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Multiple diffraction of particles by a system of point scatterers as an exactly soluble problem using the Ewald concept

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Abstract. The reflection of a de Broglie plane wave incident on a system of point scatterers (nuclei) forming an ideal semi-infinite crystal is studied using the **T**-matrix formalism of the Ewald dynamical theory of diffraction. Using from the beginning the two-dimensional translational symmetry of the crystal bordered by a surface, simple exact many-beam analytical formulae for the intensities of the reflected waves are deduced, whereby the Ewald sphere is replaced by 'the gamma diagrams' and the usual three-dimensional dispersion surface by a two-dimensional 'dispersion plot'. The results obtained are valid for arbitrary angles of incidence (including the grazing incidence, Bragg angle near $\pi/2$, near or far from the Bragg peaks) and for any directions of the reflected waves (including both the coplanar and the non-coplanar reflections). The transparent algebraic form of the final formulae allows us to discuss analytically the solutions of the dispersion relation and the intensities of the reflections.

1. Introduction

The problem of the interaction of radiation (electromagnetic waves, electrons, neutrons and atoms) with a system of scatterers (nuclei, atoms, molecules and crystals) is encountered in many branches of physics. Owing to the large variety of parameters characterizing these processes (plane wave, spherical wave, wavelength and interaction potential), different (mostly approximate) methods are used. Considering the most traditional scattering of electromagnetic waves we should mention the methods of classical optics [1], kinematical [2], dynamical [3–10] and extended dynamical [11–13] theory of diffraction.

To deal with the problem of the diffraction from crystalline materials in the short-wavelength region, mostly the approach introduced by Bethe [6] and von Laue [7], and less frequently the Darwin [14] method combined with some thin-film-optics techniques [15–17] are used. Attention is seldom paid to the Ewald [3, 4] concept [5] which was developed for studying the scattering of radiation by a system of point scatterers, which is physically closest to the diffraction of light by point dipoles (Ewald) or of neutrons by nuclei. Qualified comparison of the Bethe–von Laue method and the Ewald method has been given by Sears [18, pp 176–7].

In our previous paper [19] we dealt with the diffraction of the scalar de Broglie waves in an ideal crystal using the formalism of the **T**-matrix [18, 20]. Putting the solution of

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the quantum-mechanical Ewald equations into a suitable matrix form we have succeeded in expressing the reflection and transmission coefficients in a well arranged determinant form, which is valid quite generally. Recently we applied the results obtained to the study of the diffraction of neutrons near the Bragg peaks [21]. In the present paper we shall extend the approach developed to the regions outside the Bragg peaks, where approximations which are made in the standard dynamical theory fail. In section 2 we present the main ideas of the quantum-mechanical version of the Ewald dynamical theory of diffraction, the formula for the reflectivity (2.15) deduced previously in [19], and new forms (2.18*b*) and (2.18*c*) of the dispersion equation (2.18*a*). These general results, which are valid for all wavelengths, are analysed in detail in the short-wavelength region. The analysis of the dispersion relation and its connection with the reflectivity in section 3 differs substantially from those given in standard textbooks on the dynamical theory of diffraction [8–10]. No three-dimensional dispersion surface or approximate dispersion hyperbolae for the coplanar diffraction in the two-beam approximation are introduced. Our dispersion relation (2.18*b*) is represented for both coplanar and non-coplanar diffractions by a two-dimensional 'dispersion plot' with poles (figure 2), the position of these poles being given by the geometry and the wavelength only. The confluence of two poles occurs if the Bragg condition is satisfied. A comparison of our method with the Bethe–von Laue and the Darwin theory and some generalizations are given in section 4.

2. The Ewald equations and their solution

Let us consider the diffraction of a particle (neutron) in a system of point scatterers (nuclei). The Ewald dynamical (self-consistent field) theory of diffraction generalized to quantum mechanics leads to the following algebraic system of equations for the evaluation of the wavefunction $\Psi(\boldsymbol{r})$ [18, 20]:

$$\phi^m(\boldsymbol{R}_m) = f(\boldsymbol{R}_m) - {\sum_{n\neq m}}' Q_n \frac{\exp(\mathrm{i}k|\boldsymbol{R}_m - \boldsymbol{R}_n|)}{|\boldsymbol{R}_m - \boldsymbol{R}_n|}\phi^n(\boldsymbol{R}_n) \tag{2.1a}$$

$$\Psi(\boldsymbol{r}) = f(\boldsymbol{r}) - \sum_n Q_n \frac{\exp(\mathrm{i}k|\boldsymbol{r} - \boldsymbol{R}_n|)}{|\boldsymbol{r} - \boldsymbol{R}_n|}\phi^n(\boldsymbol{R}_n). \tag{2.1b}$$

$\Psi(\boldsymbol{r})$ is the total field at the point $\boldsymbol{r}$, $\phi^m(\boldsymbol{R}_m)$ is the 'effective field' incident on the scatterer located at $\boldsymbol{R}_m$ and Q_m the scattering length of this scatterer. The function $f(\boldsymbol{r})$ represents the primary beam of the incident particles. In our case we shall consider the scalar plane wave

$$f(\boldsymbol{r}) = A\exp(\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}). \tag{2.2}$$

Thus in the first step we have to solve the system of the algebraic equations (2.1*a*) and in the second step to insert the solution obtained into equation (2.1*b*).

2.1. Diffraction in an ideal crystal

In the following we shall deal with the diffraction of a particle in a simple crystal lattice forming a slab:

$$\begin{aligned} &\boldsymbol{R}_m = m_1\boldsymbol{a}_1 + m_2\boldsymbol{a}_2 + m_3\boldsymbol{a}_3 \qquad \boldsymbol{m} = (m_1, m_2, m_3) \qquad Q_m = Q \\ &m_1, m_2 = 0, \pm1, \pm2, \ldots, \pm\infty \qquad m_3 = 0, 1, 2, \ldots, N. \end{aligned} \tag{2.3}$$

The origin of the orthogonal coordinate system lies at the lattice point $(0, 0, 0)$; the plane Oxy coincides with the crystal surface plane $(\boldsymbol{a}_1, \boldsymbol{a}_2)$. The axis O$z$ (the unit vector

$\boldsymbol{e}_3$) and the vector $\boldsymbol{a}_1 \times \boldsymbol{a}_2$ point into the crystal and $a_{3z} > 0$. The lattice $(\boldsymbol{g}_1, \boldsymbol{g}_2, \boldsymbol{g}_3)$ is reciprocal to the three-dimensional lattice $(\boldsymbol{a}_1, \boldsymbol{a}_2, \boldsymbol{a}_3)$. The lattice $(\boldsymbol{b}_1, \boldsymbol{b}_2)$ is reciprocal to the two-dimensional lattice $(\boldsymbol{a}_1, \boldsymbol{a}_2)$; thus $\boldsymbol{b}_i \perp \boldsymbol{e}_3 (i, j = 1, 2)$.

The problem has a two-dimensional discrete translation symmetry in the plane $(\boldsymbol{a}_1, \boldsymbol{a}_2)$. Thus the components of the wavevectors of the reflected, refracted and transmitted waves parallel to the crystal surface are of the form

$$\boldsymbol{k}''_{pq} = \boldsymbol{k}'' + p\boldsymbol{b}_1 + q\boldsymbol{b}_2. \tag{2.4}$$

Here $\boldsymbol{k}''$ is the component of the wavevector $\boldsymbol{k} = \boldsymbol{k}'' + k_z\boldsymbol{e}_3 (k_z > 0)$ of the incident wave (2.2) parallel to the surface, and p, q are integers. The wavevectors $\boldsymbol{K}^-_{pq}(\boldsymbol{k})$ and $\boldsymbol{K}^+_{pq}(\boldsymbol{k})$ of the reflected and transmitted waves, respectively, are

$$\boldsymbol{K}^{\pm}_{pq} = \boldsymbol{k}''_{pq} \pm \boldsymbol{e}_3 K_{pqz}. \tag{2.5}$$

Considering merely elastic scattering processes, $|\boldsymbol{K}^{\pm}_{pq}(\boldsymbol{k})| = k = 2\pi/\lambda$ must hold, so that

$$K_{pqz} \equiv K_{pqz}(\boldsymbol{k}) = +[k^2 - (\boldsymbol{k}''_{pq})^2]^{1/2}. \tag{2.6}$$

Further let us define the quantities

$$\theta^{\pm}_{pq} \equiv \theta^{\pm}_{pq}(\boldsymbol{k}) = \boldsymbol{a}_3 \cdot \boldsymbol{K}^{\pm}_{pq}(\boldsymbol{k}) = \boldsymbol{a}''_3 \cdot \boldsymbol{k}''_{pq} \pm a_{3z} K_{pqz}. \tag{2.7}$$

From (2.6) it can be seen that there is a finite number (depending on the wavelength λ of the incident radiation and the angle γ of incidence) of reflected waves with real $K_{pqz}(\boldsymbol{k})$ and an infinite number of non-radiative waves with pure imaginary $K_{pqz}(\boldsymbol{k})$, which will be called evanescent waves.

