Acta Crystallographica Section A
Foundations of Crystallography
ISSN 0108-7673

Received 1 December 2011
Accepted 4 April 2012

Multiple wave scattering on a bordered crystal. Translation symmetry breaking and forbidden reflections

P. Dub[a]* and O. Litzman[b]

[a]Institute of Physical Engineering and CEITECH, Brno University of Technology, 616 69 Brno, Technická 2, Czech Republic, and [b]Institute of Theoretical Physics and Astrophysics, Masaryk University, 611 37 Brno, Kotlářská 2, Czech Republic. Correspondence e-mail: dub@fme.vutbr.cz

The interaction of a scalar wave (thermal neutrons) with a single Si crystal is treated using Ewald's self-consistent field method. Considering from the very beginning the two-dimensional translation symmetry of the problem, the reflectivities of allowed and forbidden reflections in the Bragg geometry valid for both coplanar and non-coplanar cases are derived. It is shown that there exists a very narrow reflectivity peak of the forbidden reflection as a result of the symmetry breaking due to a crystal surface.

1. Introduction

In the dynamical theory of diffraction of Bethe and von Laue the intensity of the reflected beam is determined by the structure factor (*e.g.* Authier, 2001; Rauch & Petraschek, 1978; Sears, 1989). Reflections for which it takes zero value are called forbidden and are of zero intensity. Based on the Darwin approach and using a very artificial model of a single ideal crystal with two identical atoms in an elementary cell, Ignatovich *et al.* (1996) have shown that the forbidden reflections are not absent but have a very small Darwin table width (DTW). In the present paper we explore the problem of forbidden reflections on a single silicon crystal utilizing the Ewald dynamical diffraction theory [see original papers by Ewald (1916,[1] 1917) and a memorial volume for P. P. Ewald (Cruickshack *et al.*, 1992)]. The Ewald concept of the dynamical diffraction theory in which the crystal is viewed as a discrete system of scatterers, and thus yields a self-consistent system of algebraic equations (Dederichs, 1972; Sears, 1989), was developed and applied both to electromagnetic waves and scalar waves (neutrons) in a series of our former papers (*e.g.* Litzman & Rózsa, 1977; Litzman, 1978, 1980, 1986; Litzman & Dub, 1982, 1990; Litzman *et al.*, 1996; Dub & Litzman, 2005). In Litzman (1986) the solution of the quantum-mechanical Ewald equations was expressed in a lucid matrix form and amplitudes of the diffracted waves were then obtained in well arranged determinant forms. The general solution of the diffraction problem found in Litzman (1986) may be applied to a lattice with an arbitrary basis. But the resulting formulae for diffracted waves in such a transparent form as derived for the crystal with one atom per cell (Dub & Litzman, 2005) have hitherto been obtained for a single atomic plane only (Dub & Litzman, 2001*b*). The challenging case of a three-dimensional lattice with a composite structure will be dealt with in the present study.

The paper is organized as follows. In §2 we expose the main ideas of the quantum-mechanical Ewald dynamical theory of diffraction and recall the general solution of the multiple scattering problem of scalar waves (neutrons) on a crystalline slab obtained by Litzman (1986). In §3 our development is applied to an ideal silicon crystal. The two-dimensional translational symmetry of the problem leads to the plane-wise summation yielding the dispersion relation with poles, the positions of which are given by the geometry and wavelength only. The confluence of two poles occurs if the Bragg condition is satisfied. In §§4 and 5 the reflection of neutrons from a semi-infinite single silicon crystal in the Bragg geometry is examined in the two-beam approximation and, in particular, in §5, it is shown that there exists a very high reflectivity peak in the forbidden reflection, *i.e.* in the direction for which the structure factor is, because of two atomic bases, zero and thus according to the Laue theory the intensity of such a reflection should be zero.

2. Basic formulae: the Ewald equations

We will study the diffraction of a scalar plane wave (neutrons)

$$\Psi_{\rm inc}(\mathbf{r}) = A\exp(i\mathbf{k}\cdot\mathbf{r}) \tag{1}$$

(where inc = incident) on a system of scattering centres fixed at points

$$\mathbf{R}^{\nu}_{\mathbf{m}} = \mathbf{R}_{\mathbf{m}} + \mathbf{r}_{\nu}, \tag{2}$$

where $\mathbf{R}_{\mathbf{m}}$, with $\mathbf{m} = (m_1, m_2, m_3)$, is a lattice point and $\mathbf{r}_{\nu}$, $\nu = 1, 2, \ldots, s$, is the position vector of atom ν in the unit cell.

[1] One hundred years ago, in early 1912, P. P. Ewald submitted his thesis *Dispersion und Doppelbrechung von Elektronengitter (Kristallen)* which was republished in *Annalen der Physik* in 1916 (Ewald, 1916). Thus our paper shall also commemorate the 100th anniversary of the first essential contribution of P. P. Ewald to the dynamical theory of diffraction.

Considering the point-like scatterers characterized by scattering lengths Q_ν the Ewald equations read (Dederichs, 1972; Sears, 1989)

$$\Psi(\mathbf{r}) = \Psi_{\rm inc}(\mathbf{r}) - \sum_{\mathbf{n},\nu} Q_\nu \frac{\exp\left(ik\left|\mathbf{r}-\mathbf{R}_{\mathbf{n}}^{\nu}\right|\right)}{\left|\mathbf{r}-\mathbf{R}_{\mathbf{n}}^{\nu}\right|}\Phi_\nu^{\mathbf{n}}(\mathbf{R}_{\mathbf{n}}^{\nu}), \quad (3a)$$

$$\Phi_\mu^{\mathbf{m}}(\mathbf{R}_{\mathbf{m}}^{\mu}) = \Psi_{\rm inc}(\mathbf{R}_{\mathbf{m}}^{\mu}) - {\sum_{\mathbf{n},\nu\neq\mathbf{m},\mu}}' Q_\nu \frac{\exp\left(ik\left|\mathbf{R}_{\mathbf{m}}^{\mu}-\mathbf{R}_{\mathbf{n}}^{\nu}\right|\right)}{\left|\mathbf{R}_{\mathbf{m}}^{\mu}-\mathbf{R}_{\mathbf{n}}^{\nu}\right|} \times \Phi_\nu^{\mathbf{n}}(\mathbf{R}_{\mathbf{n}}^{\nu}), \quad (3b)$$

where $\Psi(\mathbf{r})$ is the total field at the point $\mathbf{r}$ and $\Phi_\mu^{\mathbf{m}}(\mathbf{R}_{\mathbf{m}}^{\mu})$ is the field incident on the scatterer at $\mathbf{R}_{\mathbf{m}}^{\mu}$ (the local field). The prime in equation (3*b*) indicates that the interaction of the scatterer with its own field is excluded.

Further we will consider a crystal slab specified by a set of vectors

$$\mathbf{R}_{\mathbf{m}} = m_1\mathbf{a}_1 + m_2\mathbf{a}_2 + m_3\mathbf{a}_3, \quad (4)$$

where $m_1, m_2 = 0, \pm1, \pm2, \ldots, \pm\infty; m_3 = 0, 1, 2, \ldots, N$. The origin of the orthogonal coordinate system $Oxyz$ lies at the lattice point (0, 0, 0), the plane Oxy coincides with the entrance crystal surface plane $(\mathbf{a}_1, \mathbf{a}_2)$, and the axis Oz (the unit vector $\hat{\mathbf{e}}_3$), vectors $\mathbf{a}_3$ and $\mathbf{a}_1 \times \mathbf{a}_2$ point into the crystal. Furthermore 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.* $\mathbf{g}_i \cdot \mathbf{a}_k = 2\pi\delta_{ik}$ $(i, k = 1, 2, 3)$, whereas the lattice $(\mathbf{b}_1, \mathbf{b}_2)$ is reciprocal to the two-dimensional lattice $(\mathbf{a}_1, \mathbf{a}_2)$, *i.e.* $\mathbf{b}_i \perp \hat{\mathbf{e}}_3$, $\mathbf{b}_i \cdot \mathbf{a}_k = 2\pi\delta_{ik}$ $(i, k = 1, 2)$. Thus $\mathbf{b}_1 = \mathbf{g}_1^{||}$, $\mathbf{b}_2 = \mathbf{g}_2^{||}$, with $\mathbf{c}^{||}$ denoting the component of the vector $\mathbf{c}$ parallel to the surface.

Considering diffraction on a crystalline slab possessing the two-dimensional discrete translation symmetry in the plane $(\mathbf{a}_1, \mathbf{a}_2)$, the total field [equation (3*a*)] composed of the incident plane wave and the spherical waves excited by the scatterers may be expressed as a superposition of plane waves with the wavevectors $\mathbf{K}_{pq}^{+}(\mathbf{k})$ and $\mathbf{K}_{pq}^{-}(\mathbf{k})$ in the transmission (Laue) and reflection (Bragg) geometry, respectively, given by (Litzman, 1986)

$$\mathbf{K}_{pq}^{\pm}(\mathbf{k}) = \mathbf{k}^{||} + p\mathbf{b}_1 + q\mathbf{b}_2 \pm \mathbf{e}_3 K_{pqz}(\mathbf{k}) \quad (5)$$

with p, q being integers, where according to the photon energy conservation

$$K_{pqz}(\mathbf{k}) = +\left[k^2 - \left(\mathbf{k}^{||} + p\mathbf{b}_1 + q\mathbf{b}_2\right)^2\right]^{1/2}. \quad (6)$$

From equation (6) it can be seen that there is a finite number, say n, of different couples (p, q) (depending on the wavelength λ of the incident radiation and the angle of incidence γ) yielding $2n$ radiative waves with real $K_{pqz}(\mathbf{k})$. Other (p, q) correspond to non-radiative waves with pure imaginary $K_{pqz}(\mathbf{k})$.

Furthermore, note when studying the diffraction on a slab, three-dimensional lattice sums in equations (3*a*) and (3*b*) are decomposed into sums over n_3 in the direction perpendicular to the surface and two-dimensional ones over (n_1, n_2) in the surface plane, which may be transformed into rapidly convergent sums over (p, q) in the two-dimensional reciprocal space.

