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Ewald's Extended Dynamical Theory of Diffraction

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Abstract

The dynamical theory of diffraction usually employs some approximations that, however, cease to be valid in some cases, *e.g.* if the Bragg angle is near $\pi/2$, at grazing incidence or at skew reflection. Some difficulties of the conventional dynamical theory of diffraction are analysed using the concept of Ewald's dynamical theory of diffraction.

1. Introduction

In the so-called 'conventional' dynamical theory of diffraction some approximations are used which mean that the final formulae are not valid in some extreme cases. This unfavourable situation occurs, for example, when the Bragg angle is near $\pi/2$, at grazing incidence, or at skew reflection. The usual 'extended' dynamical theory of diffraction tries to remove these difficulties by making the von Laue conventional theory more precise (Afanas'ev & Melkonyan, 1983, 1989; Baryshevsky, 1976; Brümmer, Höche & Nieber, 1979; Härtwig, 1976, 1977; Kishino & Kohra, 1971; Rustichelli, 1975). The aim of our paper is to discuss the above problems in the frame of Ewald's conception of the dynamical theory of diffraction (Ewald, 1916, 1917).

2. The exact dynamical theory of diffraction in Ewald's picture

Let us recall briefly the main results of our former paper (Litzman, 1986)* on the dynamical theory of diffraction of particles on a periodic system of point scatterers (Fermi δ potentials). We shall deal with the diffraction on a simple lattice forming a semi-infinite crystal

$$\mathbf{R}_m = m_1\mathbf{a}_1 + m_2\mathbf{a}_2 + m_3\mathbf{a}_3, \qquad m=(m_1, m_2, m_3), \quad (1)$$

$m_1, m_2 = 0, \pm1, \pm2, \ldots, \pm\infty$, $m_3 = 0, 1, 2, \ldots, \infty$, and $a_{3z} > 0$. The origin of the orthogonal coordinate system lies at the lattice point (0, 0, 0), the plane Oxy coincides with the crystal-surface plane $(\mathbf{a}_1, \mathbf{a}_2)$. The axis Oz (the unit vector $\mathbf{e}_3$) and the vector $\mathbf{a}_1\times\mathbf{a}_2$ point into the crystal. The lattice $(\mathbf{g}_1, \mathbf{g}_2, \mathbf{g}_3)$ is reciprocal to the three-dimensional lattice $(\mathbf{a}_1, \mathbf{a}_2, \mathbf{a}_3)$, *i.e.* $\mathbf{g}_i\mathbf{a}_j = 2\pi\delta_{ij}$, $i, j = 1, 2, 3$, $\mathbf{g}_3 \parallel \mathbf{e}_3$, $|\mathbf{g}_3|a_{3z} = 2\pi$, whereas the lattice $(\mathbf{b}_1, \mathbf{b}_2)$ is reciprocal to the two-dimensional lattice $(\mathbf{a}_1, \mathbf{a}_2)$, *i.e.* $\mathbf{b}_i\mathbf{a}_j = 2\pi\delta_{ij}$, $\mathbf{b}_i \perp \mathbf{e}_3$, $i, j = 1, 2$. Further, $\mathbf{c}^{\parallel}$ and $\mathbf{c}^{\perp}$ denote the components of the vector $\mathbf{c} = \mathbf{c}^{\parallel} + \mathbf{c}^{\perp}$ parallel and perpendicular to the crystal surface, respectively. Then $\mathbf{b}_1 = \mathbf{g}_1^{\parallel}$, $\mathbf{b}_2 = \mathbf{g}_2^{\parallel}$, $\mathbf{g}_3^{\parallel} = 0$.

Let $\mathbf{k}$ be the wavevector of the incident wave, $k_z > 0$. We assign to this vector $\mathbf{k}$ and to each (p, q), where p, q are integers, three other vectors $\mathbf{k}_{pq}^{\parallel}$ and $\mathbf{K}_{pq}^{\pm}(\mathbf{k})$ as follows:

$$\mathbf{k}_{pq}^{\parallel} = \mathbf{k}^{\parallel} + p\mathbf{b}_1 + q\mathbf{b}_2, \quad (2a)$$

$$\mathbf{K}_{pq}^{\pm}(\mathbf{k}) = \mathbf{k}_{pq}^{\parallel} \pm \mathbf{e}_3 K_{pqz}(\mathbf{k}), \quad (2b)$$

where

$$K_{pqz}(\mathbf{k}) = +[k^2 - (\mathbf{k}_{pq}^{\parallel})^2]^{1/2}. \quad (2c)$$

This means that

$$|\mathbf{K}_{pq}^{\pm}(\mathbf{k})| = k. \quad (2d)$$

For $(p, q) = (0, 0)$, $\mathbf{K}_{00}^{+}(\mathbf{k}) = \mathbf{k}$ and also $K_{00z}(\mathbf{k}) = k_z$ hold. Further, we define $\theta_{pq}^{\pm}$ as

$$\theta_{pq}^{\pm} \equiv \theta_{pq}^{\pm}(\mathbf{k}) = \mathbf{a}_3\mathbf{K}_{pq}^{\pm}(\mathbf{k}) = \mathbf{a}_3^{\parallel}\mathbf{k}_{pq}^{\parallel} \pm a_{3z}K_{pqz}(\mathbf{k}). \quad (2e)$$

Now let us write down equations for the diffraction of scalar waves on Fermi δ potentials. Vector waves (electromagnetic waves) can be handled in a similar way. The wave function $\Psi(\mathbf{r})$ describing the diffraction of particles on a simple perfect lattice formed by Fermi δ potentials is [(I.9)]

$$\Psi(\mathbf{r}) = f\exp(i\mathbf{kr}) - \sum_n Q\frac{\exp(ik|\mathbf{r}-\mathbf{R}_n|)}{|\mathbf{r}-\mathbf{R}_n|}\varphi^n(\mathbf{R}_n), \quad (3)$$

which is the superposition of the incident plane wave $f\exp(i\mathbf{kr})$ and of the spherical waves excited by the

* The formulae of this paper will be referred to as I, followed by the relevant equation number.

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point scatterers forming the crystal [the second term on the RHS of (3)]. The diffraction amplitude of the nth atom is $Q\varphi^n(\mathbf{R}_n)$, where Q is the diffraction length of the scatterers (atoms), and the effective field $\varphi^n(\mathbf{R}_n)$ incident on the nth atom must satisfy equation (I.8),

$$\varphi^n(\mathbf{R}_n)=f\exp(i\mathbf{k}\mathbf{R}_n)-\sum_{m\neq n}{}' Q\frac{\exp(ik|\mathbf{R}_m-\mathbf{R}_n|)}{|\mathbf{R}_m-\mathbf{R}_n|}\varphi^m(\mathbf{R}_m). \quad (4)$$

To compare our formulae for the diffraction of scalar waves with those for X-ray diffraction (*e.g.* Litzman, 1978) we must put

$$Q_0=-k^2\Omega_0\chi_0/4\pi, \quad (5)$$

where $Q_0=Q/(1-ikQ)$ [*cf.* (12*b*)], χ_0 is 4π times the Fourier component of the susceptibility of the crystal, and $\Omega_0=(\mathbf{a}_1\times\mathbf{a}_2).\mathbf{a}_3$ is the primitive cell volume.