2.2. The Bragg reflection condition and gamma diagrams

Let $\boldsymbol{k}^{pql}_B$ be the wavevector of the incident plane wave (2.2) satisfying the Bragg condition for the reflection in the direction $\boldsymbol{K}^-_{pq}(\boldsymbol{k}^{pql}_B)$, i.e.

$$|\boldsymbol{K}^-_{pq}(\boldsymbol{k}^{pql}_B)| \equiv |\boldsymbol{k}^{pql}_B + p\boldsymbol{g}_1 + q\boldsymbol{g}_2 - l\boldsymbol{g}_3| = |\boldsymbol{k}^{pql}_B| \tag{2.8}$$

with integer l, holds. It has been shown in [19] that (2.8) is equivalent to

$$\theta^+_{00}(\boldsymbol{k}^{pql}_B) - \theta^-_{pq}(\boldsymbol{k}^{pql}_B) = 2\pi l \qquad l \text{ integer}. \tag{2.9}$$

If the wavevector $\boldsymbol{k}$ of the incident wave is near the Bragg reflection position, then

$$\theta^+_{00}(\boldsymbol{k}) = \theta^-_{pq}(\boldsymbol{k}) + 2\pi l + \eta_{pql}(\boldsymbol{k}) \qquad |\eta_{pql}(\boldsymbol{k})| \ll 1 \tag{2.10}$$

whereby $\eta_{pql}(\boldsymbol{k}) \to 0$ if $\boldsymbol{k} \to \boldsymbol{k}^{pql}_B$.

Let us mention that, when deducing (2.9) and (2.10), we have not supposed that the reflected wave $\boldsymbol{K}^-_{pq}(\boldsymbol{k})$ lies in the plane of incidence $(\boldsymbol{k}, \boldsymbol{e}_3)$. Thus the parameter $\eta_{pql}(\boldsymbol{k})$, defined by equation (2.10), allows us to handle coplanar and non-coplanar reflections by the same general procedure.

In most experiments the plane of incidence and the wavelength are kept constant and only the angle γ of incidence (measured from the normal to the surface) varies. All quantities in (2.10) are then functions of the angle γ of incidence and equation (2.9) can be expressed as

$$\Gamma^-_{pq}(\gamma) = 2\pi l + \eta_{pql}(\gamma) \tag{2.11}$$

with

$$\Gamma^-_{pq}(\gamma) \equiv \theta^+_{00}(\gamma) - \theta^-_{pq}(\gamma). \tag{2.12}$$

The connection between the parameter η and the angle γ of incidence expressed by equation (2.11) can be made more transparent using 'gamma diagrams'. Examples of the gamma diagrams Γ^-_{pq} versus $\gamma \in (-90^\circ, 90^\circ)$ are plotted in figure 1. For (p, q) other than those given in the figure the values of K_{pqz} from (2.6) are purely imaginary in the whole interval of γ, i.e. the corresponding waves are evanescent waves and thus will be excluded from our considerations. The wave $\boldsymbol{K}^-_{-1,-1}$ becomes an evanescent wave for $\gamma \in (-90^\circ, 1.531^\circ)$ and similarly for all others except the wave $\boldsymbol{K}^-_{00}$. The reflections $\boldsymbol{K}^-_{00}$, $\boldsymbol{K}^-_{-1,-2}$ and $\boldsymbol{K}^-_{12}$ are coplanar; the others are non-coplanar. Putting Γ^-_{pq} equal to $2\pi l$, $l = 0, 1, 2, -1$, we obtain the angles γ^B_{pql} of incidence for the Bragg reflections, the diffraction vector being $p\boldsymbol{g}_1 + q\boldsymbol{g}_2 - l\boldsymbol{g}_3$. It can be seen that the Bragg reflection condition cannot be satisfied in this case for $\boldsymbol{K}^-_{0-1}$, $\boldsymbol{K}^-_{01}$, $\boldsymbol{K}_{0-2}$, $\boldsymbol{K}^-_{02}$, $\boldsymbol{K}^-_{1-1}$ and $\boldsymbol{K}^-_{-11}$.

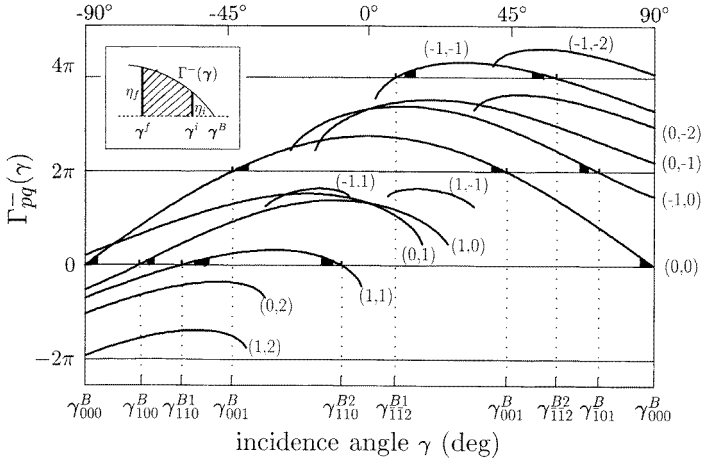

Figure 1. Gamma diagrams $\Gamma^-_{pq}(\gamma) \equiv \theta^+_{00}(\gamma) - \theta^-_{pq}(\gamma)$ for the BCC lattice with $\boldsymbol{a}_1 = a(1, 0, 0)$, $\boldsymbol{a}_2 = a(0, 1, 0)$, $\boldsymbol{a}_3 = (a/2)(1, 1, 1)$, $\lambda/a = 0.725$, $\boldsymbol{k}'' \| \boldsymbol{a}_1 + 2\boldsymbol{a}_2$, $\boldsymbol{a}_1$ and $\boldsymbol{a}_2$ lying in the crystal surface plane. The angle γ^B_{pql} is the angle of incidence, for which the Bragg diffraction condition $\Gamma^-_{pq}(\gamma^B_{pql}) = 2\pi l$ is satisfied. The inset demonstrates the distance $\eta_{pql}(\gamma) = \Gamma^-_{pq}(\gamma) - 2\pi l \geqslant 0$ determining the position and the width of the Darwin plateau (3.7). In the main picture the scale of the shaded Darwin plateaux could not be preserved.

Let us note that the gamma diagrams in figure 1 replace the traditional Ewald sphere in the reciprocal $\boldsymbol{k}$-space.

2.3. Solution of the Ewald equations

Because of the two-dimensional translational symmetry of our problem the solution of (2.1*a*) can be expressed as a superposition of plane waves [19, 21]:

$$\phi^{n_1 n_2 n_3}(\boldsymbol{R_n}) = \exp[\mathrm{i}\boldsymbol{k}'' \cdot (n_1\boldsymbol{a}_1 + n_2\boldsymbol{a}_2)] \sum_j c'_j \exp(\mathrm{i}n_3\psi_j)$$
$$= \sum_j c'_j \exp(\mathrm{i}\boldsymbol{\kappa}_j \cdot \boldsymbol{R_n}) \tag{2.13}$$

with the wavevectors

$$\boldsymbol{\kappa}_j = \boldsymbol{k}'' + (1/2\pi)(\psi_j - \boldsymbol{k}'' \cdot \boldsymbol{a}_3)\boldsymbol{g}_3 \qquad \text{i.e. } \psi_j = \boldsymbol{a}_3 \cdot \boldsymbol{\kappa}_j \tag{2.14}$$

characterizing the refracted waves. The wavevectors $\boldsymbol{\kappa}_j + p_1\boldsymbol{g}_1 + p_2\boldsymbol{g}_2 + p_3\boldsymbol{g}_3$ (p_j integers), and $\boldsymbol{\kappa}_j$ are equivalent.

