2} q_0]}. \tag{5.5}$$

The wave fields (5.4) and (5.5) do not depend on the coordinate along the crystal surface, as one would expect from general speculation. The ratio $\mathscr{E}_h/\mathscr{E}_0$ depends on, accurate to within a phase factor, the variable $q_0 + 4B(s_0 - s_h)$ only. This result was first obtained by Taupin (1964) and he used it effectively for the numerical calculation of the rocking curve.

The structure of the solutions (5.4) and (5.5) exhibits clearly the 'waveguide' nature of the Bragg diffraction of X-rays by a crystal with USG, as was pointed out above under the discussion of the general solution of the problem, (3.4), (3.8), (3.9), (3.11). Taking into account (4.8) [*cf.* (3.13)], one can prove that the spatial harmonics Re q_0 with $\mathrm{Re}(q_0 B) < 0$ undergo successive multiple reflections while those with $\mathrm{Re}(q_0 B) > 0$ are damped exponentially in the bulk of the crystal. As an example, Fig. 3 shows schematically several energy flow trajectories.

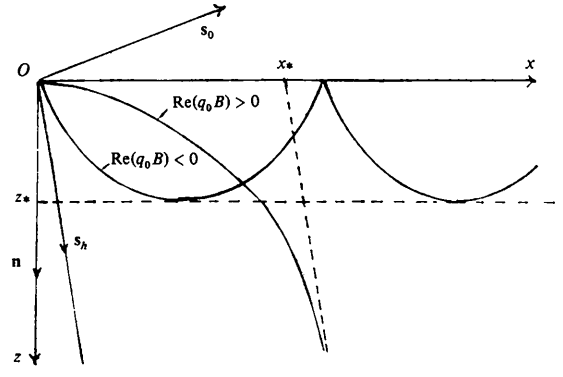

Fig. 3. Ray trajectories in Bragg case diffraction:

$x_* = \{|\mathrm{Re}\, q_0| - [(\mathrm{Re}\, q_0)^2 - 4|\sigma_h \sigma_{-h}|]^{1/2}\}/8|B|$
$z_* = (|\mathrm{Re}\, q_0| - 2|\sigma_h \sigma_{-h}|^{1/2})/4|B|.$

The angular distribution of the diffracted intensity has the form

$$\mathscr{R}_h(q_0) = |\mathscr{E}_h(s_h,s_h)|^2 = \frac{|\sigma_h|^2}{4|B|}\left| \frac{D_{-1-\nu}[-i(i/4B)^{1/2} q_0]}{D_{-\nu}[-i(i/4B)^{1/2} q_0]} \right|^2 \tag{5.6}$$

and, as a function of parameters included, is essentially determined by the variable of the kind,

$$q_0^2 - 4\sigma_h \sigma_{-h}. \tag{5.7}$$

Consider the case of $|B| \ll |\sigma_h \sigma_{-h}|$. The behaviour of the rocking curve (5.6) is sharply different in the regions $\mathrm{Re}(q_0 B) > 0$ and $\mathrm{Re}(q_0 B) < 0$. On the side of $\mathrm{Re}(q_0 B) > 0$ the curve $\mathscr{R}_h(q_0)$ coincides, accurately to within a magnitude of order

$$\left| \mathrm{Im} \frac{4B}{(q_0^2 - 4\sigma_h \sigma_{-h})^{1/2}} \right| \ll 1$$

with the corresponding function for a perfect crystal

$$\mathscr{R}_h(q_0) \simeq \left| -\frac{2\sigma_h}{q_0 + (q_0^2 - 4\sigma_h \sigma_{-h})^{1/2}} \right|^2. \tag{5.8}$$

In the region where $\mathrm{Re}(q_0 B) < 0$ it is necessary to take into account the X-ray 'waveguide' effect. Restricting ourselves to dealing with the single reflection of the wave field from the lower wall of the 'waveguide', we find

$$\mathscr{R}_h(q_0) \simeq \left| -\frac{2\sigma_h}{q_0 + (q_0^2 - 4\sigma_h \sigma_{-h})^{1/2}} \times \left\{ 1 - i\left(\frac{q_0^2 - 4\sigma_h \sigma_{-h}}{\sigma_h \sigma_{-h}}\right)^{1/2} \times \exp\left(-\frac{iq_0}{4B}(q_0^2 - 4\sigma_h \sigma_{-h})^{1/2} \right) \right\} \right|^2. \tag{5.9}$$