The solution of the system of equations (4) can be written as [(I.28)]

$$\varphi^{n_1n_2n_3}(\mathbf{R}_n)=\exp[i\mathbf{k}^{\parallel}(n_1\mathbf{a}_1+n_2\mathbf{a}_2)]\sum_j c_j'\exp(in_3\psi_j)=\sum_j c_j'\exp(i\boldsymbol{\kappa}_j\mathbf{R}_n), \quad (6)$$

where

$$\boldsymbol{\kappa}_j=\mathbf{k}^{\parallel}+(1/2\pi)(\psi_j-\mathbf{k}^{\parallel}\mathbf{a}_3)\mathbf{g}_3. \quad (7)$$

Evaluating $\varphi^n(\mathbf{R}_n)$ from (4) and inserting the results into (3) we get for the wave function $\Psi^r(\mathbf{r})$ of the reflected particles [(I.29)]

$$\Psi^r(\mathbf{r})=\sum_{pq}\frac{1}{K_{pqz}(\mathbf{k})}R_r^{\infty}(\theta_{pq}^-)\times\exp(i\theta_{pq}^-)\exp[i\mathbf{K}_{pq}^-(\mathbf{k})\mathbf{r}] \quad \text{for } z<0, \quad (8)$$

where, according to (I.55),

$$R_r^{\infty}(\theta_{pq}^-)=-fk_z\exp(-i\mathbf{k}\mathbf{a}_3)R_1(\theta_{pq}^-)R_2(\theta_{pq}^-) \quad (9)$$

with

$$R_1(\theta_{pq}^-)=\frac{\exp(i\psi_1)-\exp(i\theta_{00}^+)}{\exp(i\psi_1)-\exp(i\theta_{pq}^-)}, \quad (10a)$$

$$R_2(\theta_{pq}^-)=\prod_{j=2}^{n}\frac{\exp(i\psi_j)-\exp(i\theta_{00}^+)}{\exp(i\psi_j)-\exp(i\theta_{pq}^-)}\times\frac{\exp(i\theta_j^+)-\exp(i\theta_{pq}^-)}{\exp(i\theta_j^+)-\exp(i\theta_{00}^+)}. \quad (10b)$$

Quantities ψ_j appearing in (10*a*) and (10*b*) are solutions of the dispersion relation [(I.57)]

$$1+QS'(\mathbf{k}^{\parallel})-\sum_{pq}^{(n)}b_{pq}\left[\frac{\exp(i\theta_{pq}^+)}{\exp(i\psi)-\exp(i\theta_{pq}^+)}+\frac{\exp(-i\theta_{pq}^-)}{\exp(-i\psi)-\exp(-i\theta_{pq}^-)}\right]=0, \quad (11a)$$

where $S'(\mathbf{k}^{\parallel})$ is the two-dimensional lattice sum. Equation (11*a*) can be cast in another form (I.53) (see Appendix)

$$\sum_{pq}^{(n)}b_{pq}^0\left[\frac{i\sin(a_{3z}K_{pqz})}{\cos(\psi-\mathbf{a}_3^{\parallel}\mathbf{k}_{pq}^{\parallel})-\cos(a_{3z}K_{pqz})}+\Phi\left(\frac{iK_{pqz}}{2B}\right)\right]+PQ_0=1. \quad (11b)$$

In (11*a*) and (11*b*) we have introduced

$$b_{pq}=-\frac{2\pi iQ}{|\mathbf{a}_1\times\mathbf{a}_2|K_{pqz}}, \quad b_{pq}^0=-\frac{2\pi iQ_0}{|\mathbf{a}_1\times\mathbf{a}_2|K_{pqz}}, \quad (12a)$$

$$Q=\frac{Q_0}{1+ikQ_0},$$

and (12*b*)

$$\Phi(x)=(2/\pi^{1/2})\int_0^x\exp(-t^2)\,\mathrm{d}t.$$

The form of P can be seen when comparing (11*b*) and (I.53) (see Appendix).

In a semi-infinite crystal only the roots of the dispersion relation (11*a*) and/or (11*b*) for which Im $\psi_j>0$ have a physical meaning and thus they are to be inserted into (10*a*) and (10*b*).

The probability current densities in the incident and reflected [in the direction of $\mathbf{K}_{pq}^-(\mathbf{k})$] waves are given by

$$\mathbf{j}_{\rm inc}=(\hbar\mathbf{k}/m)|f|^2, \quad (13a)$$

$$\mathbf{j}_r(\theta_{pq}^-)=[\hbar\mathbf{K}_{pq}^-(\mathbf{k})/m]|K_{pqz}^{-1}R_r^{\infty}(\theta_{pq}^-)|^2=[\hbar\mathbf{K}_{pq}^-(\mathbf{k})/m]|(k_z/K_{pqz})R_1(\theta_{pq}^-)R_2(\theta_{pq}^-)|^2|f|^2, \quad (13b)$$

where $R_1(\theta_{pq}^-)$ and $R_2(\theta_{pq}^-)$ are defined by (10*a*) and (10*b*), respectively. The reflectivity $\mathcal{R}(\theta_{pq}^-)$ in the direction of the vector $\mathbf{K}_{pq}^-(\mathbf{k})$ is defined as

$$\mathcal{R}(\theta_{pq}^-)=|\mathbf{j}_r(\theta_{pq}^-)|\cos\xi_{pq}/|\mathbf{j}_{\rm inc}|\cos\xi \quad (14)$$

where

$$\cos\xi=k_z/k,\qquad \cos\xi_{pq}=K_{pqz}(\mathbf{k})/|\mathbf{K}_{pq}^-(\mathbf{k})|=K_{pqz}/k \quad (15)$$

(*cf.* Fig. 1 - generally the vector $\mathbf{k}$ and $\mathbf{K}_{pq}^-(\mathbf{k})$ and $\mathbf{e}_3$ do not lie in the same plane). After inserting (13*a*) and (13*b*) into definition (14) we obtain

$$\mathcal{R}(\theta_{pq}^-)=|R_1(\theta_{pq}^-)|^2|R_2(\theta_{pq}^-)|^2k_z/K_{pqz}, \quad (16)$$

which is our final exact formula for reflection in the direction of $\mathbf{K}_{pq}^-(\mathbf{k})$.

