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An Exact Solution in the Dynamical Diffraction Theory of Lamellarly Distorted Crystals

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Abstract

An exact solution of dynamical diffraction was found for a lamellarly distorted infinite crystal. Here the deviation of the lattice spacing was assumed to have the form $D_0 \tanh \alpha x$ where the spatial variable x is normal to the net plane concerned. The problem is reduced to solving an ordinary differential equation of second order. The linearly independent solutions (LIS) are represented in a very symmetric manner with the use of the U function defined here. They are essentially hypergeometric functions with three complex parameters which are specified in terms of the diffraction condition and the lattice distortion. The analytical properties of the LIS's and their physical interpretation are described. Rocking curves are calculated for both the Darwin and Ewald cases. The theory can be applied to a variety of monotonic lattice distortions.

1. Introduction

An exact analytic solution of dynamical diffraction is desirable for understanding the physics of diffraction phenomena in distorted crystals. So far, such exact solutions have been obtained in the case of the so-called constant strain gradient, both for the Laue geometry (Katagawa & Kato, 1974; Chukhovski & Petrashen', 1977) and the Bragg geometry (Chukhovski, Gabrielyan & Petrashen', 1978). However, since the assumed distortion extends over the crystal, the solution is not very useful in cases where the distortion is confined to a certain region.

Bearing this situation in mind, we shall deal with the case of a stratified distortion sandwiched between two perfect crystals which have different lattice spacings. (A more specific description will be given in § 3.) Moreover, we shall consider a distortion which changes only along the normal of the crystal surface. Although the assumed conditions seem rather stringent, we often encounter such cases in the study of solid-state devices, so that the author believes that the problem is not merely of mathematical interest.

Since our problem is essentially one-dimensional, the basic equation can be reduced to an ordinary differential equation of second order. The two linearly independent solutions (LIS) are represented by the U function introduced here, which is essentially a hypergeometric function (HGF). It includes three parameters, a, b and c, besides the independent variable z as described in any standard text book.* In our solution, the parameters are defined by physical quantities such as the deviation from the exact Bragg condition, the absorption coefficient and the magnitude and gentleness of the lattice expansion as well as the scattering strength of the net plane (structure factor), and the variable z is a function of the spatial variable x normal to the net plane.

Recently, Bensoussan, Malgrange & Sauvage-Simkin (1987) and Authier, Gronkowski & Malgrange (1989) worked out numerical calculations on a similar problem. Needless to say, the analytical solution gives more physical insight. In fact, the solution can be interpreted by the concept of dispersion surface in the perfect crystal. Moreover, once the two LIS in infinite space have been obtained for the basic equation, one can construct any concrete wave field in parallel-sided crystals and half-infinite crystals simply by taking their linear combination. Also, as discussed in § 7, one can generalize the theory to some extent for dealing with a wider range of lattice distortions.

In this paper, it is mainly the theoretical aspects of the solution that are discussed with an emphasis on the Bragg reflectivity (rocking curves). Some concrete (numerical) results and an application to the standing-wave method will be presented in later papers.

2. The basic equation

We shall write the crystal wave as

$$O \text{ wave:} \quad d_o(\mathbf{r}) \exp(i\bar{\mathbf{K}}_o.\mathbf{r}) \qquad (2.1a)$$

$$G \text{ wave:} \quad d_g(\mathbf{r}) \exp(i\bar{\mathbf{K}}_g.\mathbf{r}) \qquad (2.1b)$$

where $\bar{\mathbf{K}}_o$ and $\bar{\mathbf{K}}_g = \bar{\mathbf{K}}_o + 2\pi\bar{\mathbf{g}}$ are the kinematical wave vectors satisfying the Bragg condition exactly, and $\bar{\mathbf{g}}$ is the reciprocal-lattice vector (RLV) for a reference perfect crystal. Then the $\mathbf{r}$-dependent amplitudes, d_o

* For example, see the books edited by Abramowitz & Stegun (1964) and Bateman-Erdelyi (1953), which are referred to as AS and BE respectively in this paper.

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and d_g, obey the differential equation of Takagi-Taupin type (Takagi, 1962, 1969; Taupin, 1964; Kato, 1973)

$$\left(\frac{\partial}{\partial t}-c\frac{\partial}{\partial x}\right)d_o=-(\mu_c/2)d_o+i(M\exp i\varphi)d_g \quad (2.2a)$$

$$\left(\frac{\partial}{\partial t}+c\frac{\partial}{\partial x}\right)d_g=-(\mu_c/2)d_g+i(M\exp -i\varphi)d_o \quad (2.2b)$$

where the t axis is parallel to the net plane and the x axis is taken along the RLV. The parameter c is defined by $\tan\theta_B$, θ_B being the Bragg angle. μ_c is the linear absorption coefficient divided by $\cos\theta_B$, M is proportional to the structure factor $|F_{\pm g}|$ and φ is called the lattice phase, which will be defined later by (3.1).

Through the unitary transformation

$$u(t,x)=\exp(-i\varphi/2)d_o \quad (2.3a)$$

$$v(t,x)=\exp(i\varphi/2)d_g, \quad (2.3b)$$

(2.2) may be rewritten in the form

$$\left[\frac{\partial}{\partial t}+(i/2)\frac{\partial\varphi}{\partial t}\right]u-c\left[\frac{\partial}{\partial x}+(i/2)\frac{\partial\varphi}{\partial x}\right]u$$
$$=(-\mu_c/2)u+iMv \quad (2.4a)$$

$$\left[\frac{\partial}{\partial t}-(i/2)\frac{\partial\varphi}{\partial t}\right]v+c\left[\frac{\partial}{\partial x}-(i/2)\frac{\partial\varphi}{\partial x}\right]v$$
$$=(-\mu_c/2)v+iMu. \quad (2.4b)$$

In this paper, we shall consider a special form of the phase; namely $\varphi=\varphi(x)$. This restriction may be relaxed to some extent. Any function $\varphi_c(t,x)$ can be added to φ, provided that

$$\left(\frac{\partial^2}{\partial t^2}-c^2\frac{\partial^2}{\partial x^2}\right)\varphi_c=0 \quad (2.5)$$

is satisfied since, then, the decoupled equation (2.4) is not changed. More important is that our treatment can be generalized to the case in which $\varphi=\varphi(X)$ where X is the normal coordinate to the crystal surface (see § 7). Meanwhile, however, implicitly we assume the symmetrical Bragg geometry.