As can be seen, (5.9) oscillates, decreasing simultaneously as

$$\exp\left\{ \frac{\mathrm{Im}\, q_0}{2|B|} [(\mathrm{Re}\, q_0)^2 - 4|\sigma_h \sigma_{-h}|]^{1/2} \right\}.$$

The oscillation period is approximately equal to

$$\sim 2\pi \frac{4|B|}{[(\mathrm{Re}\, q_0)^2 - 4|\sigma_h \sigma_{-h}|]^{1/2}}. \tag{5.10}$$

It should be mentioned that the angular range of the X-ray extinction where the incident beam is totally reflected is rather insensitive to the USG parameter B in the area where $|B| \ll |\sigma_h \sigma_{-h}|$ and is practically the same as for a perfect crystal.

In the case of $|B| \gg |\sigma_h \sigma_{-h}|$ to analyse (5.6) it is necessary to use (4.1). The general type of the rocking curve is conserved. At the same time the position of $\max\{\mathscr{R}_h(q_0)\}$ shifts proportional to $\sim |B|^{1/2}$ and the maximum itself *versus* B decreases, as

$$\max[\mathscr{R}_h(q_0)] \sim |B|^{-1}. \tag{5.11}$$

The oscillation period increases linear with $|B|$.

Notice that the exponential decrease of the oscillating part of (5.9) is connected with the absorption of the wave passing along an additional path in the crystal, due to its reflection from the lower wall of the waveguide, with respect to the directly diffracted wave. Because of this, the twofold and successive multiple reflections of the wave field do not practically affect the rocking curve (5.9). Besides, for the $|B| \gg |\sigma_h \sigma_{-h}|$ the crystal depth, $(s_0 - s_h) \simeq |B|^{-1} |\sigma_h \sigma_{-h}|$, where the X-ray total reflection occurs, is much smaller than the extinction distance Λ and now the angle range $|\mathrm{Re} q_0| < 2|\sigma_h \sigma_{-h}|^{1/2}$ can no longer be distinguished.

APPENDIX I

At this point we shall derive (i) the formulae (3.6) for the Laplace transforms of the expressions including the confluent hypergeometric functions; (ii) the identity for the Weber functions, which was used in order to obtain (3.7) in the basic text.

(i) Let us consider the known integral representation of the confluent hypergeometric function (Erdelyi, 1953):

$$ {}_1F_1(a,c;\lambda t) = \frac{\Gamma(c)}{\Gamma(a)\Gamma(c-a)} \times t^{1-c} \int_0^t dx \exp(\lambda x) x^{a-1}(t-x)^{c-a-1}. \tag{AI.1}$$

This is correct under the conditions

$$\mathrm{Re}\, c > \mathrm{Re}\, a > 0. \tag{AI.2}$$

Now, assuming the particular value of the parameter,

$$\lambda = 4iB(t + s_0 - s_h), \tag{AI.3}$$

one writes down the integral (AI.1) as the convolution

$$\begin{aligned} &t^{c-1} \exp[-2iBt(t + s_0 - s_h)] \\ &\times {}_1F_1[a,c; 4iBt(t + s_0 - s_h)] \\ &= \frac{\Gamma(c)}{\Gamma(a)\Gamma(c-a)} \int_0^t dx \exp[2iBx(x + s_0 - s_h)]\, x^{a-1} \\ &\times \exp[-2iB(t-x)(t - x + s_0 - s_h)](t-x)^{c-a-1}. \end{aligned} \tag{AI.4}$$

Then the Laplace transformation of (AI.4) is readily performed on the basis of the tabulated Laplace transforms. As a result, one has

$$\begin{aligned} &\mathscr{L}\{t^{c-1} \exp[-2iBt(t + s_0 - s_h)] \\ &\times {}_1F_1[a,c; 4iBt(t + s_0 - s_h)]; p\} \\ &= \exp(-i\pi a/2)\Gamma(c)\left(-\frac{i}{4B}\right)^{c/2} \\ &\times \exp[p(s_0 - s_h)/2] D_{-1}\{(i/4B)^{1/2}[p - 2iB(s_0 - s_h)]\} \\ &\times D_{a-c}\{(-i/4B)^{1/2}[p + 2iB(s_0 - s_h)]\}. \end{aligned} \tag{AI.5}$$

(AI.5) gives formulae (3.6) in the text, if one puts, respectively,

$$\begin{aligned} a &= 1 + \nu, \quad c = 1 \\ a &= 1 + \nu, \quad c = 2. \end{aligned} \tag{AI.6}$$

The restrictions (AI.2) imposes on the parameters a,c can be removed either by means of the analytical continuation principle or by the introduction of contour integrals, as was done in § 2 when the Riemann function was constructed. Thus (3.6) are valid for arbitrary values of the USG parameter B and of the complex dynamical coefficients σ_h, σ_{-h}.

