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Three-beam X-ray diffraction in a semi-infinite perfect crystal

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The approach to multibeam X-ray diffraction by Shen [*Acta Cryst.* (1986). A**42**, 525–533] has been carefully re-examined for three-beam Laue–Laue diffraction in a perfect non-absorbing semi-infinite crystal. A modified version of Shen's theory is shown to be equivalent to the Takagi–Taupin formalism. The perturbation of the two-beam integrated power when a third beam is being excited is thus expressed by non-diverging terms. Both *Aufhellung* and *Umweganregung* terms are recovered.

1. Introduction

The theory for multibeam X-ray diffraction in a perfect crystal may be developed along different routes all emerging from Maxwell's equations for the electrical field in a spatial periodic dielectric medium.

The original approach, the Ewald–von Laue theory (Ewald, 1917; von Laue, 1931), leads to the fundamental equations that are solved numerically for the amplitudes of the constituents of the X-ray wavefield for a general n-beam situation ($n \geq 3$) (Colella, 1974; Chang, 1984; Weckert & Hümmer, 1990, 1997; Stetsko & Chang, 1997).

Takagi and Taupin (Takagi, 1962, 1969; Taupin, 1964) primarily developed their theory of diffraction for distorted crystals. It is however easily adapted to perfect crystals of finite size. The Takagi–Taupin equations may be processed analytically (Thorkildsen, 1987; Thorkildsen & Larsen, 1998) and have been shown (Thorkildsen *et al.*, 2001) to give results in perfect agreement with the numerical solution of the fundamental equations for a symmetrical three-beam Laue transmission case. The analysis is performed for a semi-infinite crystal slab, *i.e.* a parallel plate of infinite lateral extent but finite thickness t. Owing to slow convergence of the series expansions for the wave amplitudes, the results are valid for a crystal thickness limited to approximately half the value of the actual extinction lengths.

Following Jackson (1975), Shen (1986) developed a perturbative approach, analogous to the Born approximation from scattering theory in quantum mechanics, for the solution of the inhomogeneous wave equation for the displacement field within an unbounded crystal. The subsequent analysis of a three-beam situation yielded an expression for the diffracted intensity that diverged at the center of the three-beam profile. This approach has later been considerably elaborated upon (Shen, 1999*b*, 2000; Shen & Huang, 2001) through the expanded distorted-wave approximation (EDWA), especially suited to analyze reference-beam diffraction experiments (Shen, 1998, 1999*a*). Within that extended framework, analytical results are found to be in agreement with numerical solutions of the fundamental equations for the entire range of excitation of a secondary wave. However, for a valid approximation, the effective *Pendellösung* thickness associated with the distorting wave is restricted to be less than one.

The purpose of this work is to show that a modification of the original procedure outlined by Shen in fact reproduces the results from the Takagi–Taupin formalism and the thin-crystal limit of EDWA for three-beam Laue–Laue diffraction when crystal finiteness is taken into account. Anomalous-dispersion corrections are assumed to be negligible. Thus, Friedel's law applies and the influence of absorption is not treated. To include first-order *Aufhellung* and primary-extinction effects, the perturbative scheme has to be extended to third order.

The theory has a general part, §2.1, that briefly discusses the different approaches to multibeam X-ray diffraction. The procedure for calculation, restricted to a three-beam case, closely follows the work by Shen (1986) and is outlined in §§2.2 and 2.3. The effect of having a semi-infinite crystal is analyzed using the Fourier transform of the crystal shape function, §2.4. The main results are summarized in §2.6 using definitions given in §2.5.

2. Theory

2.1. General

Maxwell's wave equation for the displacement field $\mathbf{D}(\mathbf{r}, t)$ in a dielectric medium is (Authier, 2001)

$$\nabla \times \nabla \times \mathbf{D} + \frac{1}{c^2}\frac{\partial^2 \mathbf{D}}{\partial t^2} = \nabla \times \nabla \times (\chi_e \mathbf{D}), \qquad (1)$$

where $\chi_e(\mathbf{r})$, the electrical susceptiblity, is a periodic function in the crystal lattice and expressed by the Fourier expansion

$$\chi_e(\mathbf{r}) = \chi_\mathbf{o} + \chi_e'(\mathbf{r}) = \chi_\mathbf{o} + \sum_{\mathbf{p}\neq\mathbf{o}} \chi_\mathbf{p} \exp(-2\pi i \mathbf{p}\cdot\mathbf{r}). \qquad (2)$$

The Fourier amplitudes are given by

$$\chi_\mathbf{p} = -\frac{r_e \lambda^2}{\pi V_c} F_\mathbf{p}. \qquad (3)$$

Here r_e is the classical electron radius, λ the X-ray wavelength, V_c the unit-cell volume and $F_\mathbf{p}$ the structure factor associated with lattice node $\mathbf{p}$.