3. Approximate reflection curves

Formulae (10*a*), (10*b*) and (16) for the reflection given in the preceding section are generally valid without any limitation concerning the energy, direction of incident radiation or the strength of the interaction between the radiation and the crystal consisting of Fermi δ potentials.

The crucial point of the analysis of our general results is to find the character of the solutions ψ_j of the dispersion equation (11a) and/or (11b). The functions on the LHS's of these equations have poles for $\psi = \theta^{\pm}_{pq} + n2\pi$ (n integer) and we can see that each solution ψ_j of the dispersion equation may be associated with one pole $\theta^{\pm}_{pq}$. The distance $|\psi^{\pm}_{pq} - \theta^{\pm}_{pq}|$ is the smaller, the greater is the value of the quantity

$$\left|\frac{1-PQ_0}{b^0_{pq}}\right| = \left|\frac{1-PQ_0}{2\pi Q_0}\right| |\mathbf{a}_1 \times \mathbf{a}_2| K_{pqz}. \qquad (17)$$

The quantity (17) has a large value if the ratio Q_0/a is small (*i.e.* if the interaction between the radiation and the crystal is weak) and the parameter aK_{pqz} is not 'too small', which is fulfilled in the case when the wavelength of the radiation is equal in order to the lattice parameter and when the waves propagating near the surface are not included (the grazing-incidence region will be dealt with in § 4). In an absorbing crystal Im $Q_0 < 0$ holds and a simple consideration shows that the roots fulfilling the condition Im $\psi_j > 0$ lie near θ^{+}_{pq}. The root near θ^{+}_{00} will then be denoted by ψ_1.

In what follows we will suppose that

$$R_2(\theta^{-}_{pq}) \simeq 1 \quad \text{for all } (p, q). \qquad (18)$$

This is true, for example, if $|\psi_j - \theta^{+}_j| \ll 1$ for all $j \geq 2$. Then we shall be engaged in the evaluation of $R_1(\theta^{-}_{pq})$ only. $R_2(\theta^{-}_{pq}) = 1$ means physically that from the many waves $\exp(i\boldsymbol{\kappa}_j \mathbf{R}_m)$ in (6) only one with the wavevector $\boldsymbol{\kappa}_1 = \mathbf{k}^{\parallel} + (\psi_1 - \mathbf{k}^{\parallel}\mathbf{a}_3)\mathbf{g}_3/(2\pi)$ [*cf.* (7)] influences the reflectivity substantially. The wavevector $\boldsymbol{\kappa}_1$ of this wave should be evaluated of course from the exact dispersion relation (11a) and/or (11b).

The wavevector $\mathbf{k}_B$ of the incident wave is said to satisfy the Bragg condition for reflection in the direction of the vector $\mathbf{K}^{-}_{rs}(\mathbf{k}_B)$ if

$$[\mathbf{K}^{-}_{rs}(\mathbf{k}_B)]^2 = (\mathbf{k}_B + r\mathbf{g}_1 + s\mathbf{g}_2 - j\mathbf{g}_3)^2 \quad (r, s, j \text{ integers}). \qquad (19a)$$

In the Appendix it is shown that the condition (19a) is equivalent to

$$\theta^{+}_{00}(\mathbf{k}_B) = \theta^{-}_{rs}(\mathbf{k}_B) + 2\pi j. \qquad (19b)$$

We shall suppose that condition (19a) and/or (19b) is satisfied for one index triple (r, s, j) only. This condition, together with approximation (18), is similar to but not so restrictive as the usual two-beam approximation in the conventional Laue dynamical theory of diffraction since ψ_1 should be evaluated from the exact (multiple-beam) dispersion relation (11a) and/or (11b).

If the wavevector $\mathbf{k}$ of the incident wave is in the neighbourhood of vector $\mathbf{k}_B$, then

$$\theta^{+}_{00}(\mathbf{k}) = \theta^{-}_{rs}(\mathbf{k}) + 2\pi j + \eta, \quad j \text{ integer}, \quad |\eta| \ll 1. \qquad (20)$$

If $\eta \to 0$ the poles θ^{+}_{00} and θ^{-}_{rs} in the dispersion relation (11a) and/or (11b) coincide and the value of $R_1(\theta^{-}_{rs})$ defined by (10a), *i.e.*

$$R_1(\theta^{-}_{rs}) = \frac{\exp(i\psi_1) - \exp(i\theta^{+}_{00})}{\exp(i\psi_1) - \exp(i\theta^{+}_{00} - i\eta)}, \qquad (21)$$

should be handled very carefully.* For this purpose we pick out in dispersion equation (11b) the terms $(p, q) = (0, 0)$, (r, s) from the sum $\sum_{pq}$, so that this dispersion relation is rearranged in the form

$$\begin{aligned} &b^0_{00}\frac{\exp(i\theta^{+}_{00})}{\exp(i\psi) - \exp(i\theta^{+}_{00})} + b^0_{rs}\frac{\exp(-i\theta^{-}_{rs})}{\exp(-i\psi) - \exp(-i\theta^{-}_{rs})} \\ &+ b^0_{00}\left[1 + \frac{\exp(-i\theta^{-}_{00})}{\exp(-i\psi) - \exp(-i\theta^{-}_{00})} + \Phi\left(\frac{iK_{00z}}{2B}\right)\right] \\ &+ b^0_{rs}\left[1 + \frac{\exp(i\theta^{+}_{rs})}{\exp(i\psi) - \exp(i\theta^{+}_{rs})} + \Phi\left(\frac{iK_{rsz}}{2B}\right)\right] \\ &+ \sum_{\substack{(pq)\neq \\ (0,0),(r,s)}}{}' b^0_{pq}\left[\frac{i\sin(a_{3z}K_{pqz})}{\cos(\psi - \mathbf{a}_3\mathbf{k}^{\parallel}_{pq}) - \cos(a_{3z}K_{pqz})} \right. \\ &\left. + \Phi\left(\frac{iK_{pqz}}{2B}\right)\right] + PQ_0 = 1. \end{aligned} \qquad (22)$$

To evaluate $R_1(\theta^{-}_{rs})$ we have to find the solution ψ_1 of (22) near θ^{+}_{00} and to insert it into the the RHS of (21). This procedure can be simplified by using the following rule:

Let ψ fulfil the equation

$$F(\psi) - b^0_{00}\frac{\exp(i\theta^{+}_{00})}{\exp(i\psi) - \exp(i\theta^{+}_{00})} - b^0_{rs}\frac{\exp(-i\theta^{-}_{rs})}{\exp(-i\psi) - \exp(-i\theta^{-}_{rs})} = 0. \qquad (23a)$$

Then†

$$\frac{\exp(i\psi) - \exp(i\theta^{+}_{00})}{\exp(i\psi) - \exp(i\theta^{-}_{rs})}\exp(-i\eta/2)\frac{\beta_{rs}}{(\beta_{00}\beta_{rs})^{1/2}} = Y(\psi) \mp [Y^2(\psi) - 1]^{1/2} \qquad (23b)$$

* The considerations following equation (I.57) in our former paper (Litzman, 1986) are incorrect. The limit of the RHS of (21) for $\eta \to 0$ should be performed as in the present paper.