(ii) In order to prove the identity for the Weber functions,

$$D_{-\nu}(t) D_{\nu}(it) + i\nu D_{-1+\nu}(it) D_{-1-\nu}(t) = \exp(i\pi\nu/2), \tag{AI.7}$$

it is easiest to differentiate the left-hand side of (AI.7) with respect to t and, by use of the known functional relations for the Weber functions (Miller, 1955),

$$\begin{aligned} &\frac{d}{dt} D_{\nu}(t) + \tfrac{1}{2} t D_{\nu}(t) - \nu D_{-1+\nu}(t) = 0 \\ &\frac{d}{dt} D_{\nu}(t) - \tfrac{1}{2} t D_{\nu}(t) + D_{1+\nu}(t) = 0, \end{aligned}$$

one finds that the derivative of the left-hand side of (AI.7) is identically equal to zero. To calculate the constant value in the right-hand side of (AI.7) the Weber functions at $t = 0$ can be used:

$$D_{\nu}(0) = \frac{2^{\nu/2}\pi^{1/2}}{\Gamma\left(\frac{1-\nu}{2}\right)}.$$

In this case, (AI.7) is obtained, as was to be shown.

Combining (3.6) and (AI.7), we get the formulae (3.7), and (3.8).

APPENDIX II

Here we treat the symmetry relation connecting the Green function $G_{B,h0}(s_{0N},s_{hN};\ s_{0M},s_{hM})$ and the Green–Riemann function $R_B(s_{0M},s_{hM};\ s_{0N},s_{hN})$.

The Green function $G_{B,h0}(s_0,s_h;\ s_{0M},s_{hM})$, describing the X-ray diffracted wave in a crystal with any displacement field $\mathbf{hu}(\mathbf{r})$, satisfies the following equation as a function of observation point coordinates (s_0,s_h) (see, *e.g.*, Courant, 1962)

$$\frac{\partial^2 G_{B,h0}}{\partial s_0\,\partial s_h} + i\,\frac{\partial(\mathbf{hu})}{\partial s_0}\,\frac{\partial G_{B,h0}}{\partial s_h} + \sigma_h\sigma_{-h}\,\theta(s_0 - s_h)\,G_{B,h0} = \delta(s_0 - s_{0M})\,\delta(s_h - s_{hM}). \tag{AII.1}$$

According to the causality principle, we define $G_{B,h0}$ as the retarded Green function:

$$G_{B,h0}(s_0,s_h;\ s_{0M},s_{hM})$$
is equal to zero for $s_0 \geq s_{0M},\ s_h \geq s_{hM}$
and differs from zero for $s_0 < s_{0M},\ s_h < s_{hM}$. (AII.2)

Then the Green functions should obey the mixed boundary conditions

$$G_{B,h0}\Big|_{s_h = s_{hM}} = \exp\left[i\mathbf{hu}(s_{0M},s_{hM}) - i\mathbf{hu}(s_0,s_{hM})\right]$$
$$\frac{\partial G_{B,h0}}{\partial s_h}\Big|_{s_0 = s_h} = \exp\left[i\mathbf{hu}(s_{0M},s_h) - i\mathbf{hu}(s_h,s_h)\right]\delta(s_h - s_{hM}). \tag{AII.3}$$

On the other hand, the Green–Riemann function $R_B(s_0,s_h;\ s_{0N},s_{hN})$ is, apart from a phase factor, nothing but the Green function $\tilde{G}_{B,h0}(s_0,s_h;\ s_{0N},s_{hN})$ of the problem in the reciprocal geometry of the diffraction:

$$R_B(s_0,s_h;\ s_{0N},s_{hN}) = \exp\left[i\mathbf{hu}(s_0,s_h) - i\mathbf{hu}(s_{0N},s_{hN})\right] \times \tilde{G}_{B,h0}(s_0,s_h;\ s_{0N},s_{hN}). \tag{A.II.4}$$