In our former paper (Dub & Litzman, 2001*a*) we found the solution of the Ewald equations in the case of the diffraction by a single atomic plane:

$$\Psi(\mathbf{r}) = A\exp(i\mathbf{k}\cdot\mathbf{r}) - \frac{2\pi i A}{\left|\mathbf{a}_1\times\mathbf{a}_2\right|} \times \sum_{p,q}\left\{\sum_{\nu=1}^{s} Q_\nu w_\nu \exp\left[-i\mathbf{r}_\nu\cdot(p\mathbf{b}_1+q\mathbf{b}_2)\right]\right\} \times \frac{1}{K_{pqz}} \begin{cases} \exp\left(i\mathbf{K}_{pq}^{-}\cdot\mathbf{r}\right) & \text{for } z<0 \\ \exp\left(i\mathbf{K}_{pq}^{+}\cdot\mathbf{r}\right) & \text{for } z>0 \end{cases}. \quad (7)$$

It is worth noting that the unit-cell structure factor

$$F(\mathbf{G}) = \sum_{\nu=1}^{s} Q_\nu \exp(-i\mathbf{r}_\nu\cdot\mathbf{G}), \quad (8)$$

where $\mathbf{G} = u_1\mathbf{g}_1 + u_2\mathbf{g}_2 + u_3\mathbf{g}_3$ is the three-dimensional reciprocal-lattice vector, is replaced in equation (7) by an expression

$$\sum_{\nu=1}^{s} Q_\nu w_\nu \exp\left[-i\mathbf{r}_\nu\cdot(p\mathbf{b}_1+q\mathbf{b}_2)\right], \quad (9)$$

where w_ν (being slightly different from unity) are amplitudes of the local field of basis atoms.

The case of a stack of planes is more complicated (see Litzman, 1986[2]) as the local field $\Phi_\mu^{\mathbf{m}}(\mathbf{R}_{\mathbf{m}}^{\mu})$ in a slab is given by the superposition of plane waves (equation *L*28),

$$\Phi_\mu^{\mathbf{m}}(\mathbf{R}_{\mathbf{m}}^{\mu}) = \exp\left[i\mathbf{k}^{||}\cdot\left(m_1\mathbf{a}_1+m_2\mathbf{a}_2+\mathbf{r}_\mu\right)\right]\frac{\left|\mathbf{a}_1\times\mathbf{a}_2\right|}{2\pi i} \times \sum_{j}^{(n)} c_j u_\mu(\psi_j)\exp(im_3\psi_j), \quad (10)$$

which are determined by parameters ψ_j related to the z components of the local-field wavevectors $\tilde{\mathbf{k}}_j = \mathbf{k}^{||} + (1/2\pi)(\psi_j - \mathbf{k}^{||}\cdot\mathbf{a}_3)\mathbf{g}_3 = \mathbf{k}^{||} + \tilde{k}_{jz}\mathbf{e}_3$ [see equation (12*b*) in Dub & Litzman (2005)]. The parameters ψ_j, amplitudes $u_\mu(\psi_j)$, $\mu = 1, 2, \ldots, s$, and coefficients c_j can be obtained when introducing equation (10) into equation (3*b*). Thus we found that ψ_j are roots of the equation (*L*18)

$$\det\mathbf{A}(\psi) = 0, \quad (11)$$

which is an analogue of the dispersion equation of the Laue theory, and $u_1(\psi_j), u_2(\psi_j), \ldots, u_s(\psi_j)$ are solutions of the homogeneous system of linear algebraic equations (*L*20)

$$\mathbf{A}(\psi_j)\begin{Vmatrix} u_1(\psi_j) \\ u_2(\psi_j) \\ \vdots \\ u_s(\psi_j) \end{Vmatrix} = 0. \quad (12)$$

Here $\mathbf{A}(\psi)$ [see (*L*19)] is a square matrix of order s which is determined by the unit cell. Furthermore c_j are given by the inhomogeneous system of linear algebraic equations (*L*37), an equivalent to boundary conditions,

[2] The prefix *L* will be used to indicate formulae from this paper.

$$\mathbf{H}\begin{Vmatrix} c_1 \\ c_2 \\ \vdots \\ c_{2n} \end{Vmatrix} = -Ak_z \exp(-i\mathbf{a}_3\cdot\mathbf{k})\begin{Vmatrix} 1 \\ 0 \\ \vdots \\ 0 \end{Vmatrix} \tag{13}$$

where $\mathbf{H}$ [see ($L35$)] is a square matrix of order $2n$, n being the number of radiative (diffracted) waves.

In the next section we will deal with designing matrices $\mathbf{A}(\psi)$ and $\mathbf{H}$ in the case of an ideal silicon crystal.

3. Multiwave matrix formulation of Ewald theory for silicon

Applying the matrix formulation of Ewald theory explained above to a silicon crystal Si(001), instead of the conventional unit cube of a face-centred cubic (f.c.c.) lattice containing four lattice points, we use the primitive unit cell[3] defined by

$$\mathbf{a}_1 = (a,0,0),\quad \mathbf{a}_2 = \left(\frac{a}{2},\frac{a}{2},0\right),\quad \mathbf{a}_3 = \left(\frac{a}{2},0,\frac{a}{2}\right), \tag{14}$$

with $a = 0.5430$ nm, two identical motionless silicon basis atoms being placed at points

$$\mathbf{r}_1 = (0,0,0,),\quad \mathbf{r}_2 = \frac{1}{2}\left(\mathbf{a}_2+\mathbf{a}_3-\frac{\mathbf{a}_1}{2}\right) = \left(\frac{a}{4},\frac{a}{4},\frac{a}{4}\right). \tag{15}$$

Then the matrix $\mathbf{A}(\psi)$ defined by ($L19$) is of order 2,

$$\mathbf{A}(\psi) = \mathbf{I}_2 - \mathbf{C} - \sum_{pq}^{(n)}\left[L\left(-\psi,-\theta_{pq}^-\right)\mathbf{B}_{pq} + L\left(\psi,\theta_{pq}^+\right)\mathbf{D}_{pq}\right], \tag{16}$$

where $\mathbf{I}_2$ is the unit matrix of order 2, the matrix $\mathbf{C}$ renders interactions among the scatterers in one crystal plane and the sum over (p,q) expresses interactions among different atomic planes. Matrices

$$\mathbf{B}_{pq} = -\frac{2\pi i}{|\mathbf{a}_1\times\mathbf{a}_2|K_{pqz}}\,{}^1\mathbf{b}_{pq}\cdot{}^2\mathbf{b}_{pq}^T \quad\text{and}$$
$$\mathbf{D}_{pq} = -\frac{2\pi i}{|\mathbf{a}_1\times\mathbf{a}_2|K_{pqz}}\,{}^1\mathbf{d}_{pq}\cdot{}^2\mathbf{d}_{pq}^T \tag{17}$$

reflecting the structure of a unit cell are dyads of order 2 formed by the column vectors (matrix $\mathbf{d}^T$ is a transpose to matrix $\mathbf{d}$)

$${}^1\mathbf{b}_{pq} = \begin{Vmatrix} 1 \\ \exp\left(i\mathbf{r}_2\cdot\mathbf{t}_{pq}^-\right)\end{Vmatrix},\quad {}^2\mathbf{b}_{pq} = Q\begin{Vmatrix} 1 \\ \exp\left(-i\mathbf{r}_2\cdot\mathbf{t}_{pq}^-\right)\end{Vmatrix},$$
$${}^1\mathbf{d}_{pq} = \begin{Vmatrix} 1 \\ \exp\left(i\mathbf{r}_2\cdot\mathbf{t}_{pq}^+\right)\end{Vmatrix},\quad {}^2\mathbf{d}_{pq} = Q\begin{Vmatrix} 1 \\ \exp\left(-i\mathbf{r}_2\cdot\mathbf{t}_{pq}^+\right)\end{Vmatrix}.$$

Here [see also equation (5)]

$$\mathbf{t}_{pq}^{\pm} = p\mathbf{b}_1 + q\mathbf{b}_2 \pm \mathbf{e}_3 K_{pqz} = \mathbf{K}_{pq}^{\pm} - \mathbf{k}^{||} \tag{18}$$

and

$$Q = Q_0/(1+ikQ_0) \tag{19}$$

[3] Using the conventional unit cube the matrix [equation (16)] would be of order 8.

with Q_0 being the bound scattering length; for silicon Q_0 = 4.1 fm (Rauch & Petraschek, 1978). Further

$$\mathbf{C} = -QS'(\mathbf{k})\mathbf{I}_2 + \sum_{pq}^{(n)}\left(-\frac{2\pi iQ}{|\mathbf{a}_1\times\mathbf{a}_2|K_{pqz}}\right) \times \begin{Vmatrix} 0 & \exp(-i\mathbf{r}_2\cdot\mathbf{t}_{pq}^-) \\ \exp(i\mathbf{r}_2\cdot\mathbf{t}_{pq}^+) & 0\end{Vmatrix}, \tag{20}$$

where $S'(\mathbf{k})$ is the intraplanar optical lattice sum given by ($L16$), which is essential for evaluating the local field,

$$S'(\mathbf{k}) = \sum_{(n_1,n_2)\neq(0,0)}{}' \frac{\exp(ik|n_1\mathbf{a}_1+n_2\mathbf{a}_2|)}{|n_1\mathbf{a}_1+n_2\mathbf{a}_2|}\exp[i\mathbf{k}^{\|}\cdot(n_1\mathbf{a}_1+n_2\mathbf{a}_2)]$$
$$= \mathrm{Re}\,S'(\mathbf{k}) + \frac{2\pi i}{|\mathbf{a}_1\times\mathbf{a}_2|}\sum_{pq\ \text{for}\ K_{pqz}\ \text{real}}\frac{1}{K_{pqz}} - ik \tag{21}$$

with $|\mathrm{Re}\,S'(\mathbf{k})| \simeq 2\pi/ak^2 \simeq 1/a$ in the region $ak \simeq 1$ (see Appendix A). Finally,

$$L(\psi,\theta) = \frac{\exp(i\theta)}{\exp(i\psi)-\exp(i\theta)} \tag{22}$$

results from the plane-wise summation, and the quantities θ_{pq}^+ and θ_{pq}^- are determined by the geometry and the wavelength only,

$$\theta_{pq}^{\pm} \equiv \theta_{pq}^{\pm}(\mathbf{k}) = \mathbf{a}_3\cdot\mathbf{K}_{pq}^{\pm}(\mathbf{k}), \tag{23}$$

where $\mathbf{K}_{pq}^{\pm}(\mathbf{k})$ is defined in equation (5) with $K_{pqz}(\mathbf{k})$ being real. Thus the sum over (p,q) on the right-hand side of equation (16) is carried out over a finite number n of different couples (p,q), which is denoted by $\sum_{pq}^{(n)}$.

Let us note that the diagonal matrix elements A_{11} and A_{22} in equation (16) describe semi-infinite crystals with one atomic basis, and the non-diagonal ones A_{12} and A_{21} containing terms $\exp(\pm i\mathbf{r}_2\cdot\mathbf{t}_{pq}^{\pm})$ render the interaction of these two crystals shifted by the vector $\mathbf{r}_2$.