Within the fundamental theory of X-ray diffraction, the displacement vector for the case of n beams is given by the wavefield (Weckert & Hümmer, 1997):

$$\mathbf{D}(\mathbf{r}, t) = \sum_{j=1}^{4n} c_j \sum_\mathbf{p} \mathbf{D}_{j\mathbf{p}} \exp[2\pi i(\nu t - \mathbf{k}_{j\mathbf{p}}\cdot\mathbf{r})], \qquad (4)$$

where the wavevectors, $\mathbf{k}_{j\mathbf{p}}$, and corresponding amplitudes, $\mathbf{D}_{j\mathbf{p}}$, are obtained from the eigenvalue equations:

$$(k_\mathbf{o}^2 - \mathbf{k}_\mathbf{p}^2)\mathbf{D}_\mathbf{p} + \sum_{\mathbf{q}\neq\mathbf{p}} \chi_{\mathbf{p}-\mathbf{q}}[\mathbf{k}_\mathbf{p}^2 \mathbf{D}_\mathbf{q} - (\mathbf{k}_\mathbf{p}\cdot\mathbf{D}_\mathbf{q})\mathbf{k}_\mathbf{p}] = 0, \qquad (5)$$

where $k_\mathbf{o} = |\mathbf{k}_\mathbf{o}|$, while the coefficients, c_j, are determined by means of the boundary conditions. j counts the number of linearly independent solutions, corresponding to the number of tie points associated with the dispersion surface.

In the Takagi–Taupin approach to X-ray diffraction in a perfect crystal, the displacement vector is expressed by:

$$\mathbf{D}(\mathbf{r}, t) = \sum_{j=1}^{2} \sum_\mathbf{p} \mathbf{D}_{j\mathbf{p}}(\mathbf{r}) \exp[2\pi i(\nu t - \mathbf{k}_\mathbf{p}\cdot\mathbf{r})]. \qquad (6)$$

The spatial varying amplitudes, $\mathbf{D}_{j\mathbf{p}}(\mathbf{r})$, are obtained from the Takagi–Taupin equations (Takagi, 1969; Thorkildsen & Larsen, 1998):

$$(i/\pi)(\mathbf{k}_\mathbf{p}\cdot\nabla)\mathbf{D}_\mathbf{p} = (k_\mathbf{o}^2 - \mathbf{k}_\mathbf{p}^2)\mathbf{D}_\mathbf{p} + \sum_{\mathbf{q}\neq\mathbf{p}} \chi_{\mathbf{p}-\mathbf{q}}[\mathbf{k}_\mathbf{p}^2 \mathbf{D}_\mathbf{q} - (\mathbf{k}_\mathbf{p}\cdot\mathbf{D}_\mathbf{q})\mathbf{k}_\mathbf{p}], \qquad (7)$$

subject to standard boundary conditions (Authier, 2001). The wavevectors are constants, determined by the average wavenumber:

$$k_\mathbf{o} = K(1 + \tfrac{1}{2}\chi_\mathbf{o}) \qquad (8)$$

and the deviation parameters (Authier *et al.*, 1968):

$$\beta_\mathbf{p} = |\mathbf{k}_\mathbf{p}| - k_\mathbf{o}, \qquad (9)$$

$K = 1/\lambda = \nu/c$ being the wavenumber in vacuum. Here j enumerates the two polarization states.

In the approach devised by Shen, the displacement field is written

$$\mathbf{D}(\mathbf{r}, t) = \mathbf{D}(\mathbf{r}) \exp(2\pi i \nu t). \qquad (10)$$

By explicitly taking into account the average contribution to the electric susceptibility, $\chi_\mathbf{o}$, *i.e.* by implementing the DWA approximation (Daillant & Gibaud, 1999), *cf.* Fig. 1 of Shen (1999*b*), equation (1) may be written:

$$\nabla^2 \mathbf{D} + 4\pi^2 k_\mathbf{o}^2 \mathbf{D} = -\nabla \times \nabla \times (\chi_e' \mathbf{D}). \qquad (11)$$

The formal solution of this equation, given by the combination of a particular solution and a solution of the homogeneous equation, is

$$\mathbf{D}(\mathbf{r}) = \mathbf{D}_0 \exp(-2\pi i \mathbf{k}_\mathbf{o}\cdot\mathbf{r}) + \frac{1}{4\pi}\int_V \mathrm{d}^3 r' \, \frac{\exp(-2\pi i k_\mathbf{o}|\mathbf{r}-\mathbf{r}'|)}{|\mathbf{r}-\mathbf{r}'|} \times \nabla' \times \nabla' \times [\chi_e'(\mathbf{r}')\mathbf{D}(\mathbf{r}')]. \qquad (12)$$

The result of applying proper boundary conditions will be closely related to the Ewald–Oseen extinction theorem (Ewald, 1916; Oseen, 1915; Born & Wolf, 1997) and should be addressed separately. Suffice here to say that $\mathbf{D}_0$ is interpreted as the amplitude of the incoming wave. The integration subscript, V, indicates that the integration covers the volume of the crystal. The particular solution is obtained by the Green-function method. The Green function, $G(\mathbf{r}, \mathbf{r}')$, is the solution of the equation

$$\nabla^2 G(\mathbf{r}, \mathbf{r}') + 4\pi^2 k_\mathbf{o}^2 G(\mathbf{r}, \mathbf{r}') = -\delta(\mathbf{r} - \mathbf{r}').$$

This equation is solved by Fourier methods (Roman, 1965; Merzbacher, 1970) leading to the outgoing spherical wave:

$$G_{+\epsilon}(\mathbf{r}, \mathbf{r}') = \frac{1}{4\pi}\frac{\exp(-2\pi i k_\mathbf{o}|\mathbf{r}-\mathbf{r}'|)}{|\mathbf{r}-\mathbf{r}'|}.$$

The subscript $+\epsilon$ indicates that the spherical wave is linked to the plane-wave expansion:

$$\frac{\exp(-2\pi i k_\mathbf{o}|\mathbf{r}-\mathbf{r}'|)}{|\mathbf{r}-\mathbf{r}'|} = \lim_{\epsilon\to 0}\frac{1}{\pi}\int \mathrm{d}^3 k \, \frac{\exp[-2\pi i \mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')]}{k^2 - k_\mathbf{o}^2 + i\epsilon}, \qquad (13)$$

showing explicitly how to avoid singularities in the integrand along the real k axis. As will be apparent later on, *cf.* equation (27), this is a point of great importance.