The latter is defined as the solution of the equation

$$\frac{\partial^2 \tilde{G}_{B,h0}}{\partial s_0\,\partial s_h} + i\,\frac{\partial(\mathbf{hu})}{\partial s_h}\,\frac{\partial \tilde{G}_{B,h0}}{\partial s_0} + \sigma_h\sigma_{-h}\,\theta(s_0 - s_h)\,\tilde{G}_{B,h0} = \delta(s_0 - s_{0N})\,\delta s_h - s_{hN}) \tag{AII.5}$$

with the boundary conditions

$$\tilde{G}_{B,h0}|_{s_0 = s_{0N}} = \exp\left[i\mathbf{hu}(s_{0N},s_{hN}) - i\mathbf{hu}(s_{0N},s_h)\right]$$
$$\frac{\partial \tilde{G}_{B,h0}}{\partial s_0}\Big|_{s_0 = s_h} = -\exp\left[i\mathbf{hu}(s_0,s_{hN}) - i\mathbf{hu}(s_0,s_0)\right] \times \delta(s_0 - s_{0N}). \tag{AII.6}$$

Now, if one goes over to the 'proper' coordinate system for X-ray Bragg diffraction in the reciprocal geometry, namely: to the system $\tilde{s}_0 O \tilde{s}_h$, related to the initial one by

$$\tilde{s}_0 = -s_h$$
$$\tilde{s}_h = -s_0, \tag{AII.7}$$

(AII.5) and (AII.6) can be written in the form

$$\frac{\partial^2 \tilde{G}_{B,h0}}{\partial \tilde{s}_0\,\partial \tilde{s}_h} + i\,\frac{\partial[\mathbf{hu}(-\tilde{s}_h,-\tilde{s}_0)]}{\partial \tilde{s}_0}\,\frac{\partial \tilde{G}_{B,h0}}{\partial \tilde{s}_h} + \sigma_h\sigma_{-h}\,\theta(\tilde{s}_0 - \tilde{s}_h)\,\tilde{G}_{B,h0} = \delta(\tilde{s}_0 + s_{hN})\,\delta(\tilde{s}_h + s_{0N}) \tag{AII.8}$$

$$\tilde{G}_{B,h0}|_{-\tilde{s}_h = s_{0N}} = \exp\left[i\mathbf{hu}(s_{0N},s_{hN}) - i\mathbf{hu}(s_{0N},-\tilde{s}_0)\right]$$
$$\frac{\partial \tilde{G}_{B,h0}}{\partial \tilde{s}_h}\Big|_{\tilde{s}_0 = \tilde{s}_h} = \exp\left[i\mathbf{hu}(-\tilde{s}_h,s_{hN}) - i\mathbf{hu}(-\tilde{s}_h,-\tilde{s}_h)\right] \times \delta(\tilde{s}_h + s_{0N}). \tag{AII.9}$$

From comparison of (AII.1), (AII.3) with (AII.8), (AII.9) it follows that for the displacement fields with

$$\mathbf{hu}(-\tilde{s}_h,-\tilde{s}_0) = \mathbf{hu}(s_0,s_h) \tag{AII.10}$$

the Green functions $G_{B,h0}$, $\tilde{G}_{B,h0}$ of the problems in the standard and the reciprocal geometries are the same when $s_{0N} \rightleftarrows -s_{hM}$, $s_{hN} \rightleftarrows -s_{0M}$, *i.e.*

$$G_{B,h0}(s_{0N},s_{hN};\ s_{0M},s_{hM}) = \tilde{G}_{B,h0}(-s_{hN},-s_{0N};\ -s_{hM},-s_{0M}) \tag{AII.11}$$

if they are written in the same initial coordinate system.

Finally, in the particular case of the displacement field $\mathbf{hu} = 4Bs_0 s_h$, taking into account (AII.4) and (AII.11), one can find the exact Green function if the Green–Riemann function is constructed [*cf.* (3.8) and (2.13)] by means of the symmetry relation proved above:

$$G_{B,h0}(s_{0N},s_{hN};\ s_{0M},s_{hM}) = \exp\left(4iBs_{0M}s_{hM} - 4iBs_{0N}s_{hN}\right) \times R_B(-s_{hN},-s_{0N};\ -s_{hM},-s_{0M}).$$

References

BATURIN, V. E., KOVAL'CHUK, M. V., KOV'EV, E. K. & PALAPIS, V. E. (1977). *Kristallografiya*, **22**, 144–148.

BONSE, U. (1962). *Direct Observation of Imperfections in Crystals*, edited by J. B. NEWKIRK & J. H. WERNICK, pp. 431–467. New York/London: Interscience.

BONSE, U. (1964). *Z. Phys.* **177**, 385–423.