By inserting the expressions of Q and $S'(\mathbf{k})$ [equations (19) and (21)] into equation (20) we can express the matrix $\mathbf{A}(\psi)$ through a matrix $\mathbf{A}^0(\psi)$ which is determined by the bound scattering length Q_0,

$$\mathbf{A}(\psi) = \frac{Q}{Q_0}\mathbf{A}^0(\psi). \tag{24}$$

Here

$$\mathbf{A}^0(\psi) = \mathbf{I}_2 - \mathbf{C}^0 - \sum_{pq}^{(n)}\left[L\left(-\psi,-\theta_{pq}^-\right)\mathbf{B}_{pq}^0 + L\left(\psi,\theta_{pq}^+\right)\mathbf{D}_{pq}^0\right] \tag{25}$$

with $\mathbf{B}_{pq}^0$ and $\mathbf{D}_{pq}^0$ being given by equation (17) where Q has been replaced by Q_0, *i.e.*

$$\mathbf{B}_{pq}^0 = i\beta_{pq}\begin{Vmatrix} 1 & \exp(-i\mathbf{r}_2\cdot\mathbf{t}_{pq}^-) \\ \exp(i\mathbf{r}_2\cdot\mathbf{t}_{pq}^-) & 1\end{Vmatrix},$$
$$\mathbf{D}_{pq}^0 = i\beta_{pq}\begin{Vmatrix} 1 & \exp(-i\mathbf{r}_2\cdot\mathbf{t}_{pq}^+) \\ \exp(i\mathbf{r}_2\cdot\mathbf{t}_{pq}^+) & 1\end{Vmatrix} \tag{26}$$

and $\mathbf{C}^0$ is given by equation (20) where Q has also been replaced by Q_0 and $S'(\mathbf{k})$ is replaced by $\mathrm{Re}\,S'(\mathbf{k}) + (2\pi i/|\mathbf{a}_1 \times \mathbf{a}_2|)\sum_{pq\ \mathrm{for}\ K_{pqz}\ \mathrm{real}} 1/K_{pqz}$, *i.e.*

$$\mathbf{C}^0 = \left(-Q_0 \mathrm{Re}\, S'(\mathbf{k}) + i \sum_{pq\ \mathrm{for}\ \beta_{pq}\ \mathrm{real}}^{(n)} \beta_{pq}\right)\mathbf{I}_2 + i\sum_{pq}^{(n)} \beta_{pq} \begin{Vmatrix} 0 & \exp\left(-i\mathbf{r}_2 \cdot \mathbf{t}_{pq}^-\right) \\ \exp\left(i\mathbf{r}_2 \cdot \mathbf{t}_{pq}^+\right) & 0 \end{Vmatrix}. \quad (27)$$

In equations (26) and (27) we have introduced

$$\beta_{pq} = -\frac{2\pi Q_0}{|\mathbf{a}_1 \times \mathbf{a}_2| K_{pqz}} = -\frac{1}{h_0 K_{pqz} a_{3z}} \quad (28)$$

with

$$h_0 = \frac{|\mathbf{a}_1 \times \mathbf{a}_2|}{2\pi a_{3z} Q_0} = \frac{1}{2\pi}\frac{a}{Q_0} \quad (29)$$

being the fundamental parameter of our theory; in silicon $h_0 = 0.21 \times 10^5$.

It is worth noting that in $\mathbf{A}^0(\psi)$ the bound scattering length Q_0 appears instead of Q since the imaginary part of Q^{-1}, being equal to k [see equation (19)], cancels exactly with the term $-k$ in the imaginary part of the intraplanar lattice sum [equation (21)] (Dub & Litzman, 2005).

To facilitate the solution of our problem we will use the matrix $\mathbf{A}^0(\psi)$ instead of $\mathbf{A}(\psi)$. Substitution of equation (24) into equation (12) gives an equation for the amplitudes $u_\mu^0(\psi_j) = (Q/Q_0)u_\mu(\psi_j)$:

$$\mathbf{A}^0(\psi_j) \begin{Vmatrix} u_1^0(\psi_j) \\ u_2^0(\psi_j) \end{Vmatrix} = 0 \quad (30)$$

where ψ_j are roots of the equation

$$\det \mathbf{A}^0(\psi) = 0. \quad (31)$$

The dispersion relation [equation (31)] has poles for $\theta_{pq}^+ + 2\pi m$ and $\theta_{pq}^- + 2\pi m$. As $\mathbf{B}_{pq}^0$ and $\mathbf{D}_{pq}^0$ are dyads there are $2n$ solutions of the dispersion relation, each solution ψ_j being associated with one pole θ_{pq}^+ and/or θ_{pq}^-. We denote them by ψ_{pq}^+ and ψ_{pq}^-, respectively. The distance $|\psi_{pq}^+ - \theta_{pq}^+|$ and/or $|\psi_{pq}^- - \theta_{pq}^-|$ is of order h_0^{-1}. Note that by introducing a small absorption, *i.e.* putting $Q_0 = Q_0 + i0^-$, $\mathrm{Im}\,\psi_{mn}^+ > 0$ holds. The important case arises when the pole $\theta_{rs}^+(\mathbf{k})$ and/or $\theta_{rs}^-(\mathbf{k})$ and the pole $\theta_{00}^+(\mathbf{k})$ almost coincide (modulo 2π), the former case representing the Bragg diffraction condition for the transmission geometry and the latter for the reflection geometry [see Dub & Litzman (2005)].

Next we evaluate the amplitudes c_j of the local field [equation (10)]. Introducing the notation

$$y_{kl}^\pm = \exp(i\theta_{kl}^\pm) \quad (32a)$$

for all n real θ_{kl}^+ and all n real θ_{kl}^-, and

$$x_{kl}^\pm = \exp(i\psi_{kl}^\pm) \quad (32b)$$

with ψ_{kl}^+ and ψ_{kl}^- being solutions of the dispersion relation [equation (31)], the square matrix $\mathbf{H}$ of order $2n$ in equation (13) reads

$$\mathbf{H} = \begin{Vmatrix} {}^{11}\mathbf{H} & {}^{12}\mathbf{H} \\ {}^{21}\mathbf{H} & {}^{22}\mathbf{H} \end{Vmatrix} =$$

$$\left\|\begin{array}{ccc|ccc} \frac{\alpha_{00}^+(\psi_{00}^+)}{x_{00}^+ - y_{00}^+} & \frac{\alpha_{00}^+(\psi_{pq}^+)}{x_{pq}^+ - y_{00}^+} & \cdots & \frac{\alpha_{00}^+(\psi_{uv}^+)}{x_{uv}^+ - y_{00}^+} & \frac{\alpha_{00}^+(\psi_{00}^-)}{x_{00}^- - y_{00}^+} & \frac{\alpha_{00}^+(\psi_{pq}^-)}{x_{pq}^- - y_{00}^+} & \cdots & \frac{\alpha_{00}^+(\psi_{uv}^-)}{x_{uv}^- - y_{00}^+} \\ \frac{\alpha_{pq}^+(\psi_{00}^+)}{x_{00}^+ - y_{pq}^+} & \frac{\alpha_{pq}^+(\psi_{pq}^+)}{x_{pq}^+ - y_{pq}^+} & \cdots & \frac{\alpha_{pq}^+(\psi_{uv}^+)}{x_{uv}^+ - y_{pq}^+} & \frac{\alpha_{pq}^+(\psi_{00}^-)}{x_{00}^- - y_{pq}^+} & \frac{\alpha_{pq}^+(\psi_{pq}^-)}{x_{pq}^- - y_{pq}^+} & \cdots & \frac{\alpha_{pq}^+(\psi_{uv}^-)}{x_{uv}^- - y_{pq}^+} \\ \vdots & \vdots & & \vdots & \vdots & \vdots & & \vdots \\ \frac{\alpha_{uv}^+(\psi_{00}^+)}{x_{00}^+ - y_{uv}^+} & \frac{\alpha_{uv}^+(\psi_{pq}^+)}{x_{pq}^+ - y_{uv}^+} & \cdots & \frac{\alpha_{uv}^+(\psi_{uv}^+)}{x_{uv}^+ - y_{uv}^+} & \frac{\alpha_{uv}^+(\psi_{00}^-)}{x_{00}^- - y_{uv}^+} & \frac{\alpha_{uv}^+(\psi_{pq}^-)}{x_{pq}^- - y_{uv}^+} & \cdots & \frac{\alpha_{uv}^+(\psi_{uv}^-)}{x_{uv}^- - y_{uv}^+} \\ \hline \frac{\alpha_{00}^-(\psi_{00}^+)(x_{00}^+)^{N+1}}{x_{00}^+ - y_{00}^-} & \frac{\alpha_{00}^-(\psi_{pq}^+)(x_{pq}^+)^{N+1}}{x_{pq}^+ - y_{00}^-} & \cdots & \frac{\alpha_{00}^-(\psi_{uv}^+)(x_{uv}^+)^{N+1}}{x_{uv}^+ - y_{00}^-} & \frac{\alpha_{00}^-(\psi_{00}^-)(x_{00}^-)^{N+1}}{x_{00}^- - y_{00}^-} & \frac{\alpha_{00}^-(\psi_{pq}^-)(x_{pq}^-)^{N+1}}{x_{pq}^- - y_{00}^-} & \cdots & \frac{\alpha_{00}^-(\psi_{uv}^-)(x_{uv}^-)^{N+1}}{x_{uv}^- - y_{00}^-} \\ \frac{\alpha_{pq}^-(\psi_{00}^+)(x_{00}^+)^{N+1}}{x_{00}^+ - y_{pq}^-} & \frac{\alpha_{pq}^-(\psi_{pq}^+)(x_{pq}^+)^{N+1}}{x_{pq}^+ - y_{pq}^-} & \cdots & \frac{\alpha_{pq}^-(\psi_{uv}^+)(x_{uv}^+)^{N+1}}{x_{uv}^+ - y_{pq}^-} & \frac{\alpha_{pq}^-(\psi_{00}^-)(x_{00}^-)^{N+1}}{x_{00}^- - y_{pq}^-} & \frac{\alpha_{pq}^-(\psi_{pq}^-)(x_{pq}^-)^{N+1}}{x_{pq}^- - y_{pq}^-} & \cdots & \frac{\alpha_{pq}^-(\psi_{uv}^-)(x_{uv}^-)^{N+1}}{x_{uv}^- - y_{pq}^-} \\ \vdots & \vdots & & \vdots & \vdots & \vdots & & \vdots \\ \frac{\alpha_{uv}^-(\psi_{00}^+)(x_{00}^+)^{N+1}}{x_{00}^+ - y_{uv}^-} & \frac{\alpha_{uv}^-(\psi_{pq}^+)(x_{pq}^+)^{N+1}}{x_{pq}^+ - y_{uv}^-} & \cdots & \frac{\alpha_{uv}^-(\psi_{uv}^+)(x_{uv}^+)^{N+1}}{x_{uv}^+ - y_{uv}^-} & \frac{\alpha_{uv}^-(\psi_{00}^-)(x_{00}^-)^{N+1}}{x_{00}^- - y_{uv}^-} & \frac{\alpha_{uv}^-(\psi_{pq}^-)(x_{pq}^-)^{N+1}}{x_{pq}^- - y_{uv}^-} & \cdots & \frac{\alpha_{uv}^-(\psi_{uv}^-)(x_{uv}^-)^{N+1}}{x_{uv}^- - y_{uv}^-} \end{array}\right\|. \quad (33)$$

Here [*cf.* (*L*34)]

$$\alpha^{+}_{mn}\big(\psi^{\pm}_{kl}\big) = Q_0\big[u^0_1\big(\psi^{\pm}_{kl}\big) + u^0_2\big(\psi^{\pm}_{kl}\big)\exp\big(-i\mathbf{r}_2\cdot\mathbf{t}^{+}_{mn}\big)\big] \qquad (34a)$$

$$\alpha^{-}_{mn}\big(\psi^{\pm}_{kl}\big) = Q_0\big[u^0_1\big(\psi^{\pm}_{kl}\big) + u^0_2\big(\psi^{\pm}_{kl}\big)\exp(-i\mathbf{r}_2\cdot\mathbf{t}^{-}_{mn})\big] \qquad (34b)$$

with $u^0_\mu(\psi^{\pm}_{kl})$ being given by equation (30) reflect the structure of a unit cell.