† Expressing from (23a) the term $\exp(i\psi)$ as a function of $F(\psi)$ and inserting it into the expression $[\exp(i\psi) - \exp(i\theta^{+}_{00})]/[\exp(i\psi) - \exp(i\theta^{-}_{rs})]$ we get after simple algebraic manipulations the resulting equations (23b).

holds, where

$$\beta_{pq} = -ib^0_{pq} = -\frac{2\pi Q_0}{|\mathbf{a}_1 \times \mathbf{a}_2| K_{pqz}}, \quad (24)$$

$$Y(\psi) = \frac{\exp(i\eta/2)(\beta_{00}+\beta_{rs}) + 2F(\psi)\sin(\eta/2)}{2(\beta_{00}\beta_{rs})^{1/2}}$$
$$= \frac{\beta_{00}+\beta_{rs}}{2(\beta_{00}\beta_{rs})^{1/2}}\cos(\eta/2) + \frac{1}{(\beta_{00}\beta_{rs})^{1/2}}$$
$$\times [i\beta_{00}/2 + i\beta_{rs}/2 + F(\psi)]\sin(\eta/2) \quad (25)$$

with η defined by (20).

Taking into account (10a), (22), (23a), (23b), (24) and (25) we can write

$$R_1(\theta^-_{rs})\frac{\exp(-i\eta/2)\beta_{rs}}{(\beta_{00}\beta_{rs})^{1/2}} = Y(\psi_1) \mp [Y^2(\psi_1) - 1]^{1/2}, \quad (26)$$

where

$$Y(\psi_1) = \frac{\beta_{00}+\beta_{rs}}{2(\beta_{00}\beta_{rs})^{1/2}}\cos(\eta/2) + \frac{1}{(\beta_{00}\beta_{rs})^{1/2}} H_{rs}(\psi_1)\sin(\eta/2) \quad (27)$$

with

$$H_{rs}(\psi_1) = 1 - PQ_0 + \frac{2\pi i Q_0}{|\mathbf{a}_1\times\mathbf{a}_2|}\sum_{\substack{(pq)\neq\\(0,0)(rs)}}{}' \frac{1}{K_{pqz}}$$
$$\times\left[\frac{i\sin(a_{3z}K_{pqz})}{\cos(\psi_1 - \mathbf{a}_3^{\parallel}\mathbf{k}^{\parallel}_{pq}) - \cos(a_{3z}K_{pqz})} + \Phi\left(\frac{iK_{pqz}}{2B}\right)\right]$$
$$- b^0_{00}\left\{\frac{i\sin(\psi_1 - \theta^-_{00})}{2[1-\cos(\psi_1-\theta^-_{00})]} + \Phi\left(\frac{iK_{00z}}{2B}\right)\right\}$$
$$- b^0_{rs}\left\{\frac{-i\sin(\psi_1-\theta^+_{rs})}{2[1-\cos(\psi_1-\theta^+_{rs})]} + \Phi\left(\frac{iK_{rsz}}{2B}\right)\right\}. \quad (28)$$

From (26) it follows that

$$|R_1(\theta^-_{rs})|^2 k_z/K_{rsz} = |Y(\psi_1) \mp [Y^2(\psi_1)-1]^{1/2}|^2$$

so that for the reflectivity $\mathcal{R}(\theta^-_{rs})$ defined by (16) we now get

$$\mathcal{R}(\theta^-_{rs}) = |Y(\psi_1) \mp [Y^2(\psi_1)-1]^{1/2}|^2 |R_2(\theta^-_{rs})|^2, \quad (29)$$

where according to approximation (18) $R_2(\theta^-_{rs}) \doteq 1$.

To get the exact result for $R_1(\theta^-_{rs})$ one needs to insert the solution ψ_1 of the dispersion relation (11a) and/or (11b) near the pole θ^+_{00} into the expression for $H_{rs}(\psi_1)$ defined in (28). In the term $H_{rs}(\psi_1)$ the interaction of the wave $\exp(i\boldsymbol{\kappa}_1\mathbf{R}_m)$ with all waves $\exp(i\boldsymbol{\kappa}_j\mathbf{R}_m)$ with $j \neq 1$ is hidden since the dispersion relation from which ψ_1 should be computed takes into account all waves excited by the incident wave in the crystal. But in some approximation we can put

$$H_{rs}(\psi_1) \doteq H_{rs}(\theta^+_{00}) \quad \text{or} \quad H_{rs}(\psi_1) \doteq 1. \quad (30a, b)$$

Then $Y(\psi_1)$ defined by (27), needed for evaluation of the reflectivity (29), is expressed in approximation (30b) by the known quantities β_{00}, β_{rs} and η only. For η we have found in the Appendix that

$$\eta = a_{3z}[K_{rsz}(\mathbf{k}) - K_{rsz}(\mathbf{k}_B) + k_z - k_{Bz}]$$
$$= a_{3z}k(\cos\xi' - \cos\xi'_B + \cos\xi - \cos\xi_B), \quad (31a)$$

where

$$\cos\xi = k_z/k, \quad \cos\xi' \equiv \cos\xi_{rs} = K_{rsz}(\mathbf{k})/k,$$
$$\cos\xi_B = k_{Bz}/k, \quad \cos\xi'_B = K_{rsz}(\mathbf{k}_B)/k. \quad (31b)$$

Thus η can be considered to be known from the experimental arrangement.

Finally let us mention that all formulae of this section are valid for skew reflections as well.