Substituting the *Ansatz* (2.13) into (2.1*a*) we get firstly the dispersion relation (2.18*a*)–(2.18*c*) for the parameters ψ_j in (2.14) determining the wavevectors of refracted waves and secondly the inhomogeneous system of linear algebraic equations for the amplitudes c'_j.

We can then evaluate the wavefunction (2.1*b*) and the reflectivity and transmittivity of a slab. Further we confine ourselves to the case of a semi-infinite crystal (i.e. $N \to \infty$). After evaluating the wavefunction (2.1*b*) we have found for the reflectivity of semi-infinite crystal [19, 21]†

$$\mathcal{R}(\boldsymbol{K}^-_{pq}(\boldsymbol{k})) = |R_1(\boldsymbol{K}^-_{pq}(\boldsymbol{k}))|^2 |R_2(\boldsymbol{K}^-_{pq}(\boldsymbol{k}))|^2 k_z / K_{pqz} \tag{2.15}$$

where

$$R_1(\boldsymbol{K}^-_{pq}(\boldsymbol{k})) = \frac{\exp(\mathrm{i}\psi_1) - \exp(\mathrm{i}\theta^+_{00})}{\exp(\mathrm{i}\psi_1) - \exp(\mathrm{i}\theta^-_{pq})} \tag{2.16}$$

$$R_2(\boldsymbol{K}^-_{pq}(\boldsymbol{k})) = \prod_{j\neq 1}{}' \frac{\exp(\mathrm{i}\psi_j) - \exp(\mathrm{i}\theta^+_{00})}{\exp(\mathrm{i}\psi_j) - \exp(\mathrm{i}\theta^-_{pq})} \frac{\exp(\mathrm{i}\theta^+_j) - \exp(\mathrm{i}\theta^-_{pq})}{\exp(\mathrm{i}\theta^+_j) - \exp(\mathrm{i}\theta^+_{00})}. \tag{2.17}$$

The quantities ψ_j in equations (2.16) and (2.17) are solutions of the dispersion relation [19, 21]

$$1 + QS'(\boldsymbol{k}) + \frac{2\pi\mathrm{i}Q}{|\boldsymbol{a}_1 \times \boldsymbol{a}_2|} \sum_{pq} \frac{1}{K_{pqz}} \times \left[\frac{\exp(\mathrm{i}\theta^+_{pq})}{\exp(\mathrm{i}\psi) - \exp(\mathrm{i}\theta^+_{pq})} + \frac{\exp(-\mathrm{i}\theta^-_{pq})}{\exp(-\mathrm{i}\psi) - \exp(-\mathrm{i}\theta^-_{pq})} \right] = 0 \tag{2.18a}$$

where

$$S'(\boldsymbol{k}) = \sum_{(n_1 n_2)\neq(00)}{}' \frac{\exp(\mathrm{i}k|n_1\boldsymbol{a}_1 + n_2\boldsymbol{a}_2|)}{|n_1\boldsymbol{a}_1 + n_2\boldsymbol{a}_2|} \exp[\mathrm{i}\boldsymbol{k}'' \cdot (n_1\boldsymbol{a}_1 + n_2\boldsymbol{a}_2)] \tag{2.19a}$$

is the well known two-dimensional Ewald optical lattice sum [24, 25], the imaginary part of which is

$$\mathrm{Im}[S'(\boldsymbol{k})] = \frac{2\pi}{|\boldsymbol{a}_1 \times \boldsymbol{a}_2|} \sum_{K^2_{pqz}>0}{}' \frac{1}{K_{pqz}} - k. \tag{2.19b}$$

This formula follows directly from equation (16) of [19].

Further the scattering length Q can be expressed as

$$1/Q = 1/Q_0 + \mathrm{i}k \tag{2.20}$$

where $\mathrm{Im}\, Q_0 < 0$ in a crystal with absorption [18, 20].

In a semi-infinite crystal, only those solutions ψ_j of the dispersion relation (2.18*a*) and (2.18*b*) should be considered which generate decreasing waves (2.13) in a crystal with absorption ($\mathrm{Im}\, Q_0 < 0$), i.e. those ψ_j for which

$$\mathrm{Im}\,\psi_j > 0 \text{ when } \mathrm{Im}\, Q_0 < 0 \tag{2.21}$$

holds. In a crystal without absorption this condition should be understood in the limit $\mathrm{Im}\, Q_0 \to 0^-$.

† Equations (2.15)–(2.17) were deduced originally by us in [22] for the reflection of the s-polarized electromagnetic waves. The paper by Avron *et al* [23] should be mentioned too.

Using (2.19) and (2.20) the dispersion relation (2.18*a*) can be expressed in the following form appropriate for finding its solutions ψ:

$$L^{>}(\psi;\boldsymbol{k}) + L^{<}(\psi;\boldsymbol{k}) = h^0\{1 + Q_0\,\mathrm{Re}[S'(\boldsymbol{k})]\} \tag{2.18b}$$

with

$$L^{>}(\psi;\boldsymbol{k}) = -\frac{1}{2}\sum_{\substack{pq\\K^2_{pqz}>0}} \frac{1}{a_{3z}K_{pqz}}\frac{\sin(a_{3z}K_{pqz})}{\sin[(\psi-\theta^+_{pq})/2]\sin[(\psi-\theta^-_{pq})/2]} \tag{2.22}$$

$$L^{<}(\psi;\boldsymbol{k}) = \sum_{\substack{pq\\K^2_{pqz}<0}} \frac{1}{a_{3z}|K_{pqz}|}\left[1 - \frac{\sinh(a_{3z}|K_{pqz}|)}{\cosh(a_{3z}|K_{pqz}|) - \cos(\psi - \boldsymbol{a}''_3\boldsymbol{k}''_{pq})}\right] \tag{2.23}$$

being the finite and infinite sums over all (pq) for which K_{pqz} is real and/or purely imaginary, respectively. Furthermore

$$h^0 = \frac{|\boldsymbol{a}_1 \times \boldsymbol{a}_2|}{2\pi a_{3z}Q_0}. \tag{2.24}$$

Considering (2.9) we can see that the wavevector $\boldsymbol{k}$ of the incident wave is in the Bragg position for the reflection in the direction of the wavevector $\boldsymbol{K}^-_{pq}(\boldsymbol{k})$ if in the dispersion relation (2.18*a*) and (2.18*b*) the poles $\theta^+_{00}(\boldsymbol{k})$ and $\theta^-_{pq}(\boldsymbol{k})$ coincide mod 2π.

Another form of the dispersion relation will prove useful when we need to evaluate reflectivity. Hence, let us separate in (2.18*a*) and/or (2.18*b*) the terms corresponding to the poles θ^+_{00} and θ^-_{mn}, converting it into the form

$$b^0_{00}\frac{\exp(\mathrm{i}\theta^+_{00})}{\exp(\mathrm{i}\psi) - \exp(\mathrm{i}\theta^+_{00})} + b^0_{mn}\frac{\exp(-\mathrm{i}\theta^-_{mn})}{\exp(-\mathrm{i}\psi) - \exp(-\mathrm{i}\theta^-_{mn})} = F_{00,mn}(\psi) \tag{2.18c}$$

where

$$b^0_{pq} = \mathrm{i}\beta_{pq} = -\frac{i}{h^0 a_{3z}K_{pqz}} \tag{2.25}$$

and

$$F_{uv,mn}(\psi) = F^{(1)}_{uv,mn}(\psi) + \mathrm{i}F^{(2)}_{uv,mn}(\psi) \tag{2.26a}$$

$$F^{(1)}_{uv,mn}(\psi) = 1 + Q_0\,\mathrm{Re}[S'(\boldsymbol{k})] + \tfrac{1}{2}\beta_{uv}\frac{\sin(\psi-\theta^-_{uv})}{1-\cos(\psi-\theta^-_{uv})} - \tfrac{1}{2}\beta_{mn}\frac{\sin(\psi-\theta^+_{mn})}{1-\cos(\psi-\theta^+_{mn})} + \frac{1}{h^0}Z_{uv,mn}(\psi) \tag{2.26b}$$

$$F^{(2)}_{uv,mn}(\psi) = -\tfrac{1}{2}(\beta_{uv} + \beta_{mn}) \tag{2.26c}$$

$$Z_{uv,mn}(\psi) = Z_{mn,uv}(\psi) = {\sum_{\substack{pq\\(pq)\neq(uv)\\(pq)\neq(mn)\\K^2_{pqz}>0}}}' \frac{1}{2a_{3z}K_{pqz}}\frac{\sin(a_{3z}K_{pqz})}{\sin[(\psi-\theta^+_{pq})/2]\sin[(\psi-\theta^-_{pq})/2]} - L^{<}(\psi,\boldsymbol{k}). \tag{2.26d}$$

Equations (2.26*a*)–(2.26*d*) hold for $(uv) \neq (mn)$. The case $(uv) = (mn)$ is to be considered separately in a similar way.