Further, we shall consider the (mixed) Fourier component with respect to the variable t instead of the wave function itself. For this purpose we shall replace $u(t,x)$ by the following scheme:

$$u(t,x)\Rightarrow\exp(-iEt)u(E,x) \quad (2.6a)$$

$$v(t,x)\Rightarrow\exp(-iEt)v(E,x). \quad (2.6b)$$

Then, (2.4) is transformed to

$$\left\{\varepsilon-ic\left[\frac{\partial}{\partial x}+(i/2)\frac{\partial\varphi}{\partial x}\right]\right\}u(E,x)+Mv(E,x)=0 \quad (2.7a)$$

$$\left\{\varepsilon+ic\left[\frac{\partial}{\partial x}-(i/2)\frac{\partial\varphi}{\partial x}\right]\right\}v(E,x)+Mu(E,x)=0 \quad (2.7b)$$

where

$$\varepsilon=E+i(\mu_c/2). \quad (2.8)$$

For convenience, henceforth, the variable x and the parameter E will be suppressed in $u(E,x)$ and $v(E,x)$ when their inclusion is obvious. The same convention will also be used for other cases.

Equation (2.7) can be decoupled by eliminating either u or v in the form

$$\left[c^2\frac{\mathrm{d}^2}{\mathrm{d}x^2}+D^2\pm ic\frac{\mathrm{d}D}{\mathrm{d}x}-M^2\right]\begin{pmatrix}u\\v\end{pmatrix}=0. \quad (2.9)$$

The double signs $\pm$ are used for u and v respectively, and D is defined by

$$D(x)=(c/2)\,\mathrm{d}\varphi/\mathrm{d}x+\varepsilon. \quad (2.10)$$

The real part represents essentially the local deviation from the exact Bragg condition. Henceforth, it will be called D field. Thus, the problem is reduced to solving the second-order ordinary differential equation (2.9).

If the wave function u (for example) is obtained, (2.7a) and (2.10) will give the counterpart v in the form

$$v(E,x)=M^{-1}\left[ic\frac{\mathrm{d}}{\mathrm{d}x}-D(x)\right]u(E,x). \quad (2.11)$$

3. The special deformation

The lattice phase in (2.2) is defined by

$$\varphi=2\pi(\bar{\mathbf{g}}.\mathbf{u}) \quad (3.1)$$

where $\mathbf{u}$ is the displacement of the lattice point in the deformed crystal (*e.g.* Kato, 1974). The local RLV differs from $\bar{\mathbf{g}}$ by

$$\Delta g=-(\mathrm{d}/\mathrm{d}x)(\varphi/2\pi) \quad (3.2)$$

along the x axis.

In this paper, it is assumed that

$$\mathrm{d}\varphi/\mathrm{d}x=(2/c)D_0\tanh\alpha x. \quad (3.3)$$

By integrating (3.3), we shall see that

$$\varphi=(2D_0/\alpha c)\log(\cosh\alpha x), \quad (3.4)$$

the integral constant being fixed by $\varphi(0)=0$. The assumed deformation, therefore, implies an expansion or compression of the lattice spacing along the x axis, depending on the sign of D_0. (For definiteness, α is assumed to be positive.) In this context, when the Bragg condition is satisfied exactly at the depth $x=0$, the parameter E is also assumed to be zero.

Then, the D field defined by (2.10) has the form

$$D(x) = D_0[\xi(x) + \eta] \tag{3.5}$$

where

$$\xi = \tanh \alpha x, \qquad \eta = \varepsilon / D_0 \tag{3.6a, b}$$

(Fig. 1).

4. The exact wave field

4.1. *The transmitted wave*

We shall change the position variable x to ξ defined by (3.6a). Then, it turns out from (2.9) that the wave function $u(\xi)$ satisfies an equation having the form

$$\left[(1-\xi^2)\frac{d^2}{d\xi^2} - 2\xi\frac{d}{d\xi} + \nu(\nu+1) - (\mu^2 + 2\nu^2\eta\xi)/(1-\xi^2)\right] u(\xi) = 0 \tag{4.1}$$

where

$$\nu = i(D_0/\alpha c) = i\bar{\nu} \tag{4.2a}$$

$$\mu^2 = [M^2 - (D_0^2 + \varepsilon^2)]/(\alpha c)^2. \tag{4.2b}$$

If $\eta = 0$, (4.1) is nothing else but the standard form of the Legendre equation (AS, 8.1.1). To obtain the solution in general cases, the following transformation will be made:

$$u(\xi) = G(\xi)F(\xi) \tag{4.3a}$$

$$G(\xi) = (1-\xi^2)^{q/2}[(1+\xi)/(1-\xi)]^{p/2}. \tag{4.3b}$$

Then, the equation for $F(\xi)$ can be written in the form

$$g_2F'' + g_1F' + g_0F = 0 \tag{4.4}$$

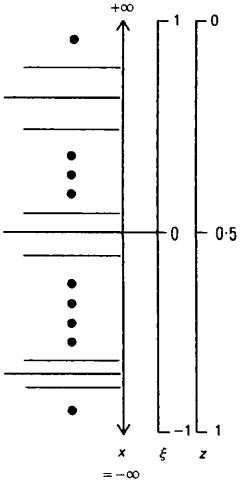

Fig. 1. The model of the lattice distortion and the relations among spatial variables; x, $\xi = \tanh \alpha x$ and $z = (1/2)(1-\xi)$. The deviation of the lattice spacing is assumed to be $D_0\xi$.

where the prime implies $d/d\xi$ and

$$g_2 = (1-\xi^2) \tag{4.5a}$$

$$g_1 = 2(p - q\xi - \xi) \tag{4.5b}$$

$$g_0 = \nu(\nu+1) - (\mu^2 - 2\bar{\nu}^2\eta\xi)/(1-\xi^2) - q(q+1) + (p^2 + q^2 - 2pq\xi)/(1-\xi^2). \tag{4.5c}$$

Therefore, one can eliminate two terms proportional to $(1-\xi^2)^{-1}$ in (4.5c) by setting

$$p^2 + q^2 = \mu^2 \tag{4.6a}$$

$$pq = \bar{\nu}^2\eta. \tag{4.6b}$$

By solving these we have

$$p + q = \pm i[(\varepsilon - D_0)^2 - M^2]^{1/2}/\alpha c \tag{4.7a}$$

$$p - q = \pm i[(\varepsilon + D_0)^2 - M^2]^{1/2}/\alpha c. \tag{4.7b}$$

We shall explain the physical interpretation of these parameters in § 4.3.

By the variable change

$$z = (1-\xi)/2, \tag{4.8}$$

(4.4) can be rewritten in an explicit form for $F(z)$,

$$z(1-z)F'' + [(1+q-p) - 2(1+q)z]F' + \lambda(\lambda + 2q + 1)F = 0 \tag{4.9}$$

where

$$\lambda = i\bar{\nu} - q. \tag{4.10}$$

Equation (4.9) is the standard form of the hypergeometric equation and the solution is well investigated (AS, chap. 15; BE, chap. II). The simplest solution is

$$F_1 = F(a, b; c; z) \tag{4.11a}$$

where

$$a = -i\bar{\nu} + q, \quad b = 1 + i\bar{\nu} + q, \quad c = 1 + q - p. \tag{4.12a, b, c}$$

In Appendix A the power series for F is presented together with some comments on its convergence.