2.2. Procedure for calculation

We associate a counting variable, ε, with the smallness quantity χ_e' in equation (12), and express the amplitude function $\mathbf{D}(\mathbf{r})$ by the series expansion

$$\mathbf{D}(\mathbf{r}) = \sum_{n=0}^{\infty} \varepsilon^n \mathbf{D}^{(n)}(\mathbf{r}). \qquad (14)$$

In the final expressions, $\varepsilon \to 1$. By inserting (14) into (12) and equating equal powers in ε, we obtain the recurrence relations:

$$\mathbf{D}^{(n)}(\mathbf{r}) = \frac{1}{4\pi}\int \mathrm{d}^3 r' \, \frac{\exp(-2\pi i k_\mathbf{o}|\mathbf{r}-\mathbf{r}'|)}{|\mathbf{r}-\mathbf{r}'|} \times \nabla' \times \nabla' \times [\chi_e'(\mathbf{r}')\mathbf{D}^{(n-1)}(\mathbf{r}')], \quad n \geq 1,$$

with

$$\mathbf{D}^{(0)}(\mathbf{r}) = \mathbf{D}_0 \exp(-2\pi i \mathbf{k}_\mathbf{o}\cdot\mathbf{r}).$$

In the string of volume integrations that will occur in the calculation of the nth-order term, the spherical wave of the

outermost integration, the radiation part, is treated in the far-field limit:

$$\frac{\exp(-2\pi i k_{\mathbf{o}}|\mathbf{r}-\mathbf{r}'|)}{|\mathbf{r}-\mathbf{r}'|} \approx \frac{\exp(-2\pi i k_{\mathbf{o}} r)}{r}\exp(2\pi i k_{\mathbf{o}}\hat{\mathbf{n}}\cdot\mathbf{r}'),$$

where we have introduced a unit vector $\hat{\mathbf{n}} = \mathbf{r}/r$ in the direction of observation, r being the crystal–detector distance. The other (internal) spherical waves are all processed using the Fourier expansion of (13). The generic integral, $I_{\boldsymbol{\psi}}$, associated with a vector function $\boldsymbol{\psi}(\mathbf{r})$, is evaluated according to

$$\begin{aligned} I_{\boldsymbol{\psi}} &= \int_V \mathrm{d}^3 r\, \exp(2\pi i\mathbf{k}\cdot\mathbf{r})\nabla\times\nabla\times\boldsymbol{\psi}(\mathbf{r}) \\ &= -(2\pi)^2\mathbf{k}\times\mathbf{k}\times\int_V \mathrm{d}^3 r\, \exp(2\pi i\mathbf{k}\cdot\mathbf{r})\boldsymbol{\psi}(\mathbf{r}). \end{aligned}$$

This result is obtained after integration by parts using the divergence theorem and by assuming vanishing contributions from the associated surface integrals. It follows that $\mathbf{D}^{(1)}(\mathbf{r})$ may be calculated from

$$\begin{aligned} \mathbf{D}^{(1)}(\mathbf{r}) = -\pi[\exp(-2\pi i k_{\mathbf{o}} r)/r]k_{\mathbf{o}}^2\,\hat{\mathbf{n}}\times\hat{\mathbf{n}}\times\mathbf{D}_0 \\ \times\int_V \mathrm{d}^3 r'\,\chi_e'(\mathbf{r}')\exp[2\pi i(k_{\mathbf{o}}\hat{\mathbf{n}}-\mathbf{k}_{\mathbf{o}})\cdot\mathbf{r}'], \end{aligned} \tag{15}$$

while

$$\begin{aligned} \mathbf{D}^{(2)}(\mathbf{r}) = \pi[\exp(-2\pi i k_{\mathbf{o}} r)/r]k_{\mathbf{o}}^2\,\hat{\mathbf{n}}\times\hat{\mathbf{n}} \\ \times\int \mathrm{d}^3 k'\,[1/(k'^2-k_{\mathbf{o}}^2)]\mathbf{k}'\times\mathbf{k}'\times\mathbf{D}_0 \\ \times\int_V \mathrm{d}^3 r'\,\chi_e'(\mathbf{r}')\exp[2\pi i(k_{\mathbf{o}}\hat{\mathbf{n}}-\mathbf{k}')\cdot\mathbf{r}'] \\ \times\int_V \mathrm{d}^3 r''\,\chi_e'(\mathbf{r}'')\exp[2\pi i(\mathbf{k}'-\mathbf{k}_{\mathbf{o}})\cdot\mathbf{r}''] \end{aligned} \tag{16}$$

and

$$\begin{aligned} \mathbf{D}^{(3)}(\mathbf{r}) = -\pi[\exp(-2\pi i k_{\mathbf{o}} r)/r]\,k_{\mathbf{o}}^2\,\hat{\mathbf{n}}\times\hat{\mathbf{n}} \\ \times\int \mathrm{d}^3 k'\,[1/(k'^2-k_{\mathbf{o}}^2)]\int \mathrm{d}^3 k''\,[1/(k''^2-k_{\mathbf{o}}^2)] \\ \times\mathbf{k}'\times\mathbf{k}'\times\mathbf{k}''\times\mathbf{k}''\times\mathbf{D}_0 \\ \times\int_V \mathrm{d}^3 r'\,\chi_e'(\mathbf{r}')\exp[2\pi i(k_{\mathbf{o}}\hat{\mathbf{n}}-\mathbf{k}')\cdot\mathbf{r}'] \\ \times\int_V \mathrm{d}^3 r''\,\chi_e'(\mathbf{r}'')\exp[2\pi i(\mathbf{k}'-\mathbf{k}'')\cdot\mathbf{r}''] \\ \times\int_V \mathrm{d}^3 r'''\,\chi_e'(\mathbf{r}''')\exp[2\pi i(\mathbf{k}''-\mathbf{k}_{\mathbf{o}})\cdot\mathbf{r}''']. \end{aligned} \tag{17}$$