Finally, using the same algebraic procedure as in [21] from (2.18*c*) it follows that

$$\frac{\exp(\mathrm{i}\psi)-\exp(\mathrm{i}\theta_{00}^{+})}{\exp(\mathrm{i}\psi)-\exp(\mathrm{i}\theta_{mn}^{-})}\exp\left(\frac{-\mathrm{i}(\theta_{00}^{+}-\theta_{mn}^{-})}{2}\right)\left(\frac{K_{00z}}{K_{mnz}}\right)^{1/2}$$
$$=-[Y_{mn}(\psi)\mp[Y_{mn}^{2}(\psi)-1]^{1/2}] \tag{2.27}$$

where

$$Y_{mn}(\psi)\equiv Y_{mn}(\psi,\boldsymbol{k})=-\frac{1}{2}\left[\sqrt{\frac{K_{00z}}{K_{mnz}}}+\sqrt{\frac{K_{mnz}}{K_{00z}}}\right]\cos\left(\frac{\theta_{00}^{+}-\theta_{mn}^{-}}{2}\right)$$
$$+h^{0}a_{3z}\sqrt{K_{00z}K_{mnz}}F_{00,mn}^{(1)}(\psi)\sin\left(\frac{\theta_{00}^{+}-\theta_{mn}^{-}}{2}\right). \tag{2.28}$$

3. Diffraction by a semi-infinite crystal

In this section we address first the problem of the solution of the dispersion relation (2.18*b*) in the short-wavelength region. Then, considering the results obtained, we express reflectivity $\mathcal{R}^{1}(\boldsymbol{K}_{pq}^{-}(\boldsymbol{k}))$ in the transparent form (3.3) appropriate for further treatment.

3.1. Solution of the dispersion relation

The left-hand side of the dispersion relation (2.18*b*) contains the variable ψ and the given parameters $\theta_{pq}^{+}(\boldsymbol{k})$, $\theta_{pq}^{-}(\boldsymbol{k})$ and $K_{pqz}(\boldsymbol{k})$ only. It is a periodic function of ψ with the period 2π, with poles for $\psi=\theta_{pq}^{+}+n2\pi$ and/or $\psi=\theta_{pq}^{-}+n2\pi$ (n integer) increasing or decreasing in the neighbourhood of the poles θ_{pq}^{+} or θ_{pq}^{-}, respectively. As the right-hand side of (2.18*b*) does not depend on the variable ψ, the real solutions ψ_j can be obtained graphically as intersections of the plot of left-hand side of (2.18*b*) with the horizontal straight line

$$h=h^{0}\{1+Q_{0}\,\mathrm{Re}[S'(\boldsymbol{k})]\}\qquad \mathrm{Im}\,Q_{0}=0. \tag{3.1}$$

Since, for neutron diffraction, $a/Q_{0}\approx 10^{4}$ and approximately $|\mathrm{Re}[S'(\boldsymbol{k})]|=2\pi/ka^{2}$, the term $Q_{0}\,\mathrm{Re}\,S'$ on the right-hand side of (3.1) can be neglected in the short-wavelength region where $ka\simeq 1$.

We can see that with each pole $\theta_{pq}^{+}(\theta_{pq}^{-})$ a solution $\psi_{pq}^{+}(\psi_{pq}^{-})$ as the function of $(\boldsymbol{k};Q_{0},\boldsymbol{a}_{i})$ can be associated. In particular, the solution near the pole θ_{00}^{+} which is the most important for the evaluation of the reflectivity will be denoted as $\psi_{1}\equiv\psi_{00}^{+}$. As the second infinite sum on the left-hand side of (2.18*b*), $L^{<}(\psi;\boldsymbol{k})$, is convergent very quickly and the approximate value of any ψ_j is known, its exact value can be evaluated with arbitrary precision by using an appropriately large number of terms on the left-hand side of (2.18*b*). In this sense the dispersion equation (2.18*a*) and/or (2.18*b*) can be solved exactly. We performed the analytical study of the dispersion relation in [26] in detail.

As an example let us consider the diffraction at an angle of incidence near the angle γ_{-101}^{B} found in figure 1. The sum $L^{>}(\psi,\gamma)$ in (2.18*a*) is now extended over $(pq)=(0,0)$, $(-1,0)$, $(0,-1)$, $(-1,-1)$, $(0,-2)$, $(-1,-2)$. In figure 2 we give 'the dispersion plot' $L^{>}(\psi;\gamma_{0})$ versus $\psi\in(-\pi,\pi)$ for $\gamma_{0}=\gamma_{-101}^{B}-0.80^{\circ}=71.85^{\circ}$, other parameters being the same as in figure 1. As $h^{0}=4.2\times10^{4}$, which corresponds to the scattering length of Si atoms [10, p 307], it can be seen that at this value of γ all solutions ψ_j^{+} of the equation $L^{>}(\psi,\gamma)=h^{0}$ are real and very close to the corresponding poles θ_j^{+}. If the wavevector $\boldsymbol{k}$ of the incident wave approaches the Bragg reflection position, in our case if $\gamma\to\gamma_{-101}^{B}$, the neighbourhood of the poles $\theta_{-1,0}^{-}$ and $\theta_{0,0}^{+}$ must be examined very carefully.

Thus in figure 3(a) we plot $L^{>}(\psi;\gamma)$ versus $\psi \in (0.557\,67, 0.558\,12)$ for the parameter $\gamma^{f}_{-101} = \gamma^{B}_{-101} - 2.12$ s in detail. At this value of γ the minimum touches the straight horizontal line at $h^0 = 4.2 \times 10^4$. On the other hand, the maximum touches this line at $\gamma = \gamma^{i}_{-101} = \gamma^{B}_{-101} - 0.01$ s (see figure 3(b)). Since both the minimum and the maximum are pushed upwards if $\gamma \to \gamma^{B}_{-101}$, we can conclude that the line h^0 does not intersect the dispersion plot for $\gamma \in (\gamma^{f}_{-101}, \gamma^{i}_{-101})$ and thus the solution ψ_1 is complex in this interval.

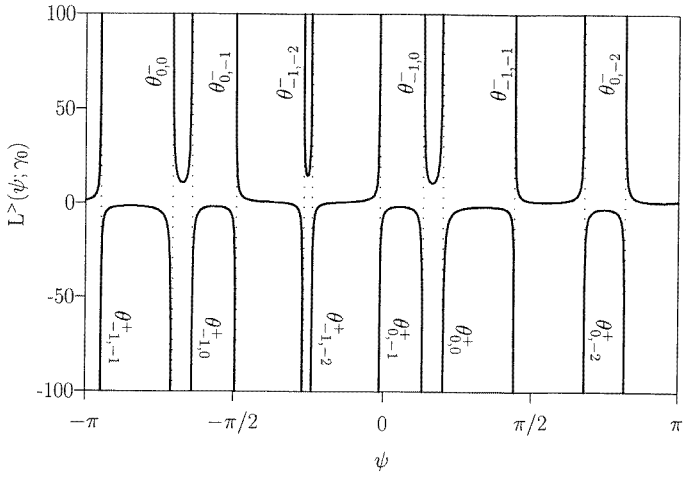

Figure 2. The dispersion plot for the incidence angle $\gamma_0 = \gamma^{B}_{-101} - 0.80^{\circ} = 71.85^{\circ}$ (for further parameters see the caption of figure 1). The intersections of the dispersion plot with the horizontal straight line h^0 give the solutions of the dispersion relation in the twelve-beam approximation.