If we inject the plane-wave superposition [equation (10)] with the coefficients c_j, given by equation (13), and the amplitudes $u^0_\mu(\psi_j)$, given by equation (30), into equation (3*a*) we get the external wavefunctions $\Psi^{<}(\mathbf{r})$ and $\Psi^{>}(\mathbf{r})$ above ($z < 0$) and below ($z > Na_{3z}$) the crystalline slab, respectively. In particular, using (*L*29) and (*L*38) the wavefunction for $z < 0$ (in the Bragg geometry) reads

$$\Psi^{<}(\mathbf{r}) = \Psi_{\rm inc}(\mathbf{r}) + \sum_{pq}^{(n)}\Psi^{<}_{pq}(\mathbf{r}), \qquad (35)$$

where

$$\Psi^{<}_{pq}(\mathbf{r}) = -A\exp(-i\mathbf{k}\cdot\mathbf{a}_3)\frac{k_z}{K_{pqz}}\frac{\det\mathbf{M}^{-}_{pq}}{\det\mathbf{H}}\exp\big(i\theta^{-}_{pq}\big) \times\exp\big(i\mathbf{K}^{-}_{pq}\cdot\mathbf{r}\big) \qquad (36)$$

is the wavefunction of the (pq)-diffracted beam. The matrix $\mathbf{M}^{-}_{pq}$ of order $2n$ differs from the matrix $\mathbf{H}$ defined by equation (33) in the first row only. Its first row reads [*cf.* (*L*40)]

$$\Big\|\big(\mathbf{M}^{-}_{pq}\big)_{1,j}\Big\| = \begin{Vmatrix} \dfrac{\alpha^{-}_{pq}(\psi^{+}_{00})}{x^{+}_{00}-y^{-}_{pq}} & \dfrac{\alpha^{-}_{pq}(\psi^{+}_{pq})}{x^{+}_{pq}-y^{-}_{pq}} & \cdots & \dfrac{\alpha^{-}_{pq}(\psi^{+}_{uv})}{x^{+}_{uv}-y^{-}_{pq}} \\ \dfrac{\alpha^{-}_{pq}(\psi^{-}_{00})}{x^{-}_{00}-y^{-}_{pq}} & \dfrac{\alpha^{-}_{pq}(\psi^{-}_{pq})}{x^{-}_{pq}-y^{-}_{pq}} & \cdots & \dfrac{\alpha^{-}_{pq}(\psi^{-}_{uv})}{x^{-}_{uv}-y^{-}_{pq}} \end{Vmatrix}. \qquad (37)$$

Equation (35) is the exact multiple-beam solution of the Ewald equations (3*a*), (3*b*) for the Bragg case which is valid for any wavelength, any angle of incidence and for both coplanar and non-coplanar diffractions.

In the following we will consider reflection on a semi-infinite crystal ($N \to \infty$). Introducing a small absorption $(x^{+}_{mn})^{N+1} = \exp[i(N+1)\psi^{+}_{mn}] \to 0$ holds for all (mn) and thus both ${}^{21}\mathbf{H} \to 0$ and ${}^{21}\mathbf{M}^{-}_{pq} \to 0$, ${}^{21}\mathbf{H} = {}^{21}\mathbf{M}^{-}_{pq}$ are submatrices of $\mathbf{H}$ and $\mathbf{M}^{-}_{pq}$. Therefore

$$\frac{\det\mathbf{M}^{-}_{pq}}{\det\mathbf{H}} = \frac{\det{}^{11}\mathbf{M}^{-}_{pq}}{\det{}^{11}\mathbf{H}}\frac{\det{}^{22}\mathbf{M}^{-}_{pq}}{\det{}^{22}\mathbf{H}}.$$

As ${}^{22}\mathbf{M}^{-}_{pq} = {}^{22}\mathbf{H}$ we get finally

$$\frac{\det\mathbf{M}^{-}_{pq}}{\det\mathbf{H}} = \frac{\det{}^{11}\mathbf{M}^{-}_{pq}}{\det{}^{11}\mathbf{H}}. \qquad (38)$$

To evaluate the quotient [equation (38)] determining the wavefunction [equation (36)] is the crucial point of our development.

4. Two-beam case

In the following we will handle the case when one pole of the dispersion equation [equation (31)] only, say θ^{-}_{rs}, in the Bragg (reflection) geometry may approach θ^{+}_{00}, which yields the Bragg diffraction condition, *i.e.*

$$\theta^{+}_{00} - \theta^{-}_{rs} = 2\pi l + \eta_{rsl}, \qquad (39)$$

with l being an integer and $\eta_{rsl} \to 0$, the other poles being well separated from both $\theta^{+}_{00} \equiv \mathbf{a}_3\cdot\mathbf{k}$ and $\theta^{-}_{rs} \equiv \mathbf{a}_3\cdot\mathbf{K}^{-}_{rs}$ (*two-beam case*). Note that the corresponding diffraction vector is $\mathbf{G}_{\mathbf{h}} = r\mathbf{g}_1 + s\mathbf{g}_2 - l\mathbf{g}_3$ with $\mathbf{h} = (r, s, -l)$ (Dub & Litzman, 2005). It is a straightforward matter to evaluate the relation between our parameter η_{rsl} and the departure from Bragg's angle of the incident beam $\Delta\Theta = \Theta - \Theta_{\rm B}$ [see equation (83) in Dub & Litzman (2005)] for the coplanar case,

$$\eta_{rsl} = a_{3z}k^2\frac{1}{K_{rsz}}\sin(2\Theta_{\rm B})\Delta\Theta + O\big[(\Delta\Theta)^2\big] \qquad (40)$$

where the component of the reflected wave K_{rsz} is given by equation (6) and $\Theta_{\rm B}$ is the Bragg angle.

As each solution of equation (31) ψ^{+}_{pq} (ψ^{-}_{pq}) lies 'very' near the corresponding pole θ^{+}_{pq} (θ^{-}_{pq}), matrix elements on the main diagonals of ${}^{11}\mathbf{H}$ and ${}^{11}\mathbf{M}^{-}_{rs}$ are of order h_0, whereas those outside the main diagonal are of order 1. Furthermore, as matrices $\mathbf{H}$ and $\mathbf{M}^{-}_{rs}$ differ in their first rows only, the quotient [equation (38)] determining the reflectivity of a semi-infinite crystal becomes

$$\frac{\det\mathbf{M}^{-}_{rs}}{\det\mathbf{H}} = \frac{\det{}^{11}\mathbf{M}^{-}_{rs}}{\det{}^{11}\mathbf{H}} = \frac{\alpha^{-}_{rs}(\psi^{+}_{00})}{\alpha^{+}_{00}(\psi^{+}_{00})}\frac{\exp\big(i\psi^{+}_{00}\big) - \exp\big(i\theta^{+}_{00}\big)}{\exp\big(i\psi^{+}_{00}\big) - \exp(i\theta^{-}_{rs})} \times\big[1 + O\big(h_0^{-2}\big)\big] \qquad (41)$$

where ψ^{+}_{00} is to be found from the dispersion relation [equation (31)]. Within the two-beam case we rearrange the matrix $\mathbf{A}^0(\psi)$ defined by equation (25) as follows:

$$\mathbf{A}^0(\psi) = \mathbf{I}_2 + \mathbf{\Phi}_{00,rs}(\psi) - L(-\psi, -\theta^{-}_{rs})\mathbf{B}^0_{rs} - L\big(\psi, \theta^{+}_{00}\big)\mathbf{D}^0_{00}, \qquad (42)$$

where

$$\mathbf{\Phi}_{00,rs}(\psi) = -\mathbf{C}^0 - \sum_{(kl)\neq(rs)} L\big(-\psi, -\theta^{-}_{kl}\big)\mathbf{B}^0_{kl} - \sum_{(kl)\neq(00)} L\big(\psi, \theta^{+}_{kl}\big)\mathbf{D}^0_{kl} \qquad (43a)$$

comprises terms of order h_0^{-1} rendering multiple reflections in one crystal plane (the matrix $\mathbf{C}^0$) and the influence of all other beams than (rs) on ψ^{+}_{00}, and thus on the reflectivity in the direction $\mathbf{K}^{-}_{rs}$ [the sums on the right-hand side of equation (43*a*)]. When looking for ψ^{+}_{00} lying near the pole θ^{+}_{00} we may adopt the following approximation:

$$\mathbf{\Phi}_{00,rs}(\psi) \simeq \mathbf{\Phi}\big(\theta^{+}_{00}\big) \equiv \mathbf{\Phi} = \begin{Vmatrix} \varphi_{11} & \varphi_{12} \\ \varphi_{21} & \varphi_{22} \end{Vmatrix}, \qquad (43b)$$

where $\varphi_{ij} = O(h_0^{-1})$, $i, j = 1, 2$. The matrix elements φ_{ij} are evaluated for the symmetric reflection in §5 [see equation (73)].