BONSE, U. & GRAEFF, W. (1973). *Z. Naturforsch. Teil A*, **28**, 558–564.

BONSE, U., KAPPLER, E. & SHILL, A. (1964). *Z. Phys.* **178**, 221–231.

CHUKHOVSKII, F. N. (1974). *Kristallografiya*, **19**, 482–488:

CHUKHOVSKII, F. N., GABRIELYAN, K. T. & PETRASHEN', P. V. (1976). *Proc. IV Conf. on the Dynamical Effects of the Scattering of X-rays and Electrons*, Leningrad, pp. 17–20. (In Russian.)

CHUKHOVSKII, F. N. & PETRASHEN', P. V. (1977). *Acta Cryst.* A**33**, 311–319.

COURANT, R. (1962). *Partial Differential Equations*. New York/London: Interscience.

ERDELYI, A. (1953). *Higher Transcendental Functions*. Vol. I. New York: McGraw-Hill.

FUKUHARA, A.. & TAKANO, Y. (1977). *Acta Cryst.* A**33**, 137–142.

KATAGAWA, T. & KATO, N. (1974). *Acta Cryst.* A**30**, 830–836.

MILLER, J. C. P. (1955). *Tables of Weber Parabolic Cylinder Functions*. London: Her Majesty's Stationery Office.

PENNING, P. & POLDER, D. (1961). *Philips Res. Rep.* **16**, 419–440.

PETRASHEN', P. V. (1973). *Fiz. Tverd. Tela*, **15**, 3131–3132.

PETRASHEN', P. V. & CHUKHOVSKII, F. N. (1975). *Zh. Eksp. Teor. Fiz.* **69**, 477–487.

PETRASHEN', P. V. & CHUKHOVSKII, F. N. (1976). *Kristallografiya*, **21**, 283–292.

PINSKER, Z. G. (1974). *The X-ray Dynamical Scattering by Perfect Crystals*. Moscow: Nauka. (In Russian.)

SMIRNOV, V. I. (1958). *Course of Higher Mathematics*, Vol. IV, pp. 795–806. Moscow: GIPML. (In Russian.)

TAUPIN, D. (1964). *Bull. Soc. Fr. Minéral. Cristallogr.* **87**, 469–514.

Acta Cryst. (1978). A**34**, 621–625

A Simple Refinement of Density Distributions of Bonding Electrons. III. Experimental Static Electron Densities for the Diborane Molecule

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From recently refined models of the electron density distribution in the diborane molecule, static density sections are calculated and presented as difference densities ρ(molecule at rest) $-$ ρ(isolated atoms at rest). The sections obtained are compared with corresponding ones derived from quantum-chemical calculations by Laws, Stevens and Lipscomb.

Introduction

In a preceding paper we described the refinement of two models of the electron density distribution in the diborane molecule (Mullen & Hellner, 1977), where the X-ray data, collected at 90 K, of Smith & Lipscomb (1965) were used. In this paper we shall compare the results of our refinement with the results of quantum-chemical calculations. For diborane, two SCF calculations were carried out by Laws, Stevens & Lipscomb (1972) (LSL), one with a minimum basis of 18 Slater-type orbitals (STO's), and one with an expanded basis of 68 STO's. Since the quantum-chemical calculations of LSL were performed for the static density of the equilibrium configuration of the molecule, we have transformed our experimentally obtained, dynamic densities (*i.e.* densities including the effects of the thermal motions of the atoms) into static densities.

Thermal deconvolution and series termination

Deconvolution of the dynamic densities for thermal smearing can be exactly performed (in the convolution approximation), if the temperature factors for all density units of the model are known. In actual practice we can assume this if the temperature factors were determined with the highest possible accuracy of the present-day methods (see below). However, the static density distribution, obtained by Fourier synthesis with structure factors, is disturbed by series-termination errors. The peaks that can be observed in difference density maps are broadened and reduced in height (Scheringer, 1977*a*). We cannot overcome the effect of series termination since the measured data are always limited, and thus we cannot reconstruct the true density distribution in the molecule, although we could reconstruct a more accurate representation of the refined density model.*

* Since the structure factors are calculated from a static density model, it is possible to calculate more structure factors than correspond to the experimental limit of $(\sin\theta)/\lambda$. With such a set of structure factors a higher degree of resolution would be suggested than is actually given by the diffraction experiment. Certain (large gradient) details in the static density maps would then be artifacts. Therefore the series should be terminated at the experimental limit of $(\sin\theta)/\lambda$ (Dietrich & Scheringer, 1978).