In (15)–(17) and all subsequent equations, multiple vector cross products are evaluated according to:

$$\begin{aligned} &\mathbf{A}_1\times\mathbf{A}_2\times\ldots\times\mathbf{A}_{n-2}\times\mathbf{A}_{n-1}\times\mathbf{A}_n \\ &\quad = (\mathbf{A}_1\times(\mathbf{A}_2\times(\ldots\times(\mathbf{A}_{n-2}\times(\mathbf{A}_{n-1}\times\mathbf{A}_n))\ldots))). \end{aligned}$$

2.3. Three-beam diffraction

We start by considering the first-order term. Substituting the Fourier expansion of the susceptibility, equation (2), into equation (15) and using equation (19), we have:

$$\mathbf{D}^{(1)}(\mathbf{r}) = -\pi[\exp(-2\pi i k_{\mathbf{o}} r)/r]\sum_{\mathbf{p}\neq\mathbf{o}}\chi_{\mathbf{p}}k_{\mathbf{o}}^2\,\hat{\mathbf{n}}\times\hat{\mathbf{n}}\times\mathbf{D}_0\mathcal{S}(\Delta\mathbf{k}_{\mathbf{p}}). \tag{18}$$

The function $\mathcal{S}(\mathbf{K})$ is the Fourier transform of the crystal shape function, $S(\mathbf{r})$:

$$\mathcal{S}(\mathbf{K}) = \int \mathrm{d}^3 r\, S(\mathbf{r})\exp(2\pi i\mathbf{K}\cdot\mathbf{r}), \tag{19}$$

where the volume integration in principle spans the whole space, since $S(\mathbf{r})\equiv 0$ for $\mathbf{r}$ outside the crystal boundaries, $S(\mathbf{r})\equiv 1$ otherwise. In the limit of an infinite crystal, $\mathcal{S}(\mathbf{K})$ converges weakly towards $\delta(\mathbf{K})$; a Dirac δ function. In (18), the characteristic mathematical properties of $\mathcal{S}$ have been utilized by defining the deviation parameter $\Delta\mathbf{k}_{\mathbf{p}}$, $|\Delta\mathbf{k}_{\mathbf{p}}| \ll K$:

$$\Delta\mathbf{k}_{\mathbf{p}} = k_{\mathbf{o}}\hat{\mathbf{n}} - \mathbf{k}_{\mathbf{o}} - \mathbf{p}.$$

Diffraction will be limited to small volume elements in reciprocal space associated with lattice nodes in the vicinity of the Ewald sphere. Three-beam diffraction is of special importance owing to its potential to carry phase information related to the triplet structure invariant Φ_Σ. The wavevectors involved are $\mathbf{k}_{\mathbf{o}}$, $\mathbf{k}_{\mathbf{h}}$ and $\mathbf{k}_{\mathbf{g}}$, linked by the reciprocal-lattice vectors $\pm\mathbf{h}$, $\pm\mathbf{g}$ and $\pm(\mathbf{h}-\mathbf{g})$. To determine the direction of the vector $\mathbf{D}^{(1)}$, we use the approximation

$$\mathbf{k}_{\mathbf{p}} = \mathbf{k}_{\mathbf{o}} + \mathbf{p} \approx K\,\hat{\mathbf{s}}_{\mathbf{p}}, \qquad \mathbf{p}\in\{\mathbf{o},\mathbf{h},\mathbf{g}\},$$

with $\hat{\mathbf{s}}_{\mathbf{p}}$ being a unit vector. $\mathbf{k}_{\mathbf{h}}$ is regarded as the wavevector of the *primary* beam and the direction of observation is set to

$$k_{\mathbf{o}}\hat{\mathbf{n}} = \mathbf{k}_{\mathbf{o}} + \mathbf{h} + \Delta\mathbf{k}_{\mathbf{h}} \approx K\hat{\mathbf{s}}_{\mathbf{h}}.$$

Thus,

$$\Delta\mathbf{k}_{\mathbf{p}} = \Delta\mathbf{k}_{\mathbf{h}} + (\mathbf{h}-\mathbf{p})$$

and the property of $\mathcal{S}$ causes a single term, $\mathbf{p}=\mathbf{h}$, in (18) to contribute. Using (3), we express the first-order contribution to the primary diffracted wave[1] indicated by the subscript $\mathbf{h}$:

$$\mathbf{D}_{\mathbf{h}}^{(1)}(\mathbf{r}) = \frac{\exp(-2\pi i k_{\mathbf{o}} r)}{r}\left(\frac{r_e}{V_c}\right)(\hat{\mathbf{s}}_{\mathbf{h}}\times\hat{\mathbf{s}}_{\mathbf{h}}\times\mathbf{D}_0)F_{\mathbf{h}}\mathcal{S}(\Delta\mathbf{k}_{\mathbf{h}}). \tag{20}$$

Equation (20) should be compared with equation (14) of Shen (1986). Although $\mathcal{S}\propto V$ for a finite crystal, the procedure by Shen does not properly take into account the spreading of the reciprocal-lattice points.