After Kummer, 24 forms of the solution of (4.9) having different analytical properties are listed (BE, pp. 105-106). Obviously, only a set of two among them is sufficient to describe any solution of (4.9). Here, as the linearly independent solution (LIS), the following pair will be adopted:

$$F_2 = F(a, b; 1 + a + b - c; 1 - z) \tag{4.11b}$$

$$F_5 = z^{1-c}(1-z)^{c-a-b}F(1-a, 1-b; 2-c; z) \tag{4.11c}$$

which are u_2 and u_5 in the notation of BE. F_2 is analytic at $z = 1$ ($x = -\infty$) but singular at $z = 0$ ($x = +\infty$). F_1 and F_5 have the reverse property. The underlying physical reason for this choice will be explained in due course.

Substituting (4.11a) into (4.3), we have

$$u(\xi) = G(\xi)F_1 = U(p, q; i\bar{\nu}; \xi) \qquad (4.13a)$$

which will be called the prototype wave in this paper. The second equation is the definition of the U function. Similarly, we obtain the set of LIS of (4.1), which we shall write as

$$u_a(\xi) = G(\xi)F_2 = U(-p, q; i\bar{\nu}; -\xi) \qquad (4.13b)$$

$$u_b(\xi) = G(\xi)F_5 = (2)^{2q}U(-p, -q, i\bar{\nu}; \xi). \qquad (4.13c)$$

In connection with these functions, a few mathematical remarks may be made. The factor $(2)^{2q}$ in (4.13c) may be dropped because a constant is irrelevant in considering LIS. To the author's knowledge, the U function is not given a specific name; however, when λ $(=-a)$ is a positive integer it is called Jacobi's polynomials (AS, 22.5.42).* In the special case of $\varepsilon = 0$ [$\eta = 0$; $p = \mu$ and $q = 0$, or $p = 0$ and $q = \mu$ after (4.6a) and (4.6b)],

$$u_a(\xi) = \Gamma(1+\mu)P_\nu^{-\mu}(-\xi) \qquad (4.14a)$$

$$u_b(\xi) = \Gamma(1+\mu)P_\nu^{-\mu}(\xi) \qquad (4.14b)$$

where $P_\nu^{-\mu}(\xi)$ is the Legendre associate function of the first kind, and $\Gamma(1+\mu)$ is the Γ function (AS, 8.1.2).

4.2. *The Bragg-reflected wave*

In this section, the expressions for $v(\xi)$, $v_a(\xi)$ and $v_b(\xi)$ corresponding to $u(\xi)$, $u_a(\xi)$ and $u_b(\xi)$ will be derived by the use of (2.11) with the explicit expression (3.5) for the D field. Since, however, a rather lengthy calculation is required, only the outline for the prototype wave, $u(\xi)$, will be explained.

It is straightforward to obtain the differential relation

$$ic\frac{\mathrm{d}}{\mathrm{d}x}U = i\alpha c(1-\xi^2)\frac{\mathrm{d}}{\mathrm{d}\xi}U(i\bar{\nu}; \xi). \qquad (4.15)$$

The right-hand side, after performing the differentiation, consists of terms proportional to $U(i\bar{\nu}; \xi)$ and a term which is essentially proportional to $(\mathrm{d}/\mathrm{d}z)F(a, b; c; z)$. The latter is given by $F(a, b; c; z)$ and $F(a+1, b-1; c; z)$ by the use of the differential formula (AS, 15.2.3) and Gauss's relation (AS, 15.2.19) for the contiguous function. Since $a+1 = 1-i\bar{\nu}+q$ and $b-1 = i\bar{\nu}+q$, and they are exchangeable in HGF, it turns out that (4.15) consists of $U(i\bar{\nu}; \xi)$ and $U(-i\bar{\nu}; \xi)$.

Inserting the result into (2.11), we notice that all terms proportional to $U(i\bar{\nu}; \xi)$ cancel out and there only remains a single term proportional to $U(-i\bar{\nu}; \xi)$. It is worth noting the following point. The suppressed parameters p and q are implicitly a function of $\bar{\nu}$ through D_0 [*cf.* (4.7a, b)]. However, they are fixed in the course of calculation so that only the sign of the explicit parameter $\bar{\nu}$ is to be changed. Finally, the following result is obtained.

$$v(\xi) = C(p, q)U(p, q; -i\bar{\nu}; \xi) \qquad (4.16a)$$

where

$$C(p, q) = M^{-1}(\alpha c/\bar{\nu})(i\bar{\nu}+p)(i\bar{\nu}-q). \qquad (4.17)$$

From the same argument, one can obtain

$$v_a(\xi) = -C(-p, q)U(-p, q; -i\bar{\nu}; -\xi) \qquad (4.16b)$$

$$v_b(\xi) = C(-p, -q)U(-p, -q; -i\bar{\nu}; \xi). \qquad (4.16c)$$

A useful expression for $C(p, q)$ in terms of ε, D_0 and M will be given in § 5.

Here, in order to show the general idea of the U function, the amplitudes of O and G waves, which are $|u_a|$ and $|v_a|$ respectively, are illustrated in Fig. 2.

4.3. *The asymptotic form of the solution*

At the beginning, we shall summarize some results so far obtained. After combining all factors introduced in several transformations [*cf.* (2.1), (2.3) and (2.6), (4.13) and (4.16)], we have the expressions

$$O \text{ wave: } d[G(\xi)F(i\bar{\nu}; \xi)]\exp(i\varphi/2) \times \exp(-iEt)\exp(i\bar{\mathbf{K}}_0.\mathbf{r}) \qquad (4.18a)$$

$$G \text{ wave: } d[C(p, q)G(\xi)F(-i\bar{\nu}; \xi)]\exp(-i\varphi/2) \times \exp(-iEt)\exp(i\bar{\mathbf{K}}_g.\mathbf{r}) \qquad (4.18b)$$

where d is the amplitude coefficient to be determined by the boundary condition (§ 5), and $F(i\bar{\nu}; \xi)$ is an abbreviation for the HGF having the form $F[-i\bar{\nu}+q, 1+i\bar{\nu}+q; 1-p+q; (1-\xi)/2]$ for the prototype wave. For the LIS of type (a) and (b), a proper sign must be assigned to ξ, p and q in $G(\xi)$ and $F(\pm i\bar{\nu}; \xi)$ as well as in $C(p, q)$.