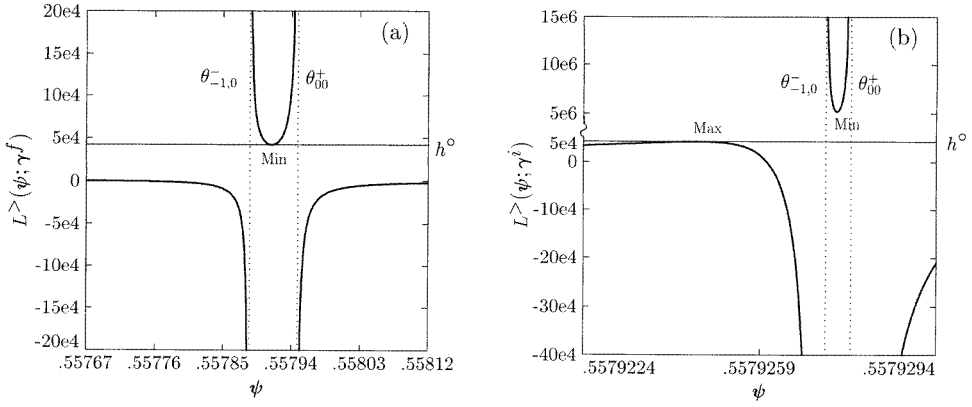

Figure 3. Details of the dispersion plot in figure 2 for the incidence angles γ^{f}_{-101} and γ^{i}_{-101}, for which $\theta^{-}_{-10}(\gamma)$ is close to $\theta^{+}_{00}(\gamma)$. (a) At $\gamma = \gamma^{f}_{-101} = \gamma^{B}_{-101} - 2.12$ s the minimum touches the horizontal straight line $h^0 = 4.2 \times 10^4$ which is the right-hand side of the dispersion relation (2.18*b*). The very flat maximum appears at $\psi = 0.557\,57$. If the pole $\theta^{-}_{01}(\gamma)$ is pushed to the pole $\theta^{+}_{00}(\gamma)$, both extremes move upwards. (b) At $\gamma = \gamma^{i}_{-101} = \gamma^{B}_{-101} - 0.01$ s the maximum touches the horizontal line h^0.

3.2. Reflectivity profiles

Let us suppose that the Bragg condition (2.9) may be fulfilled for one couple, say $(\theta_{00}^{+}, \theta_{rs}^{-})$, only. Since ψ_j^+ is very close to the corresponding pole θ_j^+ for $h^0 \gg 1$, in (2.15)–(2.17) we can make the following approximations:

$$\begin{aligned} &R_2(\boldsymbol{K}_{pq}^{-}(\boldsymbol{k})) \simeq 1, \\ &\mathcal{R}(\boldsymbol{K}_{pq}^{-}(\boldsymbol{k})) \simeq |R_1(\boldsymbol{K}_{pq}^{-}(\boldsymbol{k}))|^2 k_z / K_{pqz} \equiv \mathcal{R}^1(\boldsymbol{K}_{pq}^{-}(\boldsymbol{k})) \end{aligned} \tag{3.2}$$

where $R_1(\boldsymbol{K}_{pq}^{-}(\boldsymbol{k}))$ is given by (2.16).

First let us analyse the reflectivity $\mathcal{R}^1(\boldsymbol{K}_{pq}^{-}(\boldsymbol{k}))$ qualitatively. Since $\psi_1(\boldsymbol{k})$ is very near $\theta_{00}^{+}(\boldsymbol{k})$, the value of the numerator of the fraction $R_1(\boldsymbol{K}_{pq}^{-}(\boldsymbol{k}))$ is always small. On the other hand the value of the denominator of (2.16) depends on whether $\boldsymbol{K}_{pq}^{-}(\boldsymbol{k})$ is near to or far from the Bragg reflection. If $\theta_{pq}^{-}(\boldsymbol{k})$ and $\theta_{00}^{+}(\boldsymbol{k})$ do not satisfy condition (2.10), the difference $\exp(\mathrm{i}\theta_{pq}^{-}(\boldsymbol{k})) - \exp(\mathrm{i}\theta_{00}^{+}(\boldsymbol{k}))$ is not small and thus the respective reflectivity $\mathcal{R}^1(\boldsymbol{K}_{pq}^{-}(\boldsymbol{k}))$ is very small. If $\theta_{pq}^{-}(\boldsymbol{k})$ and $\theta_{00}^{+}(\boldsymbol{k})$ do satisfy condition (2.10), the situation changes dramatically. The numerator and denominator in (2.16) have now 'almost the same small values' and their quotient should be considered very carefully.

To evaluate the reflectivity $\mathcal{R}^1(\boldsymbol{K}_{pq}^{-}(\boldsymbol{k}))$ we shall use equation (2.27). Comparing its left-hand side with equations (2.16) and (3.2) we obtain

$$\mathcal{R}^1(\boldsymbol{K}_{pq}^{-}(\boldsymbol{k})) = |Y_{pq}(\psi_1, \boldsymbol{k}) \mp [Y_{pq}^2(\psi_1, \boldsymbol{k}) - 1]^{1/2}|^2. \tag{3.3}$$

Thus we have expressed the reflectivity $\mathcal{R}^1(\boldsymbol{K}_{pq}^{-}(\boldsymbol{k}))$ in the direction $\boldsymbol{K}_{pq}^{-}(\boldsymbol{k})$ by means of the function $Y_{pq}(\psi_1, \boldsymbol{k})$ from (2.28) with ψ_1 being the solution of the dispersion relation (2.18*b*) lying near the pole θ_{00}^{+}. The result obtained is exact and should not be confused with the so-called two-beam approximation of the Bethe–von Laue dynamical theory of diffraction because it is valid for both coplanar and non-coplanar diffractions in and outside the Bragg peak regions, including special cases such as reflections for Bragg angles near $\pi/2$ or total external reflection, and also because when evaluating ψ_1 from (2.18*b*) the influence of all beams is taken into account. Note that ψ_1 enters into (2.28) and thus into (3.3) via the term $F_{00,pq}^{(1)}(\psi_1)$, defined by (2.26*b*), rendering both the multiple diffractions in one atomic plane (the small term $Q_0 \operatorname{Re} S'$) and the influence of all other beams than (pq).

From (3.3) it follows the condition for the 'total reflection':

$$\mathcal{R}^1(\boldsymbol{K}_{pq}^{-}(\boldsymbol{k})) = 1 \qquad \text{if } Y_{pq}(\psi_1, \boldsymbol{k}) \text{ is real and } |Y_{pq}(\psi_1, \boldsymbol{k})| \leqslant 1. \tag{3.4}$$

On the other hand,

$$\mathcal{R}^1(\boldsymbol{K}_{pq}^{-}(\boldsymbol{k})) = \frac{1}{4|Y_{pq}(\psi_1, \boldsymbol{k})|^2} \qquad \text{if } |Y_{pq}(\psi_1, \boldsymbol{k})| \gg 1. \tag{3.5}$$