By the same arguments, we obtain the contributions to second and third order:

$$\begin{aligned} \mathbf{D}_{\mathbf{h}}^{(2)}(\mathbf{r}) = [\exp(-2\pi i k_{\mathbf{o}} r)/r]\left(\frac{r_e}{V_c}\right)^2(\hat{\mathbf{s}}_{\mathbf{h}}\times\hat{\mathbf{s}}_{\mathbf{h}}\times\hat{\mathbf{s}}_{\mathbf{g}}\times\hat{\mathbf{s}}_{\mathbf{g}}\times\mathbf{D}_0) \\ \times F_{\mathbf{h}-\mathbf{g}}F_{\mathbf{g}}\frac{1}{\pi}\int \mathrm{d}^3\Delta k'\,\frac{1}{|\mathbf{k}_{\mathbf{g}}+\Delta\mathbf{k}'|^2-|\mathbf{k}_{\mathbf{o}}|^2} \\ \times\mathcal{S}(\Delta\mathbf{k}_{\mathbf{h}}-\Delta\mathbf{k}')\mathcal{S}(\Delta\mathbf{k}'), \end{aligned} \tag{21}$$

[1] The zeroth-order term does not contribute to the wave in the $\mathbf{k}_{\mathbf{h}}$ direction.

$$\begin{aligned}
\mathbf{D}_{\mathbf{h}}^{(3)}(\mathbf{r}) = {} & [\exp(-2\pi i k_{\mathbf{o}} r)/r]\left(\frac{r_e}{V_c}\right)^3 \Bigg\{\Bigg[(\hat{\mathbf{s}}_{\mathbf{h}} \times \hat{\mathbf{s}}_{\mathbf{h}} \times \hat{\mathbf{s}}_{\mathbf{o}} \times \hat{\mathbf{s}}_{\mathbf{o}} \times \hat{\mathbf{s}}_{\mathbf{h}} \\
& \times \hat{\mathbf{s}}_{\mathbf{h}} \times \mathbf{D}_0) F_{\mathbf{h}} F_{-\mathbf{h}} F_{\mathbf{h}} \frac{1}{\pi^2} \int \mathrm{d}^3\Delta k' \, \frac{1}{|\mathbf{k}_{\mathbf{o}} + \Delta\mathbf{k}'|^2 - |\mathbf{k}_{\mathbf{o}}|^2} \\
& \times \int \mathrm{d}^3\Delta k'' \, \frac{1}{|\mathbf{k}_{\mathbf{h}} + \Delta\mathbf{k}''|^2 - |\mathbf{k}_{\mathbf{o}}|^2} \\
& \times \mathcal{S}(\Delta\mathbf{k}_{\mathbf{h}} - \Delta\mathbf{k}')\mathcal{S}(\Delta\mathbf{k}' - \Delta\mathbf{k}'')\mathcal{S}(\Delta\mathbf{k}'')\Bigg] \\
& + \Bigg[(\hat{\mathbf{s}}_{\mathbf{h}} \times \hat{\mathbf{s}}_{\mathbf{h}} \times \hat{\mathbf{s}}_{\mathbf{o}} \times \hat{\mathbf{s}}_{\mathbf{o}} \times \hat{\mathbf{s}}_{\mathbf{g}} \times \hat{\mathbf{s}}_{\mathbf{g}} \times \mathbf{D}_0) F_{\mathbf{h}} F_{-\mathbf{g}} F_{\mathbf{g}} \\
& \times \frac{1}{\pi^2} \int \mathrm{d}^3\Delta k' \, \frac{1}{|\mathbf{k}_{\mathbf{o}} + \Delta\mathbf{k}'|^2 - |\mathbf{k}_{\mathbf{o}}|^2} \\
& \times \int \mathrm{d}^3\Delta k'' \, \frac{1}{|\mathbf{k}_{\mathbf{g}} + \Delta\mathbf{k}''|^2 - |\mathbf{k}_{\mathbf{o}}|^2} \\
& \times \mathcal{S}(\Delta\mathbf{k}_{\mathbf{h}} - \Delta\mathbf{k}')\mathcal{S}(\Delta\mathbf{k}' - \Delta\mathbf{k}'')\mathcal{S}(\Delta\mathbf{k}'')\Bigg] \\
& + \Bigg[(\hat{\mathbf{s}}_{\mathbf{h}} \times \hat{\mathbf{s}}_{\mathbf{h}} \times \hat{\mathbf{s}}_{\mathbf{g}} \times \hat{\mathbf{s}}_{\mathbf{g}} \times \hat{\mathbf{s}}_{\mathbf{h}} \times \hat{\mathbf{s}}_{\mathbf{h}} \times \mathbf{D}_0) F_{\mathbf{h}-\mathbf{g}} F_{\mathbf{g}-\mathbf{h}} F_{\mathbf{h}} \\
& \times \frac{1}{\pi^2} \int \mathrm{d}^3\Delta k' \, \frac{1}{|\mathbf{k}_{\mathbf{g}} + \Delta\mathbf{k}'|^2 - |\mathbf{k}_{\mathbf{o}}|^2} \\
& \times \int \mathrm{d}^3\Delta k'' \, \frac{1}{|\mathbf{k}_{\mathbf{h}} + \Delta\mathbf{k}''|^2 - |\mathbf{k}_{\mathbf{o}}|^2} \\
& \times \mathcal{S}(\Delta\mathbf{k}_{\mathbf{h}} - \Delta\mathbf{k}')\mathcal{S}(\Delta\mathbf{k}' - \Delta\mathbf{k}'')\mathcal{S}(\Delta\mathbf{k}'')\Bigg]\Bigg\}. \qquad (22)
\end{aligned}$$

The factors $1/(|\mathbf{k}_{\mathbf{p}} + \Delta\mathbf{k}'|^2 - |\mathbf{k}_{\mathbf{o}}|^2)$ are denoted *resonance* terms.