4. Comparison with Laue's theory

To compare our results with those of other authors we wish to express η given by (31a) as a function of the deviation $\Delta\xi = \xi - \xi_B$ (see Fig. 1). To this goal we shall assume, as in the conventional theory of diffraction, that all the vectors $\mathbf{k}$, $\mathbf{k}_B$, $\mathbf{K}^-_{rs}(\mathbf{k})$, $\mathbf{K}^-_{rs}(\mathbf{k}_B)$ and $\mathbf{e}_3$

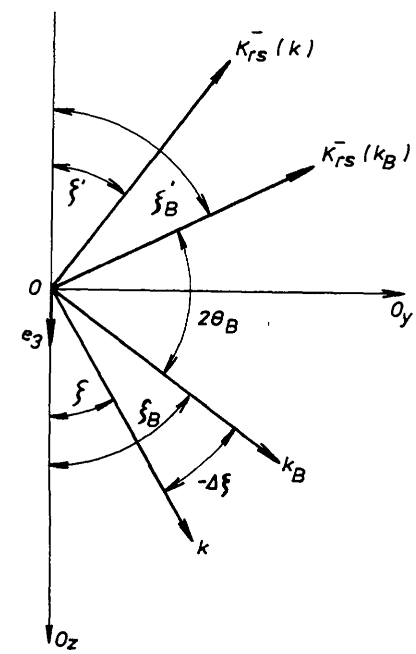

Fig. 1. The geometry of the reflection of X-rays on a crystal. $\mathbf{k}$, $\mathbf{k}_B$ and $\mathbf{K}^-_{rs}(\mathbf{k})$, $\mathbf{K}^-_{rs}(\mathbf{k}_B)$ are the wavevectors of the incident and of the reflected waves, respectively. The wavevectors of the reflected waves do not lie generally in the plane of incidence. $|\xi|$, $|\xi'|$, $|\xi_B|$, $|\xi'_B| \leq \pi/2$. The vectors $\mathbf{k}_B$ and $\mathbf{K}^-_{rs}(\mathbf{k}_B)$ fulfil the Bragg reflection condition.

lie in the same plane so that we can write $k_z = k\cos(\xi_B+\Delta\xi)$, $k^{\parallel} = k\sin(\xi_B+\Delta\xi)$ *etc.* Introducing the deviation $\Delta\xi$ into our formula for small $\Delta\xi$ we find after lengthy but easy algebraic manipulations that

$$\eta = -\frac{a_{3z}k}{\cos\xi'_B}\{\Delta\xi\sin 2\theta_B - \tfrac{1}{2}[\cos 2\theta_B - (\cos\xi_B/\cos\xi'_B)^2](\Delta\xi)^2\} + O(\Delta\xi)^3 \qquad (32)$$

where

$$2\theta_B = \pi - (\xi_B + \xi'_B).$$

To be able to make comparison of our results with formulae for X-ray diffraction we have further to take into account (5). Using it in the second-order approximation in $\Delta\xi$ we obtain from (27)

$$\begin{aligned}Y(\psi_1) &= \{\chi_0/[2(\chi_0^2)^{1/2}]\}\\ &\quad\times[(\cos\xi'/\cos\xi)^{1/2} + (\cos\xi/\cos\xi')^{1/2}]\\ &\quad-(\chi_0^2)^{-1/2}H_{rs}(\psi_1)(\cos\xi\cos\xi')^{1/2}\\ &\quad\times(\sin 2\theta_B/\cos\xi'_B)\Delta\xi + \tfrac{1}{2}(\chi_0^2)^{-1/2}\\ &\quad\times[(\cos\xi\cos\xi')^{1/2}/\cos\xi'_B]\{-\tfrac{1}{8}(a_{3z}k)^2\chi_0\\ &\quad\times[(\cos\xi)^{-1} + (\cos\xi')^{-1}]\sin^2 2\theta_B/\cos\xi'_B\\ &\quad+H_{rs}(\psi_1)[\cos 2\theta_B - (\cos\xi_B/\cos\xi'_B)^2]\}(\Delta\xi)^2.\end{aligned} \qquad (33)$$

(i) *Reflection coefficient*

Pinsker (1982) gives for the reflection coefficient of a semi-infinite crystal a formula* quite similar to our equation (29)

$$R_B = |\chi_h/\chi_{\bar h}|\,|y \pm (y^2-1)^{1/2}|^2 \qquad (\text{P.8.89})$$

but with

$$y = (\beta/2C)(\chi_h\chi_{\bar h})^{-1/2}(\gamma_0/|\gamma_h|)^{1/2} \qquad (\text{P.8.76})$$

$$\beta = 2\alpha - \chi_0(1-\gamma_h/\gamma_0), \qquad (\text{P.3.9})$$

$$\alpha = \Delta\vartheta\sin 2\vartheta, \qquad (\text{P.3.6})$$

$$C = 1 \quad\text{or}\quad |\cos 2\vartheta|. \qquad (\text{P.2.74})$$

Pinsker's $\Delta\vartheta$ and ϑ correspond to our $\Delta\xi$ and θ_B, respectively.

In our case of point scatterers we have to put $C = 1$ and $\chi_0 = \chi_h$ for all h, and then we finally obtain from (P.8.76) (in Pinsker's notation)

$$y = -\frac{\chi_0}{2(\chi_0^2)^{1/2}}\left[\left(\frac{\gamma_0}{|\gamma_h|}\right)^{1/2} + \left(\frac{|\gamma_h|}{\gamma_0}\right)^{1/2}\right] + \frac{1}{(\chi_0^2)^{1/2}}\left(\frac{\gamma_0}{|\gamma_h|}\right)^{1/2}\Delta\vartheta\sin 2\vartheta. \qquad (34)$$

* Formulae quoted from Pinsker (1982) will be referred to as P, followed by the relevant equation number. We conserve their original notation.

Putting in (33) $H_{rs}(\psi_1) = 1$ and as usual $\cos\xi = \cos\xi_B$, $\cos\xi' = \cos\xi'_B$ and neglecting terms of order $(\Delta\xi)^2$ we can see that the RHS's of (33) and (34) differ in sign only, which is unimportant for the reflectivity given by (P.8.89) and by our equation (29).

Terms of higher order in $\Delta\xi$ in the expression for the reflectivity were discussed in connection with X-ray diffraction at Bragg angles near $\pi/2$ when the term $\Delta\xi\sin 2\theta_B$ in (33) is comparable with the term of the order $(\Delta\xi)^2$. Brümmer, Höche & Nieber (1979) keep the validity of equation (P.8.89) [which agrees with our general result (29)] but improve some approximations used in the conventional theory by deducing (P.3.6) for α. They suggest for α the expression [see equation (12) of Brümmer *et al.* (1979)]

$$\begin{aligned}\alpha &= [1 + 2(\theta_B-\theta)^2\sin^2\theta_B + 2(\theta_B-\theta)\sin 2\theta_B]^{1/2} - 1\\ &\doteq (\theta_B-\theta)^2\sin^2\theta_B + (\theta_B-\theta)\sin 2\theta_B,\end{aligned} \qquad (\text{Brü.12})$$

where θ is the glancing angle, so that $\theta_B - \theta = \Delta\xi$.