Using our results we shall analyse the reflectivity in the direction $\boldsymbol{K}_{rs}^{-}(\boldsymbol{k})$ near the Bragg peak, i.e. when

$$\theta_{00}^{+}(\boldsymbol{k}) = \theta_{rs}^{-}(\boldsymbol{k}) + 2\pi l + \eta_{rsl}(\boldsymbol{k}) \qquad |\eta_{rsl}(\boldsymbol{k})| \ll 1 \tag{3.6}$$

holds. Since in neutron diffraction $Q_0/a \ll 1$ and as there is no term with a resonant denominator in $F_{00;rs}^{(1)}(\psi)$ given by (2.26*b*) for $\psi = \psi_1$ (supposing as above that only one beam (rs) satisfies the Bragg reflection condition (3.6)), we may put in (2.28) $F_{00,rs}^{(1)}(\psi_1) = 1$, so that Y_{rs} depends only on the difference $\theta_{00}^{+}(\boldsymbol{k}) - \theta_{rs}^{-}(\boldsymbol{k})$ and not on ψ_1. Expressing now in equation (2.28) for Y_{rs} the difference $\theta_{00}^{+}(\boldsymbol{k}) - \theta_{rs}^{-}(\boldsymbol{k})$ by the parameter $\eta_{rsl}(\boldsymbol{k})$ from (3.6)

and then putting approximately $\sin(\eta_{rsl}/2) = \eta_{rsl}/2$ and $\cos(\eta_{rsl}/2) = 1$, we find that the condition (3.4) for the total reflection is fulfilled for $\eta_{rsl}(\boldsymbol{k})$ satisfying the condition

$$\begin{aligned}\eta^i_{rsl}(\boldsymbol{k}^{rsl}_B) &\equiv \frac{1}{h^0}\left[\frac{1}{\sqrt{a_{3z}K_{00z}(\boldsymbol{k}^{rsl}_B)}} - \frac{1}{\sqrt{a_{3z}K_{rsz}(\boldsymbol{k}^{rsl}_B)}}\right]^2 \\ &< \eta_{rsl}(\boldsymbol{k}) \\ &< \frac{1}{h^0}\left[\frac{1}{\sqrt{a_{3z}K_{00z}(\boldsymbol{k}^{rsl}_B)}} + \frac{1}{\sqrt{a_{3z}K_{rsz}(\boldsymbol{k}^{rsl}_B)}}\right]^2 \\ &\equiv \eta^f_{rsl}(\boldsymbol{k}^{rsl}_B) \end{aligned} \tag{3.7}$$

with $\boldsymbol{k}^{rsl}_B$ satisfying the Bragg reflection condition (2.8) and h^0 being real. In our above approximate considerations we have excluded the cases when K_{00z} or K_{rsz} is very small, i.e. when the incident or diffracted wave is in the grazing position.

The connection between the interval $(\eta^i_{pql}, \eta^f_{pql})$ and the corresponding region of incidence angles $(\gamma^i_{pql}, \gamma^f_{pql})$ given by equation (2.11) is shown in figure 1. In particular, let us analyse the important case when the horizontal straight line $2\pi l$ intersects a gamma diagram, say $\Gamma_{uv}(\gamma)$, very near its extreme. It can be shown that this case corresponds to diffraction when the Bragg angle θ^{uvl}_B is near $\pi/2$, where the results of the standard dynamical theory of diffraction cannot be used [12, 27, 28]. In particular the formula for the width of the Darwin plateau, $\Delta(\theta^{uvl}_B) \equiv |\gamma^f_{uvl} - \gamma^i_{uvl}|$, diverges (see, e.g., equation (9.37) in [10]). On the other hand, our approach, which does not require the idea of a dispersion surface, is directly applicable to this special case. In figure 4, using the gamma diagram technique we treat the symmetrical reflection $\boldsymbol{K}^-_{00}$ at the Bragg angle θ_B near $\pi/2$. We can see that, when the Bragg angle is shifted from $\pi/2$, the Darwin plateau splits into two narrower parts, the width of which is given far from $\theta_B = \pi/2$ by the formula [26]

$$\Delta(\theta^{00l}_B) = \frac{2Q_0(\lambda^{00l}_B)^2}{|\boldsymbol{a}_1 \times \boldsymbol{a}_2|a_{3z}\pi\sin(2\theta^{00l}_B)} \qquad 2\theta^{00l}_B = \pi - 2\gamma^B_{00l} \tag{3.8}$$

which agrees with that yielded by the standard dynamical theory of diffraction (see, e.g., equation (9.37) in [10]). A detailed analytical study of this problem in the framework of our theory has been given in [26].

If we drop the approximation $F^{(1)}_{00,rs}(\psi_1) = 1$ in (2.28), then to apply (3.3) we need to find ψ_1 from our dispersion equation (2.18*b*). Near the Bragg peak where the pole θ^+_{00} is close to the pole $\theta^-_{rs}(\mathrm{mod}\,2\pi)$, it is possible to keep only two terms with $(pq) = (00)$ and (rs) in (2.18*b*). Thus

$$\begin{aligned}-\frac{\sin(a_{3z}K_{00z})}{2a_{3z}K_{00z}}&\frac{1}{\sin[(\psi-\theta^+_{00})/2]\sin[(\psi-\theta^-_{00})/2]} \\ &-\frac{\sin(a_{3z}K_{rsz})}{2a_{3z}K_{rsz}}\frac{1}{\sin[(\psi-\theta^+_{rs})/2]\sin[(\psi-\theta^-_{rs})/2]} = h^0. \end{aligned} \tag{3.9}$$

Introducing here again the small parameter η_{rsl} from (3.6) and using the same approximations for the trigonometric functions as above, it is possible to show that the solution ψ_1 of (3.9) becomes complex for η_{rsl} from the interval (3.7). Obviously, when omitting the approximation $F^{(1)}_{00;rs}(\psi_1) = 1$ the term $Y_{rs}(\psi_1)$ becomes complex in the Darwin plateau region (3.7) and thus the condition (3.4) cannot now be fulfilled exactly.

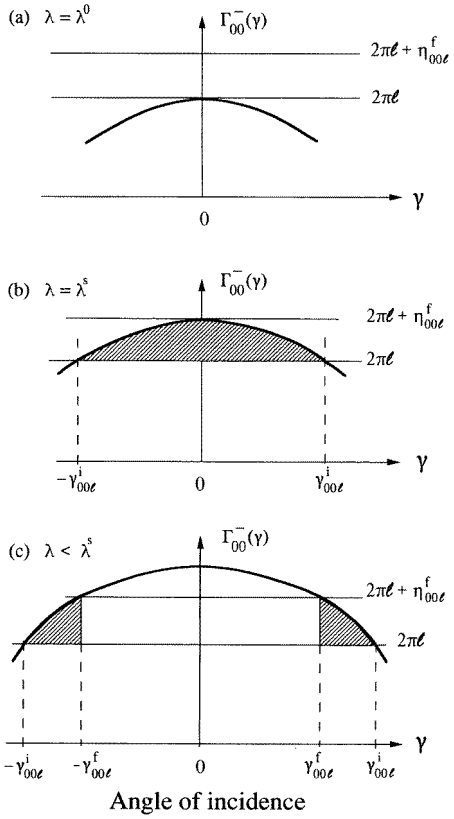

Figure 4. Gamma diagrams for the symmetrical reflection when the Bragg angle is near $\pi/2$. For the symmetrical reflection, condition (3.7) gives $\eta_{00l}^i = 0$, and thus the Darwin plateau begins just at the incidence angle $\gamma_{00l}^B(\lambda)$ at which the Bragg reflection condition (3.12) is satisfied. (a) If the Bragg reflection occurs at $\gamma_{00l}^B(\lambda^0) = 0$ (i.e. $\theta_B = \pi/2$) it can be proved that the line $2\pi l$ is a tangent to the corresponding gamma diagram and consequently the width of the Darwin plateau is zero. Decreasing the wavelength (according to (3.12) the angle $\gamma_{00l}^B(\lambda)$ is shifted from zero) the maximum of the gamma diagram moves up and the width of the Darwin plateau increases. (b) For $\lambda = \lambda^s$ the line $2\pi l + \eta_{00l}^f(\lambda^s)$ is a tangent to the corresponding gamma diagram and the width of the (shaded) Darwin plateau reaches its maximum value. (c) For $\lambda < \lambda^s$ the gamma diagram intersects the line $2\pi l + \eta_{00l}^f(\lambda)$ and the Darwin plateau splits into two parts.

In figure 5 we show the reflectivity profiles $\mathcal{R}^1(\boldsymbol{K}_{pq}^-(\gamma))$ for $(pq) = (-1, -1)$ and $(0, 0)$ calculated from (3.3) in the approximation $F_{00;\,pq}^{(1)}(\psi_1) = 1$. The former case corresponds to the non-coplanar reflection, and the latter to the symmetrical reflection where our equations (2.12) and (2.28) have very simple forms, namely

$$\Gamma_{00}^-(\gamma) = \theta_{00}^+ - \theta_{00}^- = 2a_{3z}k\cos\gamma = 2x \tag{3.10}$$

and

$$Y_{00}(\psi, \gamma) = -\cos x + h^0 F_{00,00}^{(1)}(\psi) x \sin x \tag{3.11}$$

respectively, equation (3.11) being valid for any angle γ of incidence including $\pi/2$. If the Bragg reflection condition (2.9) is satisfied for the specularly reflected wave $\boldsymbol{K}_{00}^-(\boldsymbol{k}_B^{00l})$,

from (2.9) and (3.10) it follows that

$$2a_{3z}k_B^{00l}\cos\gamma_{00l}^B = 2\pi l \tag{3.12}$$

which is the well known Bragg equation. We can see that the total reflection at grazing incidence is a special case of the Bragg reflection (2.9) for $l = 0$. The total reflection at grazing incidence was studied by us in [29] in detail.