2.4. A semi-infinite crystal

The Fourier transform of the shape function for a semi-infinite crystal of thickness t becomes

$$\begin{aligned}
S(\mathbf{K}) &= \int_0^t \mathrm{d}z \int_{-\infty}^{\infty} \mathrm{d}y \int_{-\infty}^{\infty} \mathrm{d}x \exp[2\pi i(K_x x + K_y y + K_z z)] \\
&= \delta(K_x)\delta(K_y)\exp(\pi i K_z t)\sin(\pi K_z t)/\pi K_z.
\end{aligned}$$

By introducing the dimensionless variable u,

$$u = \pi\Delta k_{\mathbf{h}z} t,$$

we have:

$$\mathcal{S}(\Delta\mathbf{k}_{\mathbf{h}}) = \delta(\Delta k_{\mathbf{h}x})\delta(\Delta k_{\mathbf{h}y})\exp(iu)t\,(\sin u)/u. \qquad (23)$$

Integrals of the type encountered in the second-order term will be evaluated according to

$$\begin{aligned}
& \frac{1}{\pi}\int \mathrm{d}^3\Delta k' \, \frac{1}{|\mathbf{k}_{\mathbf{p}} + \Delta\mathbf{k}'|^2 - |\mathbf{k}_{\mathbf{o}}|^2}\mathcal{S}(\Delta\mathbf{k}_{\mathbf{h}} - \Delta\mathbf{k}')\mathcal{S}(\Delta\mathbf{k}') \\
& \quad = (t^2/\pi)\delta(\Delta k_{\mathbf{h}x})\delta(\Delta k_{\mathbf{h}y})\exp(\pi i \Delta k_{\mathbf{h}z} t) \\
& \quad \times \int_{-\infty}^{\infty} \mathrm{d}\Delta k'_z \, \frac{1}{|\mathbf{k}_{\mathbf{p}} + \Delta k'_z \hat{\mathbf{k}}|^2 - |\mathbf{k}_{\mathbf{o}}|^2} \\
& \quad \times \frac{\sin(\pi\Delta k'_z t)}{\pi\Delta k'_z t}\frac{\sin[\pi(\Delta k_{\mathbf{h}z} - \Delta k'_z)t]}{\pi(\Delta k_{\mathbf{h}z} - \Delta k'_z)t}.
\end{aligned}$$

Here $\hat{\mathbf{k}}$ is a unit vector in the z direction, *i.e.* along the inward drawn normal vector to the entrance surface of the crystal. The denominator in the resonance term is linearized by

$$|\mathbf{k}_{\mathbf{p}} + \Delta k'_z \hat{\mathbf{k}}|^2 - |\mathbf{k}_{\mathbf{o}}|^2 \approx 2(\mathbf{k}_{\mathbf{p}} \cdot \hat{\mathbf{k}})\left[\Delta k'_z + \frac{K\beta_{\mathbf{p}}}{(\mathbf{k}_{\mathbf{p}} \cdot \hat{\mathbf{k}})}\right], \qquad (24)$$

where we have used the deviation parameter $\beta_{\mathbf{p}}$ introduced in (9). To obtain (24), squares of small terms ($\Delta k'_z \sim \beta_{\mathbf{p}} \sim K\chi_0$) have been neglected. This approximation prevents the discussion of grazing-wave diffraction modes. In what follows, we will also limit ourselves to Laue–Laue diffraction, thus $(\mathbf{k}_{\mathbf{p}} \cdot \hat{\mathbf{k}}) \approx K(\hat{\mathbf{s}}_{\mathbf{p}} \cdot \hat{\mathbf{k}}) = K\cos\gamma_{\mathbf{p}} > 0$ for $\mathbf{p} \in \{\mathbf{o}, \mathbf{h}, \mathbf{g}\}$, where $\gamma_{\mathbf{p}}$ is the angle between the direction $\hat{\mathbf{s}}_{\mathbf{p}}$ and the surface normal $\hat{\mathbf{k}}$.

By introducing the following dimensionless variables:

$$\begin{aligned}
z' &= \pi\Delta k'_z t, \\
z'' &= \pi\Delta k''_z t, \\
v_{\mathbf{p}} &= \pi\frac{K\beta_{\mathbf{p}}}{(\mathbf{k}_{\mathbf{p}} \cdot \hat{\mathbf{k}})}t \approx \pi\frac{\beta_{\mathbf{p}}}{\cos\gamma_{\mathbf{p}}}t,
\end{aligned}$$

we obtain

$$\begin{aligned}
& \frac{1}{\pi}\int \mathrm{d}^3\Delta k' \, \frac{1}{|\mathbf{k}_{\mathbf{p}} + \Delta\mathbf{k}'|^2 - |\mathbf{k}_{\mathbf{o}}|^2}\mathcal{S}(\Delta\mathbf{k}_{\mathbf{h}} - \Delta\mathbf{k}')\mathcal{S}(\Delta\mathbf{k}') \\
& \quad = \delta(\Delta k_{\mathbf{h}x})\delta(\Delta k_{\mathbf{h}y})\exp(iu)(t^2/K\cos\gamma_{\mathbf{p}}) \\
& \quad \times \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}z' \, \frac{1}{z' + v_{\mathbf{p}} + i\epsilon'}\frac{\sin(z' - u)}{z' - u}\frac{\sin z'}{z'}. \qquad (25)
\end{aligned}$$