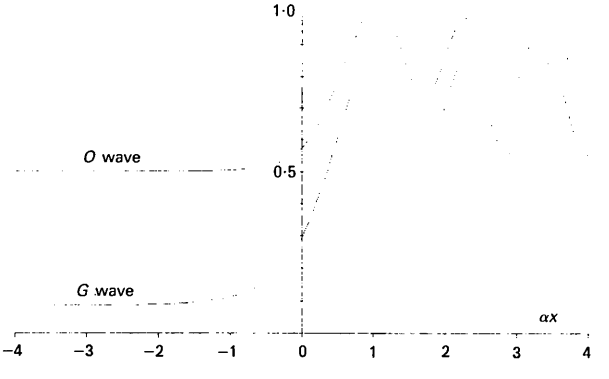

Fig. 2. A typical example of the amplitudes of O wave (u_a) and G wave (v_a) *versus* the variable x for a non-absorbing crystal; $E = 0$ and $M/\alpha c = 1$. In this particular case, $|u_a| = |F(i\bar{\nu}; -\xi)|$ and $|v_a| = |C_a F(-i\bar{\nu}; -\xi)|$ so that the figure illustrates also the behaviour of HGF's.

* This situation never happens in our problem. The standard form of Jacobi's polynomial is written as $P_n^{(\alpha,\beta)}(\xi)$. One can show that $\alpha = q-p$ and $\beta = q+p$ in the present notation.

The purpose of this section is to consider (4.18) in the region of large $|x|$, where the lattice deformation is gentle so that one can expect that they tend to the corresponding expression for the perfect crystal. Through this treatment, (4.18) will be recast in physically understandable forms [see (4.23)].

First, the factor $\exp(\pm i\varphi/2)$ is taken up. From (3.4), it is seen that

$$\varphi/2+(D_0/\alpha c)\log 2\to(D_0/c)x \quad \text{as } x\to+\infty \tag{4.19a}$$

$$\to(-D_0/c)x \quad \text{as } x\to-\infty. \tag{4.19b}$$

The constant term $(D_0/\alpha c)\log 2$ is irrelevant to our problem, so it is henceforth dropped. Also, the following arguments will be described in the case of $x\to+\infty$, unless otherwise stated.

For our purpose it is convenient to define the following vectors having (t, x) components:

$$2\pi\Delta\mathbf{g}_o\colon (D_0, D_0/c) \tag{4.20a}$$

$$2\pi\Delta\mathbf{g}_g\colon (D_0, -D_0/c). \tag{4.20b}$$

They are illustrated by the vector $(\bar{O}O)$ and the vector $(\bar{G}G)$ respectively in Fig. 3. Here, $\bar{O}$ and $\bar{G}$ are the standard RL points at the depth of $x=0$, and O and G are the local RL points in the region of $x\to+\infty$ which must lie on the Ewald sphere with its centre at L. Since we are concerned with the vicinity of $\bar{O}$ and $\bar{G}$, the sphere is approximated by the contact plane at $\bar{O}$ and $\bar{G}$. Remembering the relations (3.2) and (3.3) [$\tanh\alpha x\to 1$], one can see that $\Delta\mathbf{g}=\Delta\mathbf{g}_g-\Delta\mathbf{g}_o$ is satisfied automatically.

Next, we shall consider the factor $G(\xi)$ [*cf.* (4.3*b*)]. Returning from the variable ξ to x, we obtain the asymptotic form

$$G(p, q; \xi)=(\cosh\alpha x)^{-q}\exp(p\alpha x) \tag{4.21a}$$

$$\to(2)^q\exp(p-q)\alpha x \quad (x\to\infty) \tag{4.21b}$$

$$\to(2)^q\exp(p+q)\alpha x \quad (x\to-\infty). \tag{4.21c}$$

The explicit expressions for $\alpha(p\pm q)$ are given by (4.7). Bearing these in mind we define the deviation vector Δk having the following (t, x) components:

$$\Delta\mathbf{k}\colon \{E+D_0, \pm c^{-1}[(\varepsilon+D_0)^2-M^2]^{1/2}\}. \tag{4.22}$$

The end point D of the vector is drawn in Fig. 4 as a function of E in the case of non-absorbing crystals ($\varepsilon=E$). Using $\Delta\mathbf{k}$, $\Delta\mathbf{g}_o$ and $\mathbf{g}_g$ [(4.20) and (4.22)], we can write the total wave in the form

$$O \text{ wave: } dF(i\bar{\nu};\xi)\times\exp i[(\bar{\mathbf{K}}_o-\Delta\mathbf{k}+2\pi\Delta\mathbf{g}_o).\mathbf{r}] \tag{4.23a}$$

$$G \text{ wave: } dC(p, q)F(-i\bar{\nu};\xi)\times\exp i[(\bar{\mathbf{K}}_g-\Delta\mathbf{k}+2\pi\Delta\mathbf{g}_g).\mathbf{r}]. \tag{4.23b}$$

It is easily seen that the vector expression in (4.23) has the meaning of the wave vector in the perfect crystal where the RLV is $\bar{\mathbf{g}}+\Delta\mathbf{g}(+\infty)$ and $F(i\bar{\nu};\xi)$ and $C(p, q)F(-i\bar{\nu};\xi)$ can be interpreted as the amplitude. In fact, $F(\pm i\bar{\nu};\xi)$ tends to unity when $x\to+\infty$.

The above interpretation, however, cannot be applied straightforwardly to the region of $x\to-\infty$ although one can write down the asymptotic form of the crystal wave in the same manner as (4.23) simply by changing the sign of D_0 in $\Delta\mathbf{k}$, $\Delta\mathbf{g}_o$ and $\Delta\mathbf{g}_g$. In this case, however, $F(\pm i\bar{\nu};\xi)$ oscillates as ξ changes. In particular, they are singular at $\xi=-1$ in non-absorbing crystals. Therefore, we need a careful consideration of the phase and amplitude (see § 6).

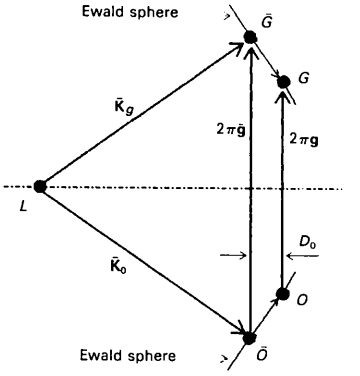

Fig. 3. The relation of the wave vectors, $\bar{\mathbf{K}}_o$ and $\bar{\mathbf{K}}_g$, and the standard RLV $\bar{\mathbf{g}}$ ($x=0$) and the local RLV $\mathbf{g}$ ($x=+\infty$). The broken line indicates the Brillouin zone boundary. *L*: the Laue point. $\bar{O}$ and $\bar{G}$: the standard RL points. *O* and *G*: the local RL points ($x=+\infty$).