Adopting the approximation [equation (43*b*)] and taking into account that matrices $\mathbf{D}^0_{00}$ and $\mathbf{B}^0_{rs}$ are dyads, the dispersion relation [equation (31)] in the two-beam case {where the Bragg diffraction condition [equation (39)] holds} yields the second-order equation in $\exp(i\psi)$,

$$\det(\mathbf{I}_2+\mathbf{\Phi}) + L\big(\psi,\theta^+_{00}\big)L(-\psi,-\theta^-_{rs})d - L\big(\psi,\theta^+_{00}\big)b - L(-\psi,-\theta^-_{rs})c = 0, \tag{44}$$

where

$$b = 2i\beta_{00} + i\beta_{00}\big[\varphi_{11}+\varphi_{22}-\varphi_{12}\exp\big(i\mathbf{r}_2\cdot\mathbf{t}^+_{00}\big) - \varphi_{21}\exp\big(-i\mathbf{r}_2\cdot\mathbf{t}^+_{00}\big)\big], \tag{45a}$$

$$c = 2i\beta_{rs} + i\beta_{rs}\big[\varphi_{11}+\varphi_{22}-\varphi_{12}\exp(i\mathbf{r}_2\cdot\mathbf{t}^-_{00}) - \varphi_{21}\exp(-i\mathbf{r}_2\cdot\mathbf{t}^-_{00})\big], \tag{45b}$$

$$d = \begin{vmatrix} D^0_{00,11} & D^0_{00,12} \\ B^0_{rs,21} & B^0_{rs,22}\end{vmatrix} + \begin{vmatrix} B^0_{rs,11} & B^0_{rs,12} \\ D^0_{00,21} & D^0_{00,22}\end{vmatrix} = -4\beta_{00}\beta_{rs}\sin^2\frac{\mathbf{r}_2\cdot\big(\mathbf{t}^+_{00}-\mathbf{t}^-_{rs}\big)}{2} = -4\beta_{00}\beta_{rs}\sin^2\frac{\mathbf{r}_2\cdot\mathbf{Q}}{2}. \tag{46}$$

Considering equation (18) and the identity in equation (46) $\mathbf{K}^+_{00}=\mathbf{k}$ we have introduced the scattering vector $\mathbf{Q} = -(\mathbf{t}^+_{00}-\mathbf{t}^-_{rs}) = \mathbf{K}^-_{rs}-\mathbf{k}$. It is worth noting that the d parameter [equation (46)] which is governed by the term $\mathbf{r}_2\cdot\mathbf{Q}$ determined by the structure of a unit cell and the scattering vector plays a crucial role in our development. Furthermore, note that the second terms on the right-hand sides of equations (45*a*) and (45*b*) being of order h_0^{-2} must be kept, as they are important when we evaluate the reflectivity of forbidden reflections.

Equation (44) yields the following expression for $\exp(i\psi^+_{00})$:

$$\exp\big(i\psi^+_{00}\big) = \exp\big(i\theta^+_{00}\big)\left\{1-\frac{\exp\big[-i(\eta_{rsl}/2)\big](bc)^{1/2}\tilde{Y}^-_{rsl}-(b+d)}{\det(\mathbf{I}_2+\mathbf{\Phi})+c}\right\}, \tag{47}$$

where we have introduced

$$\tilde{Y}^-_{rsl} = Y_{rsl} - \operatorname{sign}\big[\operatorname{Re}(Y_{rsl})\big](Y^2_{rsl}-\delta)^{1/2} \tag{48}$$

with

$$Y_{rsl} = \frac{1}{2(bc)^{1/2}}\left\{(d+b+c)\cos\Big(\frac{\eta_{rsl}}{2}\Big) + 2i\left[\det(\mathbf{I}_2+\mathbf{\Phi})+\frac{d+b+c}{2}\right]\sin\Big(\frac{\eta_{rsl}}{2}\Big)\right\} \tag{49}$$

and

$$\delta = 1-\frac{d}{bc}\det(\mathbf{I}_2+\mathbf{\Phi}), \tag{50}$$

which, as we will show in the next section, approaches one or zero for allowed and forbidden reflections, respectively. Note that the quantities [equations (48) to (50)] depend on the parameter η_{rsl}, equation (39) expressing the departure from Bragg's diffraction position.

Next, considering equations (36), (41) and (47) we find that the wavefunction $\Psi^<_{rs}(\mathbf{r})$ of the diffracted beam in the direction $\mathbf{K}^-_{rs}$ (the Bragg geometry) on a semi-infinite crystal

$$\Psi^<_{rs}(\mathbf{r}) = -A\frac{k_z}{K_{rsz}}\frac{\alpha^-_{rs}\big(\psi^+_{00}\big)}{\alpha^+_{00}\big(\psi^+_{00}\big)}\frac{L\big(\psi^+_{00},\theta^-_{rs}\big)}{L\big(\psi^+_{00},\theta^+_{00}\big)}\big[1+O\big(h_0^{-2}\big)\big] \times \exp(i\mathbf{K}^-_{rs}\cdot\mathbf{r}) \tag{51}$$

is determined by the product of the L quotient

$$\frac{L\big(\psi^+_{00},\theta^-_{rs}\big)}{L\big(\psi^+_{00},\theta^+_{00}\big)} = \frac{\exp\big(i\psi^+_{00}\big)-\exp\big(i\theta^+_{00}\big)}{\exp\big(i\psi^+_{00}\big)-\exp\big(i\theta^-_{rs}\big)}\exp\big[i\big(\theta^-_{rs}-\theta^+_{00}\big)\big] = \exp\Big(-i\frac{\eta_{rsl}}{2}\Big)\frac{-d-2i\sin(\eta_{rsl}/2)(bc)^{1/2}\tilde{Y}^-_{rsl}}{-2ic\sin(\eta_{rsl}/2)-d\exp\big[i(\eta_{rsl}/2)\big]} \tag{52}$$

and the α quotient [see equations (34*a*), (34*b*)]

$$\frac{\alpha^-_{rs}\big(\psi^+_{00}\big)}{\alpha^+_{00}\big(\psi^+_{00}\big)} = \frac{1+\big[u^0_2\big(\psi^+_{00}\big)/u^0_1\big(\psi^+_{00}\big)\big]\exp\big(-i\mathbf{r}_2\cdot\mathbf{t}^+_{00}\big)\exp\big[i\mathbf{r}_2\cdot\big(\mathbf{t}^+_{00}-\mathbf{t}^-_{rs}\big)\big]}{1+\big[u^0_2\big(\psi^+_{00}\big)/u^0_1\big(\psi^+_{00}\big)\big]\exp\big(-i\mathbf{r}_2\cdot\mathbf{t}^+_{00}\big)}. \tag{53}$$

Apparently, equation (53) is governed by the term $\mathbf{r}_2\cdot(\mathbf{t}^+_{00}-\mathbf{t}^-_{rs}) = -\mathbf{r}_2\cdot\mathbf{Q}$ and, as we will show in the next section, approaches one or zero for allowed and forbidden reflections, respectively. To evaluate equation (53) we use equation (30) yielding $u^0_2(\psi^+_{00})/u^0_1(\psi^+_{00}) = -A^0_{21}(\psi^+_{00})/A^0_{22}(\psi^+_{00})$. When replacing the matrix elements $A^0_{21}\big(\psi^+_{00}\big)$ and $A^0_{22}(\psi^+_{00})$ by equation (42) with the approximation [equation (43*b*)] we obtain that the ratio of the amplitudes of the local fields at basis atom positions is given by

$$\begin{aligned}\frac{u^0_2\big(\psi^+_{00}\big)}{u^0_1\big(\psi^+_{00}\big)} = &-\Big(-i\beta_{00}\exp\big(i\mathbf{r}_2\cdot\mathbf{t}^+_{00}\big)+\big[\varphi_{21}+i\beta_{rs}\exp(i\mathbf{r}_2\cdot\mathbf{t}^-_{rs})\big]/L\big(\psi^+_{00},\theta^+_{00}\big) \\ &+i\big[L\big(\psi^+_{00},\theta^-_{rs}\big)/L\big(\psi^+_{00},\theta^+_{00}\big)\big]\beta_{rs}\exp(i\mathbf{r}_2\cdot\mathbf{t}^-_{rs})\Big) \\ &\times\Big\{-i\beta_{00}+(1+\varphi_{22}+i\beta_{rs})/L\big(\psi^+_{00},\theta^+_{00}\big) \\ &+i\big[L\big(\psi^+_{00},\theta^-_{rs}\big)/L\big(\psi^+_{00},\theta^+_{00}\big)\big]\beta_{rs}\Big\}^{-1}.\end{aligned} \tag{54}$$

Equation (51) is our essential result which will be used for both allowed and forbidden reflections.

5. Allowed and forbidden reflections

The intensity of the diffracted beam is affected by the positions of atoms in the unit cell. In the Laue theory the influence of this arrangement on the diffraction intensity is given by the structure factor [equation (8)], which is the three-dimensional Fourier transform of the periodic potential of the three-dimensional infinite crystal lattice (see *e.g.* Authier, 2001).

Considering the primitive unit cell [equation (14)] the three-dimensional reciprocal-lattice basis vectors are

$$\mathbf{g}_1 = 2\pi\left(\frac{1}{a}, -\frac{1}{a}, -\frac{1}{a}\right),\ \mathbf{g}_2 = 2\pi\left(0, \frac{2}{a}, 0\right),\ \mathbf{g}_3 = 2\pi\left(0, 0, \frac{2}{a}\right), \tag{55}$$

and the geometric structure factor of the basis formed by two silicon atoms at the positions [equation (15)] for the reflection with the diffraction vector $\mathbf{G_h} = r\mathbf{g}_1 + s\mathbf{g}_2 - l\mathbf{g}_3$ becomes

$$\begin{aligned}\hat{F}(\mathbf{G_h}) = F(\mathbf{G_h})/Q &= \exp(-i\mathbf{G_h}\cdot\mathbf{r}_1) + \exp(-i\mathbf{G_h}\cdot\mathbf{r}_2)\\ &= 1 + \exp\left[-i\pi\left(-\frac{r}{2} + s - l\right)\right]\end{aligned}$$

so that[4]

$$\hat{F}(\mathbf{G_h}) = 0 \text{ for } s - 1 - r/2 \text{ being odd} \tag{56a}$$

$$\hat{F}(\mathbf{G_h}) = 2 \text{ for } s - 1 - r/2 \text{ being even.} \tag{56b}$$

Reflections with $\hat{F}(\mathbf{G}_{rs-l}) = 0$ are called forbidden whereas the others are called allowed. We keep this terminology in our theory although the reflectivity of the forbidden reflection will be shown to acquire significant non-zero value in a very narrow interval.

On the other hand, in our theory, which takes into account from the very beginning the two-dimensional symmetry of the slab, the influence of the basis [equation (15)] on the diffraction intensity is rendered by the term $\exp(-i\mathbf{r}_2\cdot\mathbf{Q})$ determining the d parameter [equation (46)] and the α quotient [equation (53)].