Similarly, the integrations involved in calculating the third-order terms may be reduced to the standard form:

$$\begin{aligned}
& \frac{1}{\pi^2}\int \mathrm{d}^3\Delta k' \, \frac{1}{|\mathbf{k}_{\mathbf{p}} + \Delta\mathbf{k}'|^2 - |\mathbf{k}_{\mathbf{o}}|^2}\int \mathrm{d}^3\Delta k'' \, \frac{1}{|\mathbf{k}_{\mathbf{q}} + \Delta\mathbf{k}''|^2 - |\mathbf{k}_{\mathbf{o}}|^2} \\
& \quad \times S(\Delta\mathbf{k}_{\mathbf{h}} - \Delta\mathbf{k}')S(\Delta\mathbf{k}' - \Delta\mathbf{k}'')S(\Delta\mathbf{k}'') \\
& \quad = \delta(\Delta k_{\mathbf{h}x})\delta(\Delta k_{\mathbf{h}y})\exp(iu)(t^3/K^2\cos\gamma_{\mathbf{p}}\cos\gamma_{\mathbf{q}}) \\
& \quad \times \frac{1}{4\pi^2}\int_{-\infty}^{\infty} \mathrm{d}z' \, \frac{1}{z' + v_{\mathbf{p}} + i\epsilon'}\frac{\sin(z' - u)}{z' - u} \\
& \quad \times \int_{-\infty}^{\infty} \mathrm{d}z'' \, \frac{1}{z'' + v_{\mathbf{q}} + i\epsilon''}\frac{\sin(z'' - z')}{z'' - z'}\frac{\sin z''}{z''}. \qquad (26)
\end{aligned}$$

In (25) and (26), we have explicitly indicated the smallness parameters ϵ', ϵ'' introduced to obtain the correct path of the complex integrations (limits $\epsilon', \epsilon'' \to 0$ are implied).

2.5. Definitions

We define a set of functions:[2]

$$g^{(1)}(u) = (\sin u)/u$$

and

$$g^{(2)}_{\Re}(u, v_{\mathbf{g}}) - i g^{(2)}_{\Im}(u, v_{\mathbf{g}}) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}z' \, \frac{1}{z' + v_{\mathbf{g}}}\frac{\sin(z' - u)}{z' - u}\frac{\sin z'}{z'}.$$

For the third-order contributions, we impose the conditions $v_{\mathbf{o}} = 0$ and $v_{\mathbf{h}} = -u$. The former follows from the definition of

[2] ϵ', ϵ'' are not explicitly denoted in this section.

$\beta_{\mathbf{p}}$, equation (9), and the latter one from equation (24) evaluated at exact resonance. We get:

$$g^{(3)}_{1,\Re}(u) - ig^{(3)}_{1,\Im}(u) = \frac{1}{4\pi^2}\int_{-\infty}^{\infty} \mathrm{d}z' \, \frac{1}{z'}\frac{\sin(z'-u)}{z'-u}\int_{-\infty}^{\infty} \mathrm{d}z'' \, \frac{1}{z''-u}\frac{\sin(z''-z')}{z''-z'}\frac{\sin z''}{z''},$$

$$g^{(3)}_{2,\Re}(u, v_{\mathbf{g}}) - ig^{(3)}_{2,\Im}(u, v_{\mathbf{g}}) = \frac{1}{4\pi^2}\int_{-\infty}^{\infty} \mathrm{d}z' \, \frac{1}{z'}\frac{\sin(z'-u)}{z'-u}\int_{-\infty}^{\infty} \mathrm{d}z'' \, \frac{1}{z''+v_{\mathbf{g}}}\frac{\sin(z''-z')}{z''-z'}\frac{\sin z''}{z''},$$

$$g^{(3)}_{3,\Re}(u, v_{\mathbf{g}}) - ig^{(3)}_{3,\Im}(u, v_{\mathbf{g}}) = \frac{1}{4\pi^2}\int_{-\infty}^{\infty} \mathrm{d}z' \, \frac{1}{z'+v_{\mathbf{g}}}\frac{\sin(z'-u)}{z'-u}\int_{-\infty}^{\infty} \mathrm{d}z'' \, \frac{1}{z''-u}\frac{\sin(z''-z')}{z''-z'}\frac{\sin z''}{z''}.$$

The key result used for evaluation of these integrals is the following operator equation given by Roman (1965):

$$\lim_{\epsilon\to 0}\frac{1}{z+v+i\epsilon} = P\frac{1}{z+v} - i\pi\delta(z+v). \qquad (27)$$

P indicates the Cauchy principal value. The results of the integrations, performed using the residue theorem, are summarized in Appendix A.