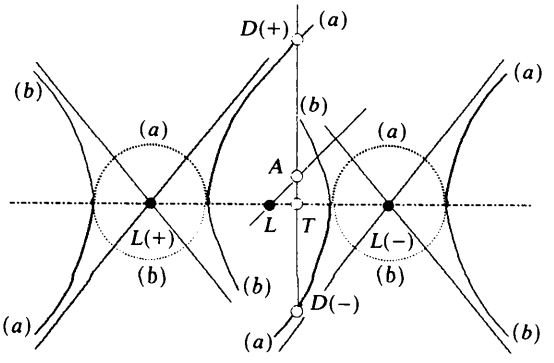

Fig. 4. The dispersion surface definable in the region of $x\to+\infty$ (left) and $x\to-\infty$ (right). (+) and (−) refer to these regions respectively. (*a*) and (*b*) denote the wings to which the waves of type (*a*) and (*b*) belong respectively. The figure illustrates a non-absorbing case. The full hyperbolic lines show the real part of $\Delta\mathbf{k}=\overrightarrow{LD}$ [equation (4.22)]. The dotted circles show the imaginary part. $\Delta\mathbf{K}_o=\overrightarrow{LA}$ [equation (5.2*a*)]. *L*: the Laue point. *A*: the dispersion point of the incident wave in vacuum. *D*: the dispersion point of the crystal wave. $\overline{LT}=E$ and $\overline{L(+)L}=\overline{LL(-)}=D_0$.

So far, we have discussed the prototype wave. The same result is obtained with the LIS of type (a) because $G(-p, q; -\xi) = G(p, q; \xi)$. However, the interpretation of the amplitude and the phase is allowed only in the region of $x = -\infty$ where $F(\pm i\bar{\nu}; -\xi)$ tends to unity.

In the case of the LIS of type (b),

$$G(-p, -q; \xi) = (\cosh \alpha x)^q \exp(-p\alpha x) \tag{4.24a}$$

$$\to (2)^{-q} \exp(-p+q)\alpha x \quad (x \to +\infty) \tag{4.24b}*$$

$$\to (2)^{-q} \exp(-p-q)\alpha x \quad (x \to -\infty). \tag{4.24c}*$$

Again the same argument can be applied also to this wave in the region of $x = +\infty$.

At this stage, it is worth commenting about the double sign of the expressions (4.7) for $(p \pm q)$. As will be seen in the next section, we are interested in the LIS of type (a) which is finite in the region $(x_e, -\infty)$, where x_e is the x coordinate of the entrance surface. From this requirement we have to assign $(-)$ to $(p \pm q)$ with the usual convention of the Riemann surface for the expression $[(\varepsilon + D_0)^2 - M^2]^{1/2}$ as a function of $(E + D_0)$. Similarly, since we are interested in the LIS of type (b) which is finite in the region $(+\infty, x_a)$ where x_a is the x coordinate of the exit surface, we must assign $(-)$ also to $(p \pm q)$. Notice that the sign of $(p \pm q)$ in (4.24b, c) is opposite to that in (4.21b, c).

Physically, the dispersion surface is definable in the region of $|x| \to \infty$. The wings (a) and (b) illustrated in Fig. 4 must be assigned to u_a and u_b, respectively. Thus, the deviation vector $\Delta\mathbf{k}$ can be regarded as a single-valued vector for a given E. The double sign in (4.7a, b) and (4.22) is redundant when u_a and u_b are treated separately.

5. The solution satisfying the boundary conditions

It is assumed that the incident plane wave impinges on a boundary at $x = x_e$ $(\xi = \xi_e;\ \mathbf{r} = \mathbf{r}_e)$ from the $+x$ side. We shall discuss the following two cases: (A) Darwin's case or a half-infinite crystal; and (B) Ewald's case or a parallel-sided crystal with an extra exit surface at $x = x_a$ $(\xi = \xi_a;\ \mathbf{r} = \mathbf{r}_a)$. Since our two LIS's with the negative sign of $(p \pm q)$ satisfy the physical requirements of finiteness, we can construct the concrete solution by the standard procedure in the theory of second-order differential equations.

The incident wave is assumed to have the form

$$D_e(\mathbf{r}) = D_e \exp[i(\bar{\mathbf{K}}_o - \Delta\mathbf{K}_o).\mathbf{r}] \tag{5.1}$$

where the deviation vector $\Delta\mathbf{K}_o$ is defined through its (t, x) components (see Fig. 4),

$$\Delta\mathbf{K}_o : (E, E/c). \tag{5.2a}$$

Since this vector is perpendicular to $\bar{\mathbf{K}}_o$, it is assured that the magnitude of $(\bar{\mathbf{K}}_o - \Delta\mathbf{K}_o)$ is $K = |\bar{\mathbf{K}}_o|$ with sufficient accuracy. Similarly, we define also the deviation vector

$$\Delta\mathbf{K}_g : (E, -E/c). \tag{5.2b}$$

Then, the diffracted wave in vacuum can be written as

$$D_g(\mathbf{r}) = D_g \exp[i(\bar{\mathbf{K}}_g - \Delta\mathbf{K}_g).\mathbf{r}]. \tag{5.3}$$

In the Ewald case, one needs the transmitted wave in vacuum. It must have the form

$$D_T(\mathbf{r}) = D_T \exp[i(\bar{\mathbf{K}}_o - \Delta\mathbf{K}_o).\mathbf{r}]. \tag{5.4}$$

The amplitudes D_g and D_T may depend on x_e and x_a but they are independent of the variable t. This is easily anticipated because the RLV has only an x component in our model.

5.1. *The Darwin case*

It is enough to take only the LIS of type (a) as the crystal wave. The boundary conditions $[D_e(\mathbf{r}_e) = d_o(\mathbf{r}_e);\ D_g(\mathbf{r}_e) = d_g(\mathbf{r}_e)]$ can be written explicitly as

$$D_e \exp[-i(E/c)x_e] = d_a u_a(\xi_e) \exp[i\varphi(x_e)/2] \tag{5.5a}$$

$$D_g \exp[i(E/c)x_e] = d_a v_a(\xi_e) \exp[-i\varphi(x_e)/2] \tag{5.5b}$$

where d_a stands for d in the expression (4.18) and u_a and v_a are given by (4.13b) and (4.16b) respectively. Solving d_a and D_g from these, we have the expressions for the O and G waves in the crystal with the use of d_a for d in (4.18a, b). Also the reflected wave in vacuum is obtained by inserting D_g into (5.3).