In Appendix B we have shown that the scattering vector $\mathbf{Q} = \mathbf{K}_{rs}^{-} - \mathbf{k}$ is related to the diffraction vector,

$$\mathbf{Q} = \mathbf{G_h} - \frac{\eta_{rsl}}{2\pi}\mathbf{g}_3. \tag{57}$$

Then

$$\exp(-i\mathbf{r}_2\cdot\mathbf{Q}) = \left[\hat{F}(\mathbf{G_h}) - 1\right]\exp\left(i\frac{\eta_{rsl}}{2}\right) = \pm\exp\left(i\frac{\eta_{rsl}}{2}\right), \tag{58}$$

where + and − correspond to allowed $[\hat{F}(\mathbf{G_h}) = 2]$ and forbidden $[\hat{F}(\mathbf{G_h}) = 0]$ reflections, respectively. Using the above result the d parameter [equation (46)] reads

$$d = -4\beta_{00}\beta_{rs}\sin^2\frac{\eta_{rsl}}{4} \tag{59a}$$

for *allowed* reflections and

$$d = -4\beta_{00}\beta_{rs}\cos^2\frac{\eta_{rsl}}{4} \tag{59b}$$

for *forbidden* reflections, and the α quotient [equation (53)] becomes

$$\frac{\alpha_{rs}^{-}(\psi_{00}^{+})}{\alpha_{00}^{+}(\psi_{00}^{+})} = \frac{1 \pm \left[u_2^0(\psi_{00}^{+})/u_1^0(\psi_{00}^{+})\right]\exp(-i\mathbf{r}_2\cdot\mathbf{t}_{00}^{+})\exp\left[i(\eta_{rsl}/2)\right]}{1 + \left[u_2^0(\psi_{00}^{+})/u_1^0(\psi_{00}^{+})\right]\exp(-i\mathbf{r}_2\cdot\mathbf{t}_{00}^{+})} \tag{60}$$

with + and − corresponding to allowed and forbidden reflections, respectively. Consequently, in the former case equation (60) is equal to $1 + O(\eta_{rsl})$ and in the latter it is equal to $O(\eta_{rsl})$. Then it may be supposed that the wavefunction [equation (51)] for a forbidden reflection, being determined by the product of the α quotient and the L quotient, is of order $O(\eta_{rsl})$, but that is not the full truth. We find there is a very narrow interval on which the L quotient [equation (52)] becomes very large so that the reflectivity for a forbidden reflection approaches one in this interval [see equation (70)].

Next we will discuss the cases of the allowed and forbidden reflections separately. While the mathematics of the former are straightforward,[5] those of the latter are cumbersome to describe the subtle effects one needs to consider in terms of orders both h_0^{-1} and h_0^{-2}.

(*i*) *Allowed reflections*. Inserting the d parameter [equation (59*a*)] into equation (50) we get $\delta = 1 + O(\eta_{rsl})$. Then equation (48) takes the form

$$\tilde{Y}_{rsl}^{-} = Y_{rsl} - \mathrm{sign}(Y_{rsl})[Y_{rsl}^2 - 1 + O(h_0^{-1})]^{1/2} \tag{61}$$

with [see equation (49)]

$$\begin{aligned}Y_{rsl}(\eta_{rsl}) &= \frac{1}{2(\beta_{00}\beta_{rs})^{1/2}}\left[(\beta_{00} + \beta_{rs}) + \frac{\eta_{rsl}}{2}\right]\left[1 + O(h_0^{-1})\right]\\ &= \frac{1}{2}\left\{-\left[\left(\frac{K_{rsz}}{k_z}\right)^{1/2} + \left(\frac{k_z}{K_{rsz}}\right)^{1/2}\right]\right.\\ &\quad\left. + \frac{h_0}{2}a_{3z}(k_zK_{rsz})^{1/2}\eta_{rsl}\right\}\left[1 + O(h_0^{-1})\right].\end{aligned} \tag{62}$$

As the d parameter is of order h_0^{-4} for the allowed reflections the L quotient [equation (52)] is simplified to $\exp(-i\eta_{rsl}/2)(\beta_{00}/\beta_{rs})^{1/2}\,\tilde{Y}_{rsl}^{-}[1 + O(h_0^{-1})]$. Furthermore, as the α quotient [equation (60)] is equal to $1 + O(\eta_{rsl})$ for the allowed reflections the wavefunction [equation (51)] becomes

$$\begin{aligned}\Psi_{rs}^{<}(\mathbf{r}) &= A(-1)^l\exp(-i\mathbf{k}\cdot\mathbf{a}_3/2)\left(\frac{k_z}{K_{rsz}}\right)^{1/2}\\ &\quad\times\left[Y_{rsl} - \mathrm{sign}(Y_{rsl})(Y_{rsl}^2 - 1)^{1/2}\right]\left[1 + O(h_0^{-1})\right]\\ &\quad\times\exp[i\mathbf{K}_{rs}^{-}\cdot(\mathbf{r} + \mathbf{a}_3/2)].\end{aligned} \tag{63}$$

The last equation has the same form as equation (11) of our former paper (Dub & Litzman, 2001*b*) deduced for the crystal with one atomic basis. Comparing equation (10) in Dub & Litzman (2001*b*) with equation (62) we can see that the influence of the basis is rendered by the term $h_0/2$ in the Y parameter [equation (62)] instead of h_0 describing a crystal with one atomic basis. Finally, note that phase factors $\exp(-i\mathbf{k}\cdot\mathbf{a}_3/2)$ and $\exp(i\mathbf{K}_{rs}^{-}\cdot\mathbf{a}_3/2)$ express the shift of the entrance crystal surface above the uppermost atomic layer $m_3 = 0$ by $\mathbf{a}_3/2$ as mentioned already by Dub & Litzman (2001*b*).

[4] In both cases r is considered to be even. The case with r odd yielding allowed reflections with a complex structure factor will not be discussed here.

[5] Here we do not consider extreme cases outside the scope of the standard dynamical theory. Handling special situations, such as *e.g.* the (allowed) diffraction at the Bragg angle near $\pi/2$, is cumbersome too as terms of orders both h_0^{-1} and h_0^{-2} have to be considered (Litzman & Dub, 1990; Litzman *et al.*, 1996).

Having found the wavefunction, we evaluate the reflectivity of allowed diffractions,

$$I^{<}_{rsl}(\eta_{rsl}) = \frac{K_{rsz}}{k_z}\frac{\left|\Psi^{<}_{rs}\right|^2}{\left|\Psi_{\rm inc}\right|^2} = \left|Y_{rsl}(\eta_{rsl}) - {\rm sign}(Y_{rsl})[Y^2_{rsl}(\eta_{rsl}) - 1]^{1/2}\right|^2 \times \left[1 + O\left(h_0^{-1}\right)\right] \quad (64)$$

with $\mathbf{h} = (r, s, -l)$ specifying the diffraction vector. The condition for the total reflection $|Y_{rsl}| \leq 1$ yields the Darwin table of the width [*cf.* equation (3.7) in Litzman *et al.* (1996)]

$$D^{\rm allowed} = 8(\beta_{00}\beta_{rs})^{1/2} \quad (65)$$

being of order h_0^{-1}, and the deviation from Bragg's angle of the middle of the reflection domain

$$\eta^{c}_{rsl} = -2(\beta_{00} + \beta_{rs}). \quad (66)$$

Finally, note that our formula [equation (64)] corresponds to equation (4.42) in Authier (2001), derived in the frame of the Laue diffraction theory, when replacing Y_{rsl} by Authier's deviation parameter η, which is also equal to the y parameter defined by equation (9.23) in Rauch & Petraschek (1978) [for details see Dub & Litzman (2005)]. However, our result is valid for both coplanar and non-coplanar diffractions.

(*ii*) *Forbidden reflections*. Inserting the d parameter [equation (59*b*)] into equation (50) after some algebraic manipulations, we get that δ is now of order h_0^{-1},

$$\delta = -(\varphi_{12} + \varphi_{21})\cos\left(\mathbf{r}_2 \cdot \mathbf{t}^{+}_{00}\right) + \frac{1}{4}(\varphi_{11} - \varphi_{22})^2 + \frac{1}{2}(\varphi_{11} + \varphi_{22}) \times (\varphi_{12} + \varphi_{21})\cos\left(\mathbf{r}_2 \cdot \mathbf{t}^{+}_{00}\right) - \left(\varphi^2_{12} + \varphi^2_{21} + \varphi_{12}\varphi_{21}\right) \times \cos^2\left(\mathbf{r}_2 \cdot \mathbf{t}^{+}_{00}\right) + \frac{1}{4}(\varphi_{12} + \varphi_{21})^2 + \left(\frac{\eta_{rsl}}{4}\right)^2. \quad (67)$$

Then $\tilde{Y}^{-}_{rsl} = Y_{rsl} - {\rm sign}(Y^2_{rsl} - \delta)^{1/2}$, with Y_{rsl} given by equation (49), is of order h_0^{-1}. Using this result in equations (52) and (54) we evaluate the α quotient and finally, after some lengthy algebraic manipulations, we get the wavefunction expressed by equation (51),

$$\Psi^{<}_{rs}(\mathbf{r}) = -A(-1)^l \exp\left(-i\mathbf{k}\cdot\mathbf{a}_3/2\right) \times \frac{k_z}{K_{rsz}}\frac{\beta_{00}}{\eta_{rsl} + 2(\beta_{00} + \beta_{rs}) + W^{(2)}_{rsl} + O\left(h_0^{-3}\right)} \times \left[W^{(1)}_{rsl} + O\left(h_0^{-2}\right)\right]\exp\left[i\mathbf{K}^{-}_{rs}\cdot\left(\mathbf{r} + \mathbf{a}_3/2\right)\right] \quad (68)$$

where

$$W^{(1)}_{rsl} = -i\frac{\eta_{rsl}}{2} + 2\left(\frac{\beta_{rs}}{\beta_{00}}\right)^{1/2}\tilde{Y}^{-}_{rsl} + \left[\varphi_{21}\exp\left(-i\mathbf{r}_2\cdot\mathbf{t}^{+}_{00}\right) - \varphi_{12}\exp\left(i\mathbf{r}_2\cdot\mathbf{t}^{+}_{00}\right)\right] \quad (69a)$$

is of order h_0^{-1}, and

$$W^{(2)}_{rsl} = 2\beta_{00}\left(\frac{\beta_{rs}}{\beta_{00}}\right)^{1/2}\tilde{Y}^{-}_{rsl} - \left(\frac{\eta_{rsl}}{2} + \beta_{00}\right)[3\varphi_{21}\exp\left(-i\mathbf{r}_2\cdot\mathbf{t}^{+}_{00}\right) + \varphi_{12}\exp\left(i\mathbf{r}_2\cdot\mathbf{t}^{+}_{00}\right)] + \frac{\eta_{rsl}}{2}\left\{\varphi_{11} + \varphi_{22} - \left[\varphi_{12}\exp\left(i\mathbf{r}_2\cdot\mathbf{t}^{-}_{00}\right) + \varphi_{21}\exp\left(-i\mathbf{r}_2\cdot\mathbf{t}^{-}_{00}\right)\right]\right\} + i\left(4\beta_{00}\beta_{rs} + \eta_{rsl}\beta_{00} + \frac{\eta_{rsl}}{2}\beta_{rs}\right) \quad (69b)$$

is of order h_0^{-2}.