For Bragg angles near $\pi/2$ the term in (33) containing $\sin^2 2\theta_B$ can be neglected. Further we express the coefficient of $(\Delta\xi)^2$ in (33) in the form

$$\begin{aligned}&\cos 2\theta_B - (\cos\xi_B/\cos\xi'_B)^2\\ &\quad= -2\sin^2\theta_B - (2\sin 2\theta_B/\cos^2\xi'_B)\\ &\quad\quad\times\cos[(\xi'_B-\xi_B)/2]\sin[(\xi'_B-\xi_B)/2].\end{aligned} \qquad (35)$$

For $\theta_B \simeq \pi/2$ the angles ξ_B, ξ'_B are very small and thus the second term on the RHS of (35) can be omitted. When we put $H_{rs}(\psi_1) = 1$ we finally obtain from (33)

$$\begin{aligned}Y(\psi_1) &= \tfrac{1}{2}\chi_0(\chi_0^2)^{-1/2}\\ &\quad\times[(\cos\xi'/\cos\xi)^{1/2} + (\cos\xi/\cos\xi')^{1/2}]\\ &\quad-(\chi_0^2)^{-1/2}[(\cos\xi\cos\xi')^{1/2}/\cos\xi'_B]\\ &\quad\times[(\sin 2\theta_B)\Delta\xi + \sin^2\theta_B(\Delta\xi)^2],\end{aligned} \qquad (36)$$

which is in agreement with the correction of Brümmer *et al.* (Brü.12).

Caticha & Caticha-Ellis (1982) follow a similar procedure to that of Brümmer, Höche & Nieber (1979). However, they do not give any analytical expressions which could be directly compared with our results. The paper of Hashizume & Nakahata (1988) agrees with our results and those of Brümmer *et al.*

(ii) *Dispersion relation and accommodation*

The fundamental task of the dynamical theory of diffraction is the evaluation of the parameters of the waves excited in the crystal by the incident radiation. For a comparison of our procedure with Laue's theory we shall follow the exposition given by Zachariasen (1946). Zachariasen gives for the wavevector of the wave within the crystal medium the formula*

$$\boldsymbol{\beta}_0 = \mathbf{k}_0^e + (k_0\delta_0/\gamma_0)\mathbf{n} \quad (Z.3.90)$$

$$= \mathbf{k}_0^{e\parallel} + (\lambda^{-1}\cos\xi + \lambda^{-1}\delta_0/\cos\xi)\mathbf{n}. \quad (37)$$

The vector $\boldsymbol{\beta}_0$ corresponds to our $\boldsymbol{\kappa}_1$ defined by (7):

$$2\pi\boldsymbol{\beta}_0 = \boldsymbol{\kappa}_1 = \mathbf{k}^{\parallel} + (2\pi)^{-1}(\psi_1 - \mathbf{k}^{\parallel}\mathbf{a}_3)\mathbf{g}_3. \quad (7a)$$

Comparing (37) and (7*a*) we obtain the connection between the accommodation δ_0 introduced in Laue's theory and our quantity ψ_1:

$$\psi_1 - \theta_{00}^+ = a_{3z}k\,\delta_0/\cos\xi; \quad (38a)$$

then

$$\psi_1 - \theta_{rs}^- = \psi_1 - (\theta_{00}^+ - \eta - 2\pi j)$$
$$= a_{3z}k\,\delta_0/\cos\xi + \eta + 2\pi j \quad (38b)$$

also holds.

The accommodation δ_0 satisfies (if $\chi_h = \chi_0$) the dispersion equation

$$(2\delta_0 - \chi_0)[(2/b)\delta_0 - \chi_0 + \alpha] = \chi_0^2, \quad (Z.3.119)$$

$$\alpha = 2(\theta_B - \theta)\sin 2\theta_B, \quad (Z.3.116)$$

where θ and θ_B are again the glancing angles. In the notation used in the preceding section and in (Z.3.116), equation (Z.3.119) reads

$$\delta_0^2 - (\cos\xi_B/\cos\xi'_B)\tfrac{1}{2}[2(\theta_B-\theta)\sin 2\theta_B$$
$$-\chi_0(1-\cos\xi'_B/\cos\xi_B)]\delta_0$$
$$+\chi_0(\cos\xi_B/\cos\xi'_B)[(\theta_B-\theta)/2]\sin 2\theta_B = 0. \quad (39)$$

On the other hand, inserting (7*a*), (38*a*) and (38*b*) into our exact dispersion equation (23*a*) we get for δ_0 the equation

$$F(\theta_{00}^+ + a_{3z}k\,\delta_0/\cos\xi)$$
$$- b_{00}^0[\exp(ia_{3z}k\delta_0/\cos\xi) - 1]^{-1}$$
$$- b_{rs}^0/\{\exp[-i(a_{3z}k\delta_0/\cos\xi + \eta)] - 1\}^{-1} = 0. \quad (40)$$

In the approximation

$$F(\psi_1) = 1, \quad \exp(ix) - 1 = ix \quad (41a)$$

and [see (32)]

$$\eta = -(a_{3z}k/\cos\xi'_B)\sin 2\theta_B\,\Delta\xi, \quad (41b)$$

we finally obtain from (40) [through (5)]

$$\delta_0^2 + \frac{1}{2}\frac{\cos\xi}{\cos\xi'}\left[-\chi_0\left(\frac{\cos\xi'}{\cos\xi} - 1\right)\right.$$
$$\left. -2\Delta\xi\sin 2\theta_B\frac{\cos\xi'}{\cos\xi'_B}\right]\delta_0 + \chi_0\frac{\cos\xi}{\cos\xi'_B}\frac{\Delta\xi}{2}\sin 2\theta_B = 0. \quad (42)$$

As $\Delta\xi = \xi - \xi_B = \theta_B - \theta$, then neglecting the difference between $\cos\xi$ and $\cos\xi_B$ and/or between $\cos\xi'$ and $\cos\xi'_B$ we can see that (42) agrees with (39) following from the conventional theory. From the foregoing considerations we can see how the 'conventional' dispersion equation (Z.3.119) and/or (39) is found from the 'exact' one (40). In the extended dynamical theory of diffraction one replaces the approximate dispersion equation of the second order (Z.3.119) by a more general one (*e.g.* Bedynska, 1974; Brümmer, Höche & Nieber, 1979; Kishino & Kohra 1971)

$$\left(\frac{K_0^2-k^2}{K_0^2} - \chi_0\right)\left(\frac{K_h^2-k^2}{K_h^2} - \chi_0\right) = \chi_0^2, \quad (43)$$

where $\mathbf{K}_h = \mathbf{K}_0 + \mathbf{h} = 2\pi\boldsymbol{\beta}_0 + \mathbf{h}$, which leads to an equation of the fourth order for δ_0. It is not quite clear what the connection between the approximate equation (43) and our exact one (40) is.