It is straightforward to calculate the reflectivity in the form

$$R_g = |D_g/D_e|^2 = |v_a(\xi_e)/u_a(\xi_e)|^2 \tag{5.6a}$$

$$= |C_a(-)|^2 \times \left| \frac{F(i\bar{\nu}+q, 1-i\bar{\nu}+q; 1+p+q; (1+\xi_e)/2)}{F(-i\bar{\nu}+q, 1+i\bar{\nu}+q; 1+p+q; (1+\xi_e)/2)} \right|^2. \tag{5.6b}$$

The amplitude ratio $C_a(-) = -C(-p, q)$ can be written as follows from the definition (4.17) of $C(p, q)$ and the expression of pq [(4.6b)] and $p+q$ [(4.7a)]:

$$C_a(-) = M^{-1}\{[(\varepsilon - D_0)^2 - M^2]^{1/2} - (\varepsilon - D_0)\}. \tag{5.7}$$

In the case of $x_e \to -\infty$ where the HGF's tend to unity, obviously, the expression (5.6b) is identical to the reflectivity of the perfect crystal. It shows the famous

* If we retain the factor $(2)^{2q}$ in (4.13c), the factor $(2)^{-q}$ can be replaced by $(2)^q$ as the LIS of type (a) [*cf.* (4.21b) and (4.21c)].

silk-hat rocking curve in non-absorbing crystals, and a total reflection is expected in the domain of $|E-D_0|<M$.

The second factor of (5.6*b*) including HGF's is complicated in general. However, in non-absorbing crystals and in the domain of $|E-D_0|<M$ ($p+q$ is real), one can show that the factor is unity independently of ξ_e (Appendix *B*). This implies that total reflection always occurs. The result seems strange at first sight because the crystal is distorted. However, it is reasonable because the crystal is a perfect reflector at $x=-\infty$ and, after all, the reflected wave comes out from the entrance surface. This phenomenon may occur in neutron cases but never does in X-ray cases because of absorption.

In another extreme case, $x_e=+\infty$, the situation is again a little complicated. In fact, if the crystal is perfectly non-absorbing, the HGF's in (5.6*b*) are singular. If, however, the crystal is absorbing (even to a very small degree) R_g can be written asymptotically in the form (Appendix *C*)

$$R_g \to |C_a(+)|^2 |\text{HGF factor}|^2 \text{ as } \xi_e \to 1, \qquad (5.8a)$$

where the second factor is same as the expression in (5.6*b*) with the opposite sign in p and ξ_e, and $C_a(+)=C(p,q)$ is given by

$$C_a(+)=M^{-1}\{[(\varepsilon+D_0)^2-M^2]^{1/2}-(\varepsilon+D_0)\}. \qquad (5.8b)$$

Now, the HGF's tend to unity. Therefore, one can expect that in the absorbing crystal the rocking curve tends to the form of the perfect crystal in the region of $x_e=+\infty$. The result is reasonable because the effective crystal limited by absorption is perfect.

5.2. *The Ewald case*

We have to determine the two amplitude coefficients d_a and d_b. The procedure for calculation is standard so that here only the final results of reflectivity and transmissivity are presented.

$$R_g=|v_a(\xi_e)v_b(\xi_a)-v_a(\xi_a)v_b(\xi_e)|^2/|\Delta|^2 \qquad (5.9a)$$

$$T_g=|u_a(\xi_a)v_b(\xi_a)-u_b(\xi_a)v_a(\xi_a)|^2/|\Delta|^2 \qquad (5.9b)$$

where

$$\Delta=u_a(\xi_e)v_b(\xi_a)-u_b(\xi_e)v_a(\xi_a). \qquad (5.10)$$

When ξ_a tends to -1, $|v_b(\xi_a)| \gg |v_a(\xi_a)|$ in absorbing crystals. Therefore, the expression (5.9*a*) tends to that given by (5.6*b*).

6. Creation of a Bloch wave of another type

In this section, we shall supplement the argument in § 4.3 and discuss the creation of a Bloch wave owing to the lattice distortion. As an example, we shall take up the LIS of type (*a*), $u_a(\xi)=G(\xi)F_2$ and the associated v_a.

The key to the present argument is based on the mathematical relation [*cf.* equations (1), (5), (17) and (35) of § 2.9 of BE; pp. 105–106]

$$F_2=F(a,b;1+a+b-c;1-z) \qquad (6.1a)$$

$$=W_1F(a,b;c;z)+W_2(z)^{1-c}F(1+a-c,1+b-c;2-c;z) \qquad (6.1b)$$

where

$$W_1=\frac{\Gamma(1+a+b-c)\Gamma(1-c)}{\Gamma(1+a-c)\Gamma(1+b-c)} \qquad (6.2a)$$

$$W_2=\frac{\Gamma(1+a+b-c)\Gamma(c-1)}{\Gamma(a)\Gamma(b)} \qquad (6.2b)$$

and the parameters a, b, c are defined by (4.12) in our problem. Multiplying the asymptotic form of $G(\xi)$ [(4.21*b*, *c*)] and using $1-c=p-q$, we easily see that

$$u_a \to (2)^q[W_1\exp(p-q)\alpha x+W_2\exp(q-p)\alpha x] \quad \text{as } x\to+\infty \qquad (6.3a)$$

$$\to (2)^q\exp(p+q)\alpha x \quad \text{as } x\to-\infty \qquad (6.3b)$$

where HGF's are omitted because their asymptotic values tend to unity in the respective region. A similar result is also obtained for the diffracted wave v_a as follows:

$$v_a \to (2)^q C_a(-)[\bar{W}_1\exp(p-q)\alpha x+W_2\exp(q-p)\alpha x] \quad \text{as } x\to\infty \qquad (6.4a)$$

$$\to (2)^q C_a(-)\exp(p+q)\alpha x \quad \text{as } x\to-\infty \qquad (6.4b)$$

where $\bar{W}_1$ and $\bar{W}_2$ are the same expression as W_1 and W_2, respectively, with the use of the opposite sign for the parameter $\bar{\nu}$.

The physical interpretation is given as follows. The first terms of (6.3*a*) and (6.4*a*) constitute the incident Bloch wave belonging to the wing (*a*) of the dispersion surface in the region of $x\to+\infty$. It propagates through the distorted region and emerges in the form of (6.3*b*) and (6.4*b*). This is the transmitted Bloch wave and, in fact, one can assign it to the wing (*a*) of the dispersion surface defined in the region of $x\to-\infty$. On the other hand, the incident wave is always associated with a Bloch wave in the form of the second terms of (6.3*a*) and (6.4*a*), which belong to the wing (*b*) of the dispersion surface in $x\to+\infty$. This wave can be interpreted as the reflected Bloch wave created by the lattice distortion.