Finally, the reflectivity of the ideal Si(001) crystal, which is valid for both symmetric and non-symmetric and both coplanar and non-coplanar forbidden diffractions, reads

$$I^{<}_{rsl}(\eta_{rsl}) = \frac{K_{rsz}}{k_z}\frac{\left|\Psi^{<}_{rs}\right|^2}{\left|\Psi_{\rm inc}\right|^2} = \frac{k_z}{K_{rsz}} \times \frac{\beta^2_{00}\left|W^{(1)}_{rsl}\left(h_0^{-1}\right)\right|^2}{\left\{\eta_{rsl} + 2(\beta_{00} + \beta_{rs}) + {\rm Re}\left[W^{(2)}_{rsl}\left(h_0^{-2}\right)\right]\right\}^2 + \left\{{\rm Im}\left[W^{(2)}_{rsl}\left(h_0^{-2}\right)\right]\right\}^2}. \quad (70)$$

Analysing equation (70) we see that for $\eta_{rsl} = -2(\beta_{00} + \beta_{rs}) + O(h_0^{-2})$ the denominator is of order h_0^{-4}, and thus the reflectivity $I^{<}_{rsl}(\eta_{rsl})$ is of order one here, while elsewhere it is of order h_0^{-2}. Consequently, there is a very narrow peak in the reflectivity [equation (70)] in the vicinity of $\eta^{c}_{rsl} = -2(\beta_{00} + \beta_{rs})$ [see also equation (66) derived for the allowed reflections]. Note that the reflectivity peak width of forbidden reflections is of order h_0^{-2}, whereas the DTW of allowed reflections is of order h_0^{-1} [see equation (65)]. Thus the ratio of the widths of the former and the latter cases is of the order $Q_0/a \simeq 10^{-5}$ which agrees with the result by Ignatovich *et al.* (1996) obtained for symmetric reflections.

5.1. Forbidden symmetric reflection of the Si(001) crystal in the Bragg geometry

As an example we will concern ourselves with the forbidden (l is odd) symmetric $[(r, s) = (0, 0)]$ reflection. Here the deviation from Bragg's angle of the middle of the reflection domain $\eta^{c}_{001} = -4\beta_{00}$, the Bragg diffraction condition [equation (39)] reads

$$\theta^{+}_{00} - \theta^{-}_{00} = ak\cos\gamma_{\rm B} = 2\pi l \quad (71)$$

and

$$\mathbf{r}_2\cdot\mathbf{t}^{\pm}_{00} = \pm(ak\cos\gamma_{\rm B})/4 = \pm\frac{\pi}{2}l \quad (72)$$

with $\gamma_{\rm B} = \pi/2 - \Theta_{\rm B}$ being the angle of incidence (measured from the inner normal to the surface) at the Bragg reflection. In the following we consider that l = 1. Then equation (28) yields $\beta_{00} = -2(Q_0/a)$ and the Bragg angle equals $\Theta_{\rm B} = \arcsin(\lambda/a)$. Furthermore, averaging over oscillations in equation (43*a*), matrix elements in equation (43*b*) read

$$\varphi_{11} = \varphi_{22} = -i\beta_{00} + \beta_{00}\sigma, \varphi_{12} = \varphi_{21} = \beta_{00}, \quad (73)$$

where the term

$$\sigma = Q_0{\rm Re}\,S'(\mathbf{k})/\beta_{00}, \quad (74)$$

being of order one, expresses local-field corrections.

To evaluate the reflectivity $I_{001}^{<}$ in the vicinity of $\eta_{001}^{c} = -4\beta_{00}$ we express the deviation parameter η_{001} as

$$\eta_{001} = \eta_{001}^{c} + 4\beta_{00}^{2}\xi \tag{75}$$

with ξ being of order one. Considering equation (75), using equation (72) and adopting the approximations [equation (73)], the δ parameter [equation (67)] reduces to $\delta = 2\beta_{00}^{2} + O(h_0^{-3})$, equation (49) yields $Y_{001} = -\beta_{00}(\xi - \sigma) + O(h_0^{-2})$, and equations (69*a*) and (69*b*) give $W_{001}^{(1)} = 2\tilde{Y}_{001}^{-} + O(h_0^{-2})$ and $W_{001}^{(2)} = 2\beta_{00}\tilde{Y}_{001}^{-} - 4\beta_{00}^{2}\sigma + O(h_0^{-3})$, respectively, with [see equation (48)]

$$\begin{aligned}\tilde{Y}_{001}^{-} &= Y_{001} - \mathrm{sign}(Y_{001})(Y_{001}^{2} - \delta)^{1/2} \\ &= \beta_{00}\left\{-(\xi - \sigma) + \mathrm{sign}(\xi - \sigma)\left[(\xi - \sigma)^{2} - 2\right]^{1/2}\right\} + O\left(h_0^{-2}\right).\end{aligned} \tag{76}$$

Finally, inserting the above formulae into equation (68) we get the reflectivity[6]

$$\begin{aligned}I_{001}^{<} &= \frac{\left|\Psi_{00}^{<}\right|^{2}}{\left|\Psi_{\mathrm{inc}}\right|^{2}} = \left|\frac{\tilde{Y}_{001}^{-}}{2\beta_{00}(\xi - \sigma) + \tilde{Y}_{001}^{-}}\right|^{2} \\ &= \left|\frac{-(\xi - \sigma) + \mathrm{sign}(\xi - \sigma)[(\xi - \sigma)^{2} - 2]^{1/2}}{(\xi - \sigma) + \mathrm{sign}(\xi - \sigma)[(\xi - \sigma)^{2} - 2]^{1/2}}\right|^{2}.\end{aligned} \tag{77}$$

From here we can see that if $-2^{1/2} < \xi - \sigma < 2^{1/2}$ holds, the reflectivity $I_{001}^{<}$ equals unity whereas outside this interval it goes rapidly to zero. Hence we may conclude that there exists the Darwin table of the width

$$D_{001}^{\mathrm{forbidden}} = 8(2)^{1/2}\beta_{00}^{2} = 32(2)^{1/2}\left(\frac{Q_0}{a}\right)^{2} \tag{78}$$

centred at $\eta_{001}^{(c)} + 4\beta_{00}^{2}\sigma = 8(Q_0/a)[1 + 2(Q_0/a)\sigma]$ in the forbidden symmetric reflection (see Fig. 1). Note that σ defined by equation (74) causes a tiny shift of the table centre while it does not affect the table width.

Finally, we put down the angular width of the Darwin table $|\Delta\Theta|_{\mathrm{FDT}}$ of the forbidden reflection, and the angular deviation from Bragg's angle of the middle of the reflection domain, $\Delta\Theta_c = \Theta_c - \Theta_{\mathrm{B}}$. Considering equations (40) and (71) with $l = 1$, we get for the symmetric Bragg reflection (001) that $|\Delta\Theta|_{\mathrm{FDT}} = (D_{001}^{\mathrm{forbidden}}/2\pi)\tan\Theta_{\mathrm{B}}$ and $\Delta\Theta_c = (\eta_{001}^{c}/2\pi)\tan\Theta_{\mathrm{B}}$. Inserting here for $D_{001}^{\mathrm{forbidden}}$ from equation (78) and $\eta_{001}^{c} = -4\beta_{00} = 8(Q_0/a)$, respectively, we obtain

$$|\Delta\Theta|_{\mathrm{FDT}} = \frac{16(2)^{1/2}}{\pi}\left(\frac{Q_0}{a}\right)^{2}\tan\Theta_{\mathrm{B}} \tag{79}$$

and

$$\Delta\Theta_c = \frac{4}{\pi}\left(\frac{Q_0}{a}\right)\tan\Theta_{\mathrm{B}}, \tag{80}$$

where $\Theta_{\mathrm{B}} = \arcsin(\lambda/a)$ is yielded by equation (71). Considering diffraction of thermal neutrons ($\lambda = 0.1$ nm) on a single Si crystal ($Q_0 = 4.1$ fm, $a = 0.5430$ nm) we get $|\Delta\Theta|_{\mathrm{FDT}} = 1.6 \times 10^{-5}$ arcsec and $\Delta\Theta_c = 0.4$ arcsec.

[6] When using the conventional unit cube notation this case represents the (002) diffraction.

6. Summary and concluding remarks

In this paper we have treated the diffraction of neutrons on an ideal semi-infinite single silicon crystal Si(001) as a multiple scattering problem based upon the Ewald equations (3*a*), (3*b*). As we have considered from the very beginning the bordered crystal, we have naturally overcome the boundary conditions problem which, as pointed out by von Laue (1941), is 'the weakest point of the dynamical diffraction theory'. The question of where to locate the boundary taken as a mathematical plane is of fundamental importance particularly in crystals with a distributed cell content (Juretschke, 1992). We have found {see wavefunctions [equations (63) and (68)] for allowed and forbidden reflections, respectively} that a mathematical boundary plane lies above the uppermost atomic layer, the shift vector being equal to $\mathbf{a}_3/2$. Thus we have confirmed the result which we already derived for the crystal with the cell containing one atom only (Dub & Litzman, 2001*b*). Furthermore, when considering from the very beginning the two-dimensional translation symmetry of the problem, we have found that no structure factor being the Fourier transform of a three-dimensional crystal appears. Instead, in our development the influence of the two atomic silicon basis on diffraction intensity in the direction $\mathbf{K}_{rs}^{-}$ is rendered by the term $\exp(-i\mathbf{r}_2 \cdot \mathbf{Q})$ with $\mathbf{Q} = \mathbf{K}_{rs}^{-} - \mathbf{k}$ being