(iii) *Grazing reflection*

The wavevector of the specularly reflected wave is, in our notation, $(k_x, k_y, -k_z) = \mathbf{K}_{00}^-$ and, as for the intensity of the reflected beam, we need to evaluate, following (10*a*), the term

$$R_1(\theta_{00}^-) = \frac{\exp(i\psi_1) - \exp(i\theta_{00}^+)}{\exp(i\psi_1) - \exp(i\theta_{00}^-)} \quad (44)$$

where as before ψ_1 is the solution of the dispersion relation (11*a*). If $\psi_1 \simeq \theta_{00}^+$ (this is true for X-ray and neutron diffraction) the specular reflectivity $\mathcal{R}(\theta_{00}^-)$, given by [*cf.* (16) and (18)]

$$\mathcal{R}(\theta_{00}^-) = |R_1(\theta_{00}^-)|^2 \quad (45)$$

is very small except for the case when

$$\theta_{00}^- \simeq \theta_{00}^+,$$

i.e.

$$\mathbf{a}_3\mathbf{k}^{\parallel} - a_{3z}k_z \simeq \mathbf{a}_3\mathbf{k}^{\parallel} + a_{3z}k_z,$$

i.e.

$$a_{3z}k_z \simeq 0, \quad (46)$$

which is true in the grazing-incidence region. Thus the grazing-incidence specular reflection is from our point of view a special case of the Bragg reflection in which the poles θ_{00}^- and θ_{00}^+ of the dispersion equation (11*a*) nearly coincide. Then we can use the method expained in subsection (i) by putting [see (20)]

$$\eta = \theta_{00}^+(\mathbf{k}) - \theta_{00}^-(\mathbf{k}) = 2a_{3z}k_z = 2a_{3z}k\cos\xi. \quad (47)$$

The dispersion relation (11*a*) should now be rearranged in the form [*cf.* (23*a*)]

$$\tilde{F}(\psi) - b_{00}^0\left[\frac{\exp(i\theta_{00}^+)}{\exp(i\psi) - \exp(i\theta_{00}^+)}\right.$$
$$\left. + \frac{\exp(-i\theta_{00}^-)}{\exp(-i\psi) - \exp(-i\theta_{00}^-)}\right] = 0. \quad (48)$$

* Formulae quoted from Zachariasen (1946) will be referred to as Z, followed by the relevant equation number. We conserve their original notation.

Again using rule (23b) for $(r, s) = (0, 0)$ we obtain

$$\frac{\exp(i\psi_1) - \exp(i\theta_{00}^+)}{\exp(i\psi_1) - \exp(i\theta_{00}^-)} \exp(-i\eta/2) \frac{\beta_{00}}{(\beta_{00}^2)^{1/2}} = Y(\psi_1) \mp [Y^2(\psi_1) - 1]^{1/2}, \quad (49)$$

where in the approximation (30b) $Y(\psi_1)$ reads

$$Y(\psi_1) = -\frac{Q_0}{(Q_0^2)^{1/2}} + \left(\frac{\Omega_0}{2\pi(Q_0^2)^{1/2}} + \frac{1}{2} a_{3z}^2 \frac{Q_0}{(Q_0^2)^{1/2}}\right) \times k^2 \cos^2 \xi + O(\eta^4). \quad (50)$$

Since for neutron diffraction the parameter Q_0/a is very small we can write

$$Y(\psi_1) \doteq -\frac{Q_0}{(Q_0^2)} + \frac{\Omega_0}{2\pi(Q_0^2)^{1/2}} k^2 \cos^2 \xi. \quad (51)$$

Introducing (51) into (49) we get for the grazing-incidence reflectivity $\mathcal{R}(\theta_{00}^-)$ defined by (45) the same formula as given by Sears (1978). But let us emphasize that the approaches used by Sears and by us are quite different.

5. Concluding remarks

Methods for making more precise some formulae of the 'conventional' dynamical theory of diffraction from the point of view of the 'extended' theory has been discussed in many papers in the framework of Laue's method. In the present paper we have tried to build up an extended dynamical theory of diffraction using Ewald's fundamental ideas.

The starting points of our procedure were the exact formulae (9), (10a), (10b), (11a) and (11b) for the reflection on a semi-infinite crystal consisting of Fermi δ potentials, which are valid even in the case of a skew reflection. For the reflectivity $\mathcal{R}(\theta_{pq}^-)$ we have deduced formula (29) which in approximation $R_2(\theta_{pq}^-) = 1$ coincides formally with the well known expression (P.8.89) of Laue's conventional theory. But the meaning of parameter $Y(\psi_1)$ given by (27) and/or (33) and of parameter y defined in (P.8.76) is different. We shall show in a future paper that the formal similarity between formulae of the conventional and extended theory of Ewald is valid in the more general case of a slab of finite thickness as well.

Since we deal from the very beginning with the semi-infinite crystal bordered by a surface it turns out that a good parameter of our theory (especially at skew reflection) is not the deviation $\Delta\xi$ from the Bragg reflection position but the parameter η defined by (20) and/or (31).

A comparison of our results with those following from the conventional and extended Laue theory of diffraction has been drawn in § 4: (i) The former suggested extension of Laue's theory for Bragg angles near $\pi/2$ (Brümmer, Höche & Nieber, 1979) has been confirmed as a special case of the general formula (29). (ii) The exact dispersion relation (11a) and/or (11b) yields the conventional one (Z.3.119) and/or (39) by adopting approximations (41a) and (41b). (iii) The total reflection at grazing incidence is from our point of view formally equivalent to Bragg reflection when $\theta_{00}^+(\mathbf{k}) \simeq \theta_{00}^-(\mathbf{k})$ [see (46)].

The extreme asymmetric diffraction in the Bragg case of grazing incidence (Rustichelli, 1975; Brümmer, Höche & Nieber, 1976) means that $\theta_{00}^+(\mathbf{k}) \simeq \theta_{00}^-(\mathbf{k}) \simeq \theta_{rs}^-(\mathbf{k}) + 2\pi\mathbf{n}$, *i.e.* three poles of the dispersion equation (11a) and/or (11b) coincide. We have not discussed this case in the present paper.

The great drawback of the Ewald procedure is that the crystal is supposed to be built up of point scatterers. Thus it is difficult to take into account the atomic factors and the theory is more appropriate for neutron than for X-ray diffraction. Secondly, in the present paper the case of a simple lattice without a basis has been dealt with. The theory can, however, be extended to the case of a general crystal lattice with a basis (Litzman, 1986), but the resulting formulae are more complicated than those used in our present paper. Nevertheless, we think that the formulae deduced here can be used to test different approximations adopted in the extended dynamical theory of diffraction.