In the case of half-infinite crystals, in which we are most interested here, the above interpretation needs an extension for the incident and reflected Bloch waves. Strictly speaking, the concept of the Bloch wave is allowed only in the perfect crystal. Here, we

consider the modified Bloch wave (MBW) as introduced in the eikonal theory (Kato, 1963, 1974).

For convenience, the first and second HGF's appearing in the expression (6.1*b*) are denoted by F^1 and F^2. Notice that the parameters are different. If they are gently changed near the entrance surface, one can define the incident and reflected MBW similarly to the Bloch waves as has been done in the region of $x \to +\infty$. This procedure is unique provided that z_e is small, say less than 0·5 depending on the parameters involved in HGF's. If z_e is close to unity, the expression (6.1*a*) should be used as the wave function. Then, no wave is created and we are not interested in such cases.

On the entrance surface, the intensity (energy) carried by the incident MBW can be written in the form, neglecting the interference term,

$$I_B = |G|^2[|W_1 F^1(i\bar{\nu}; z_e)|^2 + |C_a \bar{W}_1 F^1(-i\bar{\nu}; z_e)|^2]. \quad (6.5a)$$

Also, the intensity of the reflected MBW has the form

$$R_B = |G z_e^{(p-q)}|^2[|W_2 F^2(i\bar{\nu}; z_e)|^2 + |C_a \bar{W}_2 F^2(-i\bar{\nu}; z_e)|^2]. \quad (6.5b)$$

Similarly, the transmitted Bloch wave can be written as

$$T_B = |G|^2(1+|C_a|^2) \quad (6.5c)$$

by the use of the original expressions for u_a and v_a, where the HGF can be omitted.

7. Summary and discussion

Starting from the fundamental equation of Takagi–Taupin type, we arrived at equation (2.9) which is suitable for studying the diffraction in a crystal with lamellar distortion. We investigated the symmetrical Bragg case in detail for a specific distortion described by (3.2) and (3.3).

First, we obtained the two linearly independent solutions u_a and u_b for the O wave [(4.13*b*, *c*)] and the associated G waves v_a and v_b [(4.16*b*, *c*)] in an infinite crystal. All of them can be represented by the U function defined by (4.13*a*) in a very symmetrical manner with respect to the parameters involved (p, q, $i\bar{\nu}$) and the position variable $\xi = \tanh \alpha x$.

The U function is a product of the algebraic function $G(\xi)$ and one HGF. HGF's are well investigated in applied mathematics so that, in principle, it is not very difficult to handle them analytically and even numerically. Nevertheless, as the parameters p, q mentioned above are in general complex, their physical meanings are not very straightforward.

For this reason, next, an attempt was made to obtain the asymptotic form of the LIS's in the regions of large $|x|$ (§§ 4.3 and 6). Then, it can be shown that $(p \pm q)$ are related to the deviation wave vector $\Delta\mathbf{k}$ in $\mp\infty$ regions, respectively. Moreover, through this analysis, the crystal wave can be interpreted by the modified Bloch waves.

The MBW creates another MBW in the distorted region. It is the reflected MBW of the original one. Thus, one can expect a kind of *Pendellösung* oscillation in the region $x > 0$ owing to the presence of two MBW's. A typical example is shown in Fig. 2. It is worth noting that, although HGF's may be singular at either $\xi = 1$ or -1, the wave functions are regular as a function of the physical coordinate x. The singularity is merely mathematical, due to the contraction of the infinite variable region $x(+\infty, -\infty)$ into the finite region $\xi(1, -1)$.

Incidentally, the creation of a new Bloch wave does not occur in perfect crystals because the Bloch wave is an eigenfunction in the perfect crystal. It is well known that such a creation occurs only on the exit surface, or at a plate-like defect such as stacking faults or twin boundaries. The present theory illustrates explicitly that such phenomena occur in any continuously distorted region.

As an application, the formulae for rocking curves are presented corresponding to the Darwin and the Ewald case. Once the two LIS's have been obtained, one can calculate the reflectivity and transmissivity by an algebraic manipulation of the values of the LIS's on the entrance and exit surfaces. This is a great merit of the analytical approach. In the Darwin case, some analytical properties were discussed in § 5.

Finally, we shall discuss briefly the extension of the present theory. It is not difficult to generalize the treatment to the asymmetric case and even to the Laue case, provided that the lattice phase φ [(3.1)] has the same form as (3.4) as a function of X normal to the crystal surface. Then, the problem remains of solving a one-dimensional differential equation, because the relevant differentiations $\partial/\partial t$ and $\partial/\partial x$ can be replaced by $\mathrm{d}/\mathrm{d}X$ multiplied by definite geometrical constants.

Because of the relaxation of strain on the crystal surface, the above model is physically plausible at least in a limited area of the surface. If this area is sufficiently wide from the viewpoint of diffraction, one can apply the present theory. In this context, it is to be noted also that the theory can be used in practice for any monotonic one-dimensional distortion. Then, we are able to select two positions x_e and x_a and the parameters D_0 and α in such a way that the functional form ($D_0 \tanh \alpha x$) describes closely the distortion concerned.

Remembering these flexibilities, as worked out by Authier *et al.* (1989), one interesting topic is the application to the standing wave method, which affords valuable information about the atomic structure on the crystal surface and an interface between two perfect crystals. When the surface and the interface are associated with an intrinsic lattice distortion,

the present theory must be useful for calculating the standing wave field.

APPENDIX *A*

The power series for a HGF

The standard HGF is defined by the power series

$$F(\alpha, \beta; \gamma; \zeta) = \sum_{n=0}^{\infty} \frac{(\alpha)_n (\beta)_n}{(\gamma)_n n!} \zeta^n \qquad (A.1)$$

where $(\alpha)_n$ are Pochhammer's symbols; $(\alpha)_n = \Gamma(\alpha+n)/\Gamma(\alpha)$, and ζ stands for $1-z$ and z for u_a and u_b, respectively. The series is regular at $\zeta = 0$ and absolutely converges for $\zeta < 1$, if γ is not $0, -1, -2, \ldots$ (condition 1). If $\mathrm{Re}\,(\alpha+\beta-\gamma) < 0$ (condition 2), the series converges absolutely also at $\zeta = 1$.

In our problem, when the crystal is absorbing, $R_e(p \pm q)$ are positive except that they take zero for the case of a special value of E. This ensures in practice the above two conditions for LIS's. In non-absorbing crystals, the convergence of u_a and u_b is conditional at $z = 0$ and $z = 1$, respectively. Physically, we can eliminate such cases by limiting the crystal by the entrance and exit surfaces. Condition 1 is always satisfied.