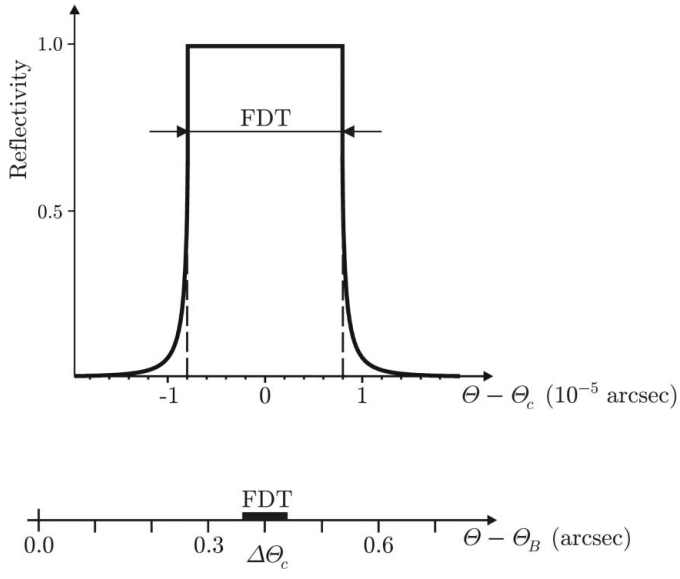

Figure 1
Reflectivity profile of the forbidden symmetric (001) reflection on the Si(001) thick crystal. The deviation from Bragg's angle of the middle of the reflection domain is given by equation (80), $\Delta\Theta_c = \Theta_c - \Theta_{\mathrm{B}} = (4/\pi)(Q_0/a)\tan\Theta_{\mathrm{B}} = 0.4$ arcsec, where $\Theta_{\mathrm{B}} = \arcsin(\lambda/a) = 10.6^{\circ}$. The angular width of the forbidden Darwin table (FDT) is given by equation (79), $|\Delta\Theta|_{\mathrm{FDT}} = [16(2)^{1/2}/\pi](Q_0/a)^{2}\tan\Theta_{\mathrm{B}} = 1.6 \times 10^{-5}$ arcsec. Numerical values characterize diffraction of thermal neutrons ($\lambda = 0.1$ nm) on a single Si crystal ($Q_0 = 4.1$ fm, $a = 0.5430$ nm). Using the conventional unit cube notation this case corresponds to the (002) reflection. (Note, if the reflection was allowed the Darwin table width would broaden, becoming $2\Delta\Theta_c$.)

the scattering vector. The term $\exp(-i\mathbf{r}_2 \cdot \mathbf{Q})$ acquires opposite signs for allowed and forbidden reflections [see equation (58)] and thus affects substantially the value of the d parameter [equations (59*a*), (59*b*)] determining the solution of the dispersion relation [see equations (47) to (50)], and also the L quotient [equation (52)] and α quotient [equation (60)] controlling the wavefunction of the reflected beam [equation (51)]. Moreover, the scattering vector is related to the diffraction one by equation (57), $\mathbf{Q} = \mathbf{G_h} - (\eta_{rsl}/2\pi)\mathbf{g}_3$. While in the case of allowed reflections the scattering vector $\mathbf{Q}$ may be put equal to the diffraction vector $\mathbf{G_h}$, the small correction given by the parameter η_{rsl} [equation (39)] expressing the departure from Bragg's diffraction position must be considered and provides the narrow Darwin table to forbidden reflections. Finally, note that, applying the matrix formulation of the Ewald theory to a silicon crystal, instead of the conventional unit cube of an f.c.c. lattice containing four lattice points, we have used the primitive unit cell containing two Si atoms.

In summary, the main results of our paper are the dispersion equation (44) and formula (51) for the wavefunction $\Psi^{<}_{rs}(\mathbf{r})$ of the reflected beam in the direction $\mathbf{K}^{-}_{rs}$ in the Bragg geometry. By using them, we have analysed the cases of allowed and forbidden reflections. Whereas in the former case our reflectivity formula [equation (64)] corresponds to that derived in the frame of the Laue diffraction theory, in the latter one we have found that unlike the Laue theory predicting zero intensity there exists a very narrow peak in reflectivity [equation (70)], which is the result of the broken symmetry due to the border of a crystal. The results obtained hold for both symmetric and non-symmetric reflections and for both coplanar and non-coplanar ones. It is worth noting that elements of the Φ matrix [equation (43*b*)] also enter equation (70), rendering the influence of beams other than (rs) on the reflectivity in the direction $\mathbf{K}^{-}_{rs}$, and local-field correction. In the case of forbidden reflections we have confirmed the result by Ignatovich *et al.* (1996) who found, using an artificial model of a crystal with two identical atoms in an elementary cell, that a forbidden symmetric reflection possesses a finite DTW which differs from that of a non-forbidden (allowed) one by order 10^{-5}. They utilized the approach in which the crystal is imagined to be cut in slices parallel to the entrance surface and the scattering on a slice is described by direct transmission and specular reflection. The formalisms used by Ignatovich *et al.* and by us differ substantially. Nevertheless, their interpretation of the effect corresponds to our finding that the slight difference between the scattering and diffraction vectors given by the departure from Bragg's diffraction position accounts for the finite DTW in forbidden reflections. Furthermore, their remark that the first plane screens the second one inside a single period since amplitudes of the waves reflected by the two planes are different is related to our equation (54), indicating that the amplitudes of the local fields at the positions of the first and second basis atom differ.[7]

Concluding, let us note that the method explained may be applied to any crystal but matrices $\mathbf{B}_{pq}$ and $\mathbf{D}_{pq}$ given by equation (17) reflecting the structure of a unit cell will change, the order of them being given by the number of atoms in a unit cell.

APPENDIX *A*
The intraplanar lattice sum [equation (21)]

Looking for the approximate value of the sum [equation (21)] we can write (Dub *et al.*, 1996)

$$S'(\mathbf{k}) = \frac{1}{a^2}\int_a^\infty \int_0^{2\pi} \exp(ikr)\exp\left(ik^{||} r\cos\varphi\right) \mathrm{d}r\,\mathrm{d}\varphi$$
$$= \frac{2\pi}{a^2}\int_a^\infty \exp(ikr) J_0\left(k^{||} r\right) \mathrm{d}r, \qquad (81)$$

where $J_0(x)$ is the Bessel function. As $\int_0^\infty \cos(kr) J_0(k^{||} r)\,\mathrm{d}r = 0$ for $k^{||} < k$, in the region where $ak \simeq 1$ we can put approximately $|\mathrm{Re}\, S'(\mathbf{k})| \simeq 2\pi/ak^2$.

APPENDIX *B*
Relation between the scattering and diffraction vectors

The vector $\mathbf{K}^{-}_{rs}$ defined by equation (5) may be expressed as $\mathbf{K}^{-}_{rs} = \mathbf{k} + y_1\mathbf{g}_1 + y_2\mathbf{g}_2 + y_3\mathbf{g}_3$. Since $\mathbf{g}_1^{||} = \mathbf{b}_1$, $\mathbf{g}_2^{||} = \mathbf{b}_2$, we get $y_1 = r$ and $y_2 = s$ and thus $\mathbf{K}^{-}_{rs} = \mathbf{k} + r\mathbf{g}_1 + s\mathbf{g}_2 + y_3\mathbf{g}_3$ [see also equation (84) in Dub & Litzman (2005)]. Multiplying the last equation by $\mathbf{a}_3$ we obtain $\theta^{-}_{rs} = \theta^{+}_{00} + 2\pi y_3$ [*cf.* definition (23)]. After using the Bragg diffraction condition [equation (39)] we get $y_3 = -l - \eta_{rsl}/2\pi$ and finally

$$\mathbf{Q} = \mathbf{K}^{-}_{rs} - \mathbf{k} = r\mathbf{g}_1 + s\mathbf{g}_2 - l\mathbf{g}_3 - \frac{\eta_{rsl}}{2\pi} = \mathbf{G_h} - \frac{\eta_{rsl}}{2\pi}, \qquad (82)$$

where $\mathbf{G_h} = r\mathbf{g}_1 + s\mathbf{g}_2 - l\mathbf{g}_3$ is the diffraction vector.

We thank Professor V. Holý for critical reading of the manuscript, Dr R. Kalousek for stimulating discussions and our referees for their comments which helped to improve our paper. The work has been supported by the research plan MSM0021630508 of the Ministry of Education, Youth and Sport of the Czech Republic, and the European Regional Development Fund (grant No. CEITEC-CZ.1.05/1.1.00/02.0068).

References

Authier, A. (2001). *Dynamical Theory of X-ray Diffraction.* Oxford University Press.

Cruickshack, D. W. J., Juretschke, H. J. & Kato, N. (1992). Editors. *P. P. Ewald and his Dynamical Theory of X-ray Diffraction.* Oxford University Press.

[7] Ignatovich *et al.* (1996) noticed that in Ewald theory the screening is not taken into account. Apparently, by Ewald theory they did not mean the theory of multiple scattering of waves based on the Ewald equations (3*a*), (3*b*) [see *e.g.* Ignatovich (1993), p. 590].

Dederichs, P. H. (1972). *Solid State Physics*, Vol. 27, edited by H. Ehrenreich, F. Seitz & D. Turnbull, pp. 135–236. New York: Academic Press.
Dub, P. & Litzman, O. (2001*a*). *Acta Cryst.* A**57**, 212–216.
Dub, P. & Litzman, O. (2001*b*). *Acta Cryst.* A**57**, 686–689.
Dub, P. & Litzman, O. (2005). *Acta Cryst.* A**61**, 209–222.
Dub, P., Litzman, O. & Mikulík, P. (1996). *Scr. Fac. Sci. Nat. Univ. Masaryk. Brun. (Physics)*, **24–26**, 5–38.
Ewald, P. P. (1916). *Ann. Phys. (Leipzig)*, **49**, 117–143.
Ewald, P. P. (1917). *Ann. Phys. (Leipzig)*, **54**, 519–597.
Ignatovich, V. K. (1993). *Sov. Phys. Crystallogr.* **35**, 588–595.
Ignatovich, V. K., Protopopescu, D. & Utsuro, M. (1996). *Phys. Rev. Lett.* **77**, 4202–4205.
Juretschke, H. J. (1992). *P. P. Ewald and his Dynamical Theory of X-ray Diffraction*, edited by D. W. J. Cruickshank, H. J. Juretschke & N. Kato, p. 96. Oxford University Press.
Laue, M. von (1941). *Röntgenstrahlinterferenzen.* Leipzig: Akademische Verlagsgesellschaft.
Litzman, O. (1978). *Opt. Acta*, **25**, 509–526.
Litzman, O. (1980). *Opt. Acta*, **27**, 231–240.
Litzman, O. (1986). *Acta Cryst.* A**42**, 552–559.
Litzman, O. & Dub, P. (1982). *Opt. Acta*, **29**, 1317–1330.
Litzman, O. & Dub, P. (1990). *Acta Cryst.* A**46**, 247–254.
Litzman, O., Mikulík, P. & Dub, P. (1996). *J. Phys. Condens. Matter*, **8**, 4709–4725.
Litzman, O. & Rózsa, P. (1977). *Surf. Sci.* **66**, 542–558.
Rauch, H. & Petraschek, D. (1978). *Dynamical Neutron Diffraction and its Applications. Neutron Diffraction*, edited by H. Dachs, *Topics in Current Physics*, Vol. 6, pp. 303–357. Berlin: Springer-Verlag.
Sears, V. F. (1989). *Neutron Optics.* Oxford University Press.