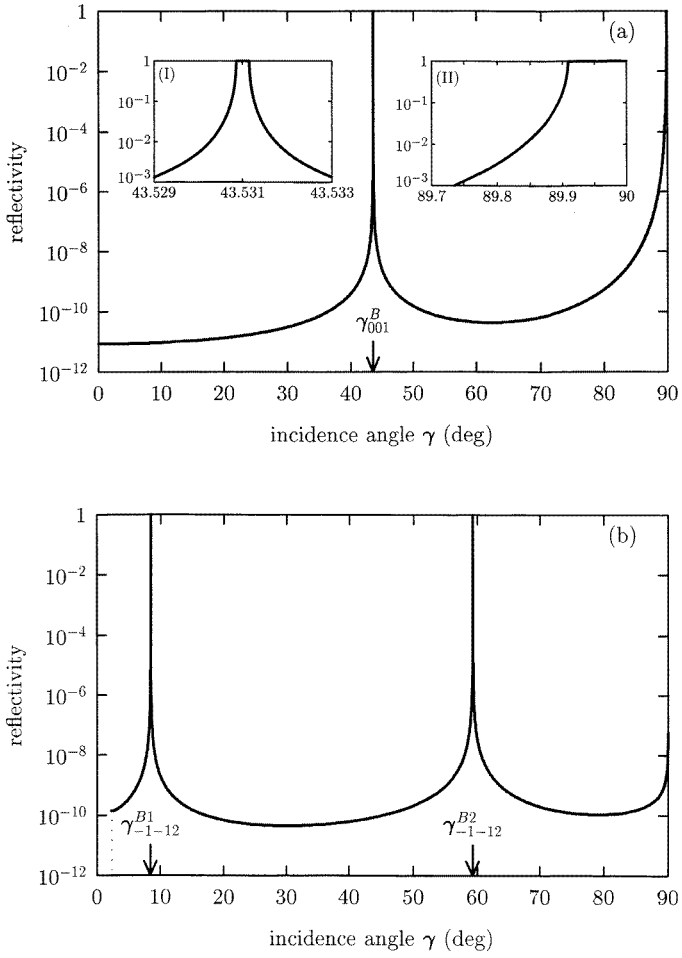

Figure 5. Reflectivities of a semi-infinite crystal into the directions (a) $\boldsymbol{K}_{00}^-$ and (b) $\boldsymbol{K}_{-1-1}^-$ as functions of the incidence angle measured from the normal to the surface. Note that both (a) the coplanar and (b) the non-coplanar reflectivities have been calculated from the same equation (3.3) in the approximation $F^{(1)}_{00;pq}(\Psi_1) = 1$, $(pq) = (0,0)$ or $(-1,-1)$; for the discussion of this approximation see the end of section 3.2. The crystal lattice, the wavelength and the plane of incidence are defined in the caption of figure 1. The insets show details of (I) the specular reflectivity around the Bragg reflection peak at γ_{001}^B and (II) the total external reflection at grazing incidence.

Near the symmetrical Bragg reflection we can write

$$Y_{00}(\psi,\gamma) \equiv Y_{00l}(\psi,\gamma;\gamma_{00l}^B) = (-1)^l[-1 - (\tfrac{1}{2}h^0(a_{3z}k_B^{00l})^2 \sin(2\gamma_{00l}^B)\delta\gamma + \mathrm{O}(\delta\gamma)^2)F^{(1)}_{00,00}(\psi)] \tag{3.13}$$

where $\delta\gamma = \gamma - \gamma^B_{00l}$ is the deviation from the Bragg reflection position. Introducing instead of the incidence angle γ the glancing angle $\theta = \pi/2 - \gamma$, we can see that equation (3.13) reminds us of the expression

$$y = -1 - \frac{E}{V(0)}(\theta_B - \theta)\sin(2\theta_B) \tag{3.14}$$

well known from the von Laue dynamical theory of diffraction (see equation (9.23) in [10]) for the potential V from (4.4).

On the other hand, far from the Bragg peaks we get from (3.5) and (3.11) in the approximation $F^{(1)}_{00;00}(\psi_1) = 1$ for the reflectivity of the specularly reflected beam:

$$\mathcal{R}^1(\boldsymbol{K}^-_{00}) = [2h^0(a_{3z}k\cos\gamma)\sin(a_{3z}k\cos\gamma)]^{-2}. \tag{3.15}$$

Let us consider for example the reflectivity $\mathcal{R}^1(\boldsymbol{K}^-_{-1-1})$ outside Bragg peaks for the incidence angle γ close to γ^B_{-101} for which Bragg reflection in the direction $\boldsymbol{K}^-_{10}$ appears (see figure 1). For $\gamma \to \gamma^B_{-101}$, according to (2.10), $\theta^+_{00}(\gamma) \to \theta^-_{-10}(\gamma)$ (mod 2π) holds and the same holds for ψ_1 which lies near $\theta^+_{00}(\gamma)$. Thus in equation (2.26*b*) for $F^{(1)}_{00;-1-1}(\psi_1)$ a term with the resonant denominator $1-\cos(\psi_1-\theta^-_{-10})$ appears, which destroys the approximation $F^{(1)}_{00;-1-1}(\psi_1) = 1$. Generally, this approximation fails always when the other beam satisfies the Bragg reflection condition. Hence the profile of $\mathcal{R}^1(\boldsymbol{K}^-_{-1-1}(\gamma))$ in figure 5(b) should be corrected in the neighbourhood of γ^B_{001}, γ^B_{-1-01} and $\gamma^B_{000} = \pi/2$. A more detailed analysis of this 'fine structure' of the reflectivity outside Bragg peaks, which could be of interest in truncation rod scattering [16, 17, 30], would exceed the aim and extent of this paper and will be published later.

In the dynamical theory of the diffraction of x-rays or particles it is usual to consider the thermal vibrations by means of the Debye–Waller factor. Thus in our case the scattering length becomes temperature dependent: $Q_0(T) = Q_0\exp(-W(T,\theta))$ [10, p 306]. The Debye–Waller factor narrows the region of the total reflection and deforms the profile of the reflectivity between the Bragg peaks.

4. Concluding remarks

4.1. Comparison with the Bethe–von Laue method

In the dynamical theory of diffraction due to Bethe [6] and von Laue [7] the crystal is considered as a continuous medium and the propagation of de Broglie waves in an infinite crystal is studied. Thus we have to solve the Schrödinger equation with the periodic potential $V(\boldsymbol{r}) = V(\boldsymbol{r}+\boldsymbol{R}_n) = \sum_{\boldsymbol{G}} V(\boldsymbol{G})\exp(\mathrm{i}\boldsymbol{G}\cdot\boldsymbol{r})$.

Applying the Bloch theorem we write the solution of the Schrödinger equation as

$$\psi(\boldsymbol{r}) = \sum_{\boldsymbol{G}} C_{\boldsymbol{\kappa}}(\boldsymbol{G})\exp[\mathrm{i}(\boldsymbol{\kappa}+\boldsymbol{G})\cdot\boldsymbol{r}]. \tag{4.1}$$

Using the well known procedure (see, e.g., [10, 18]) we get for the unknown vectors $\boldsymbol{\kappa}$ and for the expansion coefficients $C_{\boldsymbol{\kappa}}(\boldsymbol{G})$ the following system of homogeneous equations:

$$\left[E - V(0) - \frac{\hbar^2}{2m}(\boldsymbol{\kappa}+\boldsymbol{G})^2\right] C_{\boldsymbol{\kappa}}(\boldsymbol{G}) = \sum_{\boldsymbol{G}'\neq\boldsymbol{G}} V(\boldsymbol{G}-\boldsymbol{G}')C_{\boldsymbol{\kappa}}(\boldsymbol{G}'). \tag{4.2}$$

The determinant of the infinitely large system of algebraic equations (4.2) will be denoted as $\mathcal{D}(\boldsymbol{\kappa})$ and the equation for the unknown vector $\boldsymbol{\kappa}$ reading $\mathcal{D}(\boldsymbol{\kappa}) = 0$ is called the dispersion relation.