APPENDIX

1. Bragg reflection condition

Each vector $\mathbf{K}_{rs}^-(\mathbf{k})$ defined by (2b) can be expresed in two coordinate systems, *viz* $(\mathbf{b}_1, \mathbf{b}_2, \mathbf{e}_3)$ or $(\mathbf{g}_1, \mathbf{g}_2, \mathbf{g}_3)$:

$$\begin{aligned}\mathbf{K}_{rs}^-(\mathbf{k}) &= \mathbf{k}^{\parallel} + r\mathbf{b}_1 + s\mathbf{b}_2 - \mathbf{e}_3 K_{rsz}(\mathbf{k}) \\ &= \mathbf{k} + y_1\mathbf{g}_1 + y_2\mathbf{g}_2 + y_3\mathbf{g}_3 \\ &= \mathbf{k}^{\parallel} + y_1\mathbf{g}_1^{\parallel} + y_2\mathbf{g}_2^{\parallel} + \mathbf{k}^{\perp} + y_1\mathbf{g}_1^{\perp} + y_2\mathbf{g}_2^{\perp} + y_3\mathbf{g}_3^{\perp}.\end{aligned}$$

As $\mathbf{b}_1 = \mathbf{g}_1^{\parallel}$, $\mathbf{b}_2 = \mathbf{g}_2^{\parallel}$ hold we get $y_1 = r$, $y_2 = s$, and thus

$$\begin{aligned}\mathbf{K}_{rs}^-(\mathbf{k}) &= \mathbf{k}^{\parallel} + r\mathbf{b}_1 + s\mathbf{b}_2 - \mathbf{e}_3 K_{rsz}(\mathbf{k}) \\ &= \mathbf{k} + r\mathbf{g}_1 + s\mathbf{g}_2 + y_3\mathbf{g}_3. \end{aligned} \quad (A1)$$

Multiplying the last equation by $\mathbf{a}_3$ we obtain [*cf.* (2e)]

$$\theta_{rs}^-(\mathbf{k}) = \theta_{00}^+(\mathbf{k}) + 2\pi y_3(\mathbf{k}),$$

so that (A1) may be read as

$$\mathbf{K}_{rs}^-(\mathbf{k}) = \mathbf{k} + r\mathbf{g}_1 + s\mathbf{g}_2 + \frac{\theta_{rs}^-(\mathbf{k}) - \theta_{00}^-(\mathbf{k})}{2\pi} \mathbf{g}_3. \quad (A2)$$

The vector $\mathbf{k}_B$ satisfies the Bragg reflection condition if

$$(\mathbf{k}_B + r\mathbf{g}_1 + s\mathbf{g}_2 - n\mathbf{g}_3)^2 = k^2 \quad (r, s, n \text{ integers})$$

holds. Since $|\mathbf{K}_{rs}^-(\mathbf{k})|^2 = k^2$ must always hold [see

$(2d)$], it can be seen from $(A2)$ that the Bragg reflection condition for the vector $\mathbf{K}_{rs}^{-}(\mathbf{k}_B)$ reads

$$\theta_{00}^{+}(\mathbf{k}_B) = \theta_{rs}^{-}(\mathbf{k}_B) + 2\pi n. \qquad (A3)$$

2. η parameter

We have introduced η in (20):

$$\eta = \theta_{00}^{+}(\mathbf{k}) - \theta_{rs}^{-}(\mathbf{k}) - n2\pi. \qquad (A4)$$

Let $\mathbf{k}_B$ be a vector in the Bragg reflection position, *i.e.*

$$\mathbf{K}_{rs}^{-}(\mathbf{k}_B) = \mathbf{k}_B + r\mathbf{g}_1 + s\mathbf{g}_2 - n\mathbf{g}_3, \qquad (A5a)$$

$$\theta_{00}^{+}(\mathbf{k}_B) = \theta_{rs}^{-}(\mathbf{k}_B) + 2\pi n. \qquad (A5b)$$

Let $\mathbf{k}$ be a vector in the neighbourhood of the vector $\mathbf{k}_B$. Then using $(A4)$, $(A5b)$ and $(2e)$ we obtain

$$\begin{aligned}\eta &= \theta_{00}^{+}(\mathbf{k}) - \theta_{rs}^{-}(\mathbf{k}) - 2\pi n \\ &= \theta_{00}^{+}(\mathbf{k}) - \theta_{rs}^{-}(\mathbf{k}) - [\theta_{00}^{+}(\mathbf{k}_B) - \theta_{rs}^{-}(\mathbf{k}_B)] \\ &= \mathbf{a}_3\mathbf{k} - \mathbf{a}_3\mathbf{K}_{rs}^{-}(\mathbf{k}) - [\mathbf{a}_3\mathbf{k}_B - \mathbf{a}_3\mathbf{K}_{rs}^{-}(\mathbf{k}_B)] \\ &= a_{3z}[K_{rsz}(\mathbf{k}) - K_{rsz}(\mathbf{k}_B) + k_z - k_{Bz}], \qquad (A6)\end{aligned}$$

so that the η parameter is a function of the known vectors $\mathbf{k}_B$ and $\mathbf{k}$.

3. Correction of a misprint in equation (I.53)

There is a misprint in equation (I.53); the term

$$-\frac{1}{K_{pqz}}\,\Phi\left(\frac{iK_{pqz}}{2B}\right)$$

should be replaced by

$$-\frac{i}{K_{pqz}}\,\Phi\left(\frac{iK_{pqz}}{2B}\right).$$

4. P parameter appearing in equation (11b)

When comparing (11b) and (I.53) we obtain

$$\begin{aligned}P &= ik\Phi(ik/2B) + (2/\pi^{1/2})B\exp(k^2/4B^2) \\ &\quad - \sum_{\substack{(n_1n_2)\\ \neq(00)}}{}' \frac{\exp[i\mathbf{k}^{\parallel}(n_1\mathbf{a}_1 + n_2\mathbf{a}_2)]}{2|n_1\mathbf{a}_1 + n_2\mathbf{a}_2|} \\ &\quad \times\{\exp(-ik|n_1\mathbf{a}_1 + n_2\mathbf{a}_2|) \\ &\quad \times[1 - \Phi(|n_1\mathbf{a}_1 + n_2\mathbf{a}_2|B - ik/2B)] + \text{c.c.}\}. \qquad (A7)\end{aligned}$$

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About the Coulomb Potential in Crystals

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Abstract

The Coulomb potential in a crystal is discussed. It is shown that its Fourier series expansion has a singularity for the $V(0, 0, 0)$ component, which is important when comparing different compounds, or when using the Coulomb potential as a probe for reactivity. Methods to calculate this term are discussed. Sum rules for multipolar moments of crystals in terms of structure factors are derived, which are of interest for

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