APPENDIX *B*

Proof of $|F^*(i\bar{\nu}; q)| = |F(-i\bar{\nu}; q)|$ when $(p+q)$ is real in non-absorbing crystals

Here, the following abbreviation is used:

$$F(i\bar{\nu}; q) = F(-i\bar{\nu}+q, 1+i\bar{\nu}+q; 1+p+q; 1-z). \qquad (B.1)$$

The value of $(p-q)$ is either purely real or imaginary. For convenience we shall write

$$q = (1/2)[(p+q)-(p-q)]. \qquad (B.2)$$

If $(p-q)$ is purely real, q itself is real. In this case, the proof is obvious.

If $(p-q)$ is purely imaginary, $q^* = p$ from (*B*.2). Therefore, the HGF (*B*.1) can be rewritten with the use of equation (6) on p. 105 of BE in the form

$$F(i\bar{\nu}; q) = z^{(p-q)} F(i\bar{\nu}; p) = z^{(p-q)} F^*(-i\bar{\nu}; q). \qquad (B.3)$$

Since $(p-q)\log z$ is purely imaginary, namely $|z^{(p-q)}| = 1$, the proof is completed.

APPENDIX *C*

The mathematical supplement to u_a and u_b

We shall start with the expression of u_a [(4.13*b*)]. The same argument can also be applied to u_b. As often mentioned, $\mathrm{Re}\,(p \pm q) > 0$ for absorbing crystals. We shall discuss only this case in this Appendix.

(*a*) *The asymptotic form of u_a near $z = 1$*

The definition of F_2 [(6.1*a*)] will give the expression

$$u_a = (2)^q (z)^{-(p-q)/2} (1-z)^{(p+q)/2} \times F(a, b; 1+a+b-c; 1-z). \qquad (C.1)$$

Here, F is analytic. The singularity may arise only from the third factor, which is

$$S = \exp[(1/2)(p+q)\log(1-z)]. \qquad (C.2)$$

Since $\log(1-z)$ is real and negative, S is an exponentially damping function towards $x = -\infty$ so that u_a is in fact regular.

(*b*) *The asymptotic form of u_a near $z = 0$*

If the expression (6.1*b*) is used for F_2 we shall have another expression,

$$u_a = (2)^q (1-z)^{(p+q)/2} [W_1 z^{-(p-q)/2} F(a, b; c; z) + W_2 z^{(p-q)/2} F(1+a-c, 1+b-c; 2-c; z)] \qquad (C.3)$$

where $1-c = p-q$ is used. Now, the two HGF's are analytic. The singularity may arise through the factor $z^{\pm(p-q)/2}$. Similarly to the argument on S, one can conclude that the first term in square brackets increases exponentially as x increases and the second term attenuates.

Therefore, the leading term of u_a has the form

$$u_a = (2)^q \exp[-(1/2)(p-q)\log z] \times \frac{\Gamma(1+p+q)\Gamma(p-q)}{\Gamma(-i\bar{\nu}+p)\Gamma(1+i\bar{\nu}+p)} \times F(a, b; c; z). \qquad (C.4)$$

(*c*) *The reflectivity in the Darwin case*

The expression (5.6*b*) cannot be used in practice when ξ_e is close to 1, because the HGF's involved have a singular behaviour. In this case we use the original form (5.6*a*), and the leading term (*C*.4) for u_a and a similar one for v_a [$\bar{\nu} \rightarrow -\bar{\nu}$]. Then we shall have an analytic expression (5.8*a*) even at $\xi_e = 1$. In fact, the singular factors of u_a and v_a and the Γ functions in the nominator of (*C*.4) are cancelled and we obtain

$$v_a/u_a = -C(-p, q)(p+i\bar{\nu})/(p-i\bar{\nu})(\text{HGF factor}) = C(p, q)(\text{HGF factor}). \qquad (C.5)$$

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Experimental Distinction of Elements with Similar Atomic Number using Anomalous Dispersion (δ Synthesis): an Application of Synchrotron Radiation in Crystal Structure Analysis

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Abstract

A two-wavelength method (δ synthesis) which deals with the experimental distinction of elements with similar atomic number by means of single-crystal X-ray diffraction measurements is presented. This method uses the characteristic wavelength dependence of the anomalous-dispersion correction terms f' and f'' close to the absorption edges of the corresponding elements. In case of a properly chosen wavelength combination the resulting difference electron density map (δ map) mainly shows peaks at the positions of the 'near-edge' element. The basic mathematical formalism is described and selection rules for the determination of the optimal wavelength combination are derived. The applicability in case of mixed occupancy and/or occupancy deficiency is discussed. A series of δ maps which are based on theoretical data sets and calculated according to the formalism of the δ synthesis are shown. From this the influence of errors on the interpretation of the δ map can be estimated and the requirements on the experimental conditions can be determined. First experimental results concerning the distinction of Pb/Bi in the well known crystal structure of galenobismutite ($PbBi_2S_4$) are presented. They show that the δ synthesis is not only a theoretical model, but can be applied successfully in practice. For experimental reasons the applicability of the δ synthesis is restricted to elements with atomic numbers greater than $Z = 22$.

Introduction

The experimental distinction of elements with similar atomic number Z (*e.g.* Pb/Bi) in crystal structures by use of conventional X-ray diffraction techniques is usually not possible due to the small difference of the corresponding atomic scattering power f_j. The only way to solve this problem is to enlarge these differences. This can be done using anomalous dispersion effects.

The atomic scattering factor f_j is usually written as

$$f_j(\mathbf{h}, \lambda) = f_j^0(\mathbf{h}) + f_j'(\lambda) + if_j''(\lambda),$$
$$f_j^0 = Z \text{ in case of } (\sin\theta)/\lambda = 0.$$

The terms f_j' and f_j'' due to anomalous scattering vary drastically only close to the absorption edges. Apart from that they are nearly wavelength independent. This physical property can be used to solve the crystallographic problem outlined above by choosing a suitable wavelength to contrast the electron density map.

However this method remained generally theoretical in the past, as long as experimental requirements such as (1) free choice of the wavelength and additionally high photon flux in the X-ray region and (2) small bandwidth (*i.e.* small $\Delta\lambda/\lambda$) were not available. Recently the rapid development of improved synchrotron radiation sources has made it an experimental method.

In 1984, a paper was presented (Ohsumi, Tsutsui, Takeuchi & Tokonami, 1984) in which an attempt was made to determine the Pb/Bi distribution in the crystal structure of lillianite ($3PbS.Bi_2S_3$). The experiment was carried out at the Photon Factory in Japan at a wavelength close to the L_{III} absorption edge of lead. The data were analysed using the usual difference Fourier synthesis. But the atomic positions were not distinguished definitely due to insufficient

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