If the external wave $A\exp(\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r})$ impinges upon a semi-infinite crystal bordered by the surface Oxy, the wavevectors $\boldsymbol{\kappa}_j$ of the waves (4.1) propagating inside the crystal must satisfy besides (4.3) the conditions

$$\boldsymbol{\kappa}_j = \boldsymbol{k}'' + \kappa_{jz}\boldsymbol{e}_3. \tag{4.3}$$

Introducing (4.3) into equation $\mathcal{D}(\boldsymbol{\kappa}) = 0$ we get the fundamental equation of the Bethe–von Laue theory $\mathcal{D}(\kappa_z) = 0$ for the evaluation of the z components of the wavevectors of the de Broglie waves in the crystal. Solving (4.2) and using the standard boundary conditions the intensity of the refracted and reflected waves can be computed.

Let us compare the methods and results of the dynamical theory of diffraction on a system of point scatterers, based on the solution of the Schrödinger differential equation and on the Ewald system of algebraic equations (2.1*a*) and (2.1*b*).

(i) When studying the diffraction of neutrons on a crystal with a simple lattice, the potential

$$V(\boldsymbol{r}) = \frac{2\pi\hbar^2}{m}Q_0\sum_{\boldsymbol{n}}\delta(\boldsymbol{r}-\boldsymbol{R}_{\boldsymbol{n}}) \qquad V(\boldsymbol{G}) = V(\boldsymbol{0}) = \frac{2\pi\hbar^2 Q_0}{m(\boldsymbol{a}_1\times\boldsymbol{a}_2)\cdot\boldsymbol{a}_3} \tag{4.4}$$

is used. Following to [18, p 177], this potential neglects the local-field effects whereas in the Ewald equations (2.1*a*) and (2.1*b*) they are rigorously taken into account. Let us also recall the critical remark made by James [8, p 61] concerning the use of the classical boundary conditions in the von Laue method. No boundary conditions are necessary for the Ewald system of algebraic equations (2.1*a*) and (2.1*b*).

(ii) The system of equations (4.2) is exact and different n-beam approximations for its solution are used [9, 10, 31]. We are not aware of any formulae for the reflectivity based on (4.2) which are of such simple analytical form and universal use as our formulae. The dispersion relation (2.18*a*)–(2.18*c*) and equation (2.15) for the reflectivity by a semi-infinite ideal crystal with a simple basis are exact. They hold for all wavelengths, for any value of the scattering length Q and any directions of the wavevectors of the incident or reflected waves, including the grazing incidence, Bragg angle near $\pi/2$, and both coplanar and non-coplanar reflections. Owing to the simple analytical form of our formulae their solutions can be found easily with arbitrary precision.

(iii) In the short-wavelength region an analytical comparison shows that equation (3.3) for the reflectivity $\mathcal{R}^1(\boldsymbol{K}^-_{pq})$ agrees with that obtained in the Bethe–von Laue theory when omitting terms higher than $\Delta(\theta-\theta_B)$ [21]. This omission is not possible for example for θ_B near $\pi/2$ (see section 3.2). On the other hand, in the long-wavelength region the Ewald equations yield the usual formulae of classical optics, while in the transient soft-ray region the corrections of the Snell law and the Fresnel reflection coefficient have been derived [32]. In this connection let us recall Ewald's words: 'Besides, my work has been attempting to establish the unity of classical optics throughout the entire range of wavelengths from infrared to x-rays. This general aspect has received little resonance.' [4].

4.2. Comparison with the Darwin method

Recently the classical Darwin method combined with different thin-film-optics algebraic manipulations has become very popular particularly in connection with the study of diffraction outside the Bragg peaks in truncation rod scattering [16, 17]. Considering the two-beam case of the Darwin theory, Takahashi and Nakatani [17] recently published the formula for the reflectivity of x-rays by a semi-infinite crystal formally identical with our result (3.3). The main difference is in the absence of the term $F^{(1)}_{00,pq}(\psi_1)$ in their deviation

parameter appearing in (2.25) in [17]. As shown in section 3.2 this term cannot be put equal to unity when the other beam satisfies the Bragg condition. Naturally, in this case the two-beam approximation is insufficient. In [17] the extension of the Darwin theory to the multiple-beam case is shown by studying the coplanar three-beam Bragg case. The general multiple-beam algebraic method in the Darwin dynamical theory of the diffraction of scalar waves was developed by Ignatovich [15] by employing the recurrence procedure [33]. He applies his method to the tetragonal simple lattice and the generalization of his resulting formulae to general lattices or to lattices with a basis is not evident. Note that, when calculating the reflection and transmission amplitudes of one atomic plane, he takes into account the multiple diffractions inside the atomic plane. Evaluating the two-dimensional lattice sum (2.19*a*), he did not use the traditional method introduced by Ewald [3] but employed a special procedure. It would be interesting to investigate whether his sophisticated procedure is applicable to more general lattice sums appearing for example in LEED [34] or x-ray diffraction [24].

4.3. Further perspectives

In this paper we have confined ourselves to the study of waves outside the crystal. The analysis of the wave field inside the crystal, leading to a new insight into the well known Ewald–Oseen theorem, was performed by one of us in [35].

Let us briefly mention the results obtained for more complicated cases. The problem of diffraction of particles by a crystal of finite thickness and simple crystal lattice was dealt with in [36]. The dispersion relation is the same as in the case of a semi-infinite crystal, but the formula for the reflectivity does not have such a simple form as (2.15)–(2.17). In the case of a crystal with s atoms of basis, the dispersion equation has the more complicated form [19]

$$\det \|\mathbf{I} - \mathbf{C} - \sum_{pq} (\{\exp[\mathrm{i}(\theta^{-}_{pq} - \psi)] - 1\}^{-1}\mathbf{B}_{pq} + \{\exp[-\mathrm{i}(\theta^{+}_{pq} - \psi)] - 1\}^{-1}\mathbf{D}_{pq})\| = 0 \tag{4.5}$$

where $\mathbf{I}$, $\mathbf{C}$, $\mathbf{B}_{pq}$ and $\mathbf{D}_{pq}$ are matrices of order s. The dispersion relation of the same algebraic form appears in the dynamical theory of diffraction of electromagnetic waves based on the Ewald discrete dipole model of a crystal, which has been reformulated and further developed in [37]. Neither the dispersion relation (4.5) nor the formulae for the intensities of the reflected and transmitted waves have been analysed yet. Moreover, in [38] we have dealt with the diffraction by a crystal with isolated defects. A further problem to address is the case when two (or more) beams $\boldsymbol{K}^{-}_{pq}(\boldsymbol{k})$ satisfy the Bragg reflection condition (2.8) simultaneously. Then three (or more) poles in the dispersion relation (2.18) coincide and thus when evaluating the reflectivity the term $R_2(\boldsymbol{K}^{-}_{pq}(\boldsymbol{k}))$ from (2.17) should be considered too. The Bragg reflection and simultaneous surface reflection at grazing incidence are an important example of this problem [13].

Ewald elaborated his procedure for the diffraction of electromagnetic waves by a system of point dipoles and thus it is difficult to take into account the atomic factors (appearing for example in x-ray diffraction or LEED) without losing the simple form of the resulting formulae [38]. Nevertheless, the Ewald discrete model of a crystal and the methods explained above were successfully applied to problems of surface optics in the long-wavelength region. In particular, the optical response of an adsorbed monolayer [25, 39] and surface-induced optical anisotropy [40] have been addressed. Recently the Ewald approach was developed to describe the propagation of electromagnetic

waves in magnetoelectric crystals [41] and the surface second-harmonic generation by centrosymmetric semiconductors [42].

The quantum-mechanical generalization of the Ewald method, giving exact and simple analytical formulae, can be used for testing some approximations used in the Bethe–von Laue and the Darwin theories. Besides this, the problem of the diffraction of particles by a semi-infinite crystal of delta-like potentials represents one of the rare problems of quantum mechanics which can be solved exactly in an analytical form and represents a problem of more general interest [43].

Acknowledgments

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