We introduce polarization factors $\mathbf{p}^{(i)}_j$ by the definitions:

$$\begin{aligned}
|\mathbf{D}_0|\mathbf{p}^{(1)} &= -(\hat{\mathbf{s}}_{\mathbf{h}}\times\hat{\mathbf{s}}_{\mathbf{h}}\times\mathbf{D}_0),\\
|\mathbf{D}_0|\mathbf{p}^{(2)} &= \quad(\hat{\mathbf{s}}_{\mathbf{h}}\times\hat{\mathbf{s}}_{\mathbf{h}}\times\hat{\mathbf{s}}_{\mathbf{g}}\times\hat{\mathbf{s}}_{\mathbf{g}}\times\mathbf{D}_0),\\
|\mathbf{D}_0|\mathbf{p}^{(3)}_1 &= -(\hat{\mathbf{s}}_{\mathbf{h}}\times\hat{\mathbf{s}}_{\mathbf{h}}\times\hat{\mathbf{s}}_{\mathbf{o}}\times\hat{\mathbf{s}}_{\mathbf{o}}\times\hat{\mathbf{s}}_{\mathbf{h}}\times\hat{\mathbf{s}}_{\mathbf{h}}\times\mathbf{D}_0),\\
|\mathbf{D}_0|\mathbf{p}^{(3)}_2 &= -(\hat{\mathbf{s}}_{\mathbf{h}}\times\hat{\mathbf{s}}_{\mathbf{h}}\times\hat{\mathbf{s}}_{\mathbf{o}}\times\hat{\mathbf{s}}_{\mathbf{o}}\times\hat{\mathbf{s}}_{\mathbf{g}}\times\hat{\mathbf{s}}_{\mathbf{g}}\times\mathbf{D}_0),\\
|\mathbf{D}_0|\mathbf{p}^{(3)}_3 &= -(\hat{\mathbf{s}}_{\mathbf{h}}\times\hat{\mathbf{s}}_{\mathbf{h}}\times\hat{\mathbf{s}}_{\mathbf{g}}\times\hat{\mathbf{s}}_{\mathbf{g}}\times\hat{\mathbf{s}}_{\mathbf{h}}\times\hat{\mathbf{s}}_{\mathbf{h}}\times\mathbf{D}_0).
\end{aligned}$$

The choice of signs causes the term $\hat{\mathbf{d}}_0 = \mathbf{D}_0/|\mathbf{D}_0|$ to enter with a + sign for all factors, *e.g.*

$$\begin{aligned}
\mathbf{p}^{(1)} &= \hat{\mathbf{d}}_0 - \hat{\mathbf{s}}_{\mathbf{h}}(\hat{\mathbf{s}}_{\mathbf{h}}\cdot\hat{\mathbf{d}}_0),\\
\mathbf{p}^{(2)} &= \hat{\mathbf{d}}_0 - \hat{\mathbf{s}}_{\mathbf{h}}(\hat{\mathbf{s}}_{\mathbf{h}}\cdot\hat{\mathbf{d}}_0) - \hat{\mathbf{s}}_{\mathbf{g}}(\hat{\mathbf{s}}_{\mathbf{g}}\cdot\hat{\mathbf{d}}_0) + \hat{\mathbf{s}}_{\mathbf{h}}(\hat{\mathbf{s}}_{\mathbf{h}}\cdot\hat{\mathbf{s}}_{\mathbf{g}})(\hat{\mathbf{s}}_{\mathbf{g}}\cdot\hat{\mathbf{d}}_0),\\
&\vdots
\end{aligned}$$

Furthermore, we use dimensionless parameters $\eta_{\mathbf{pq}}$,

$$\eta_{\mathbf{pq}} = (\cos\gamma_{\mathbf{p}}\cos\gamma_{\mathbf{q}})^{-1/2}(t\lambda\, r_e/V_c)F_{\mathbf{p-q}},$$

and explicitly introduce the phase of the triplet structure invariant, $\Phi_\Sigma = \varphi_{-\mathbf{h}} + \varphi_{\mathbf{h-g}} + \varphi_{\mathbf{g}}$, in the calculation by

$$\eta_{\mathbf{hg}}\eta_{\mathbf{go}}/\eta_{\mathbf{ho}} = \Gamma\exp(i\Phi_\Sigma).$$

Γ, the modulus of the left-hand side, is thus proportional to $|F_{\mathbf{h-g}}||F_{\mathbf{g}}|/|F_{\mathbf{h}}|$.

2.6. Diffracted intensity and integrated power

Using the definitions of §2.5, we arrive at the final expression for the amplitude of the primary diffracted wave, valid to third order:[3]

$$\begin{aligned}
\mathbf{D}_{\mathbf{h}}(\mathbf{r}) = &-[\exp(-2\pi i k_{\mathbf{o}} r)/r]\delta(\Delta k_{\mathbf{h}x})\delta(\Delta k_{\mathbf{h}y})\exp(iu)\\
&\times t(r_e/V_c)F_{\mathbf{h}}|\mathbf{D}_0|\{\mathbf{p}^{(1)}g^{(1)}(u)\\
&- \mathbf{p}^{(2)}\Gamma(\cos\Phi_\Sigma + i\sin\Phi_\Sigma)[g^{(2)}_{\Re}(u, v_{\mathbf{g}}) - ig^{(2)}_{\Im}(u, v_{\mathbf{g}})]\\
&+ \mathbf{p}^{(3)}_1|\eta_{\mathbf{oh}}|^2 g^{(3)}_{1,\Re}(u)\\
&+ \mathbf{p}^{(3)}_2|\eta_{\mathbf{og}}|^2[g^{(3)}_{2,\Re}(u, v_{\mathbf{g}}) - ig^{(3)}_{2,\Im}(u, v_{\mathbf{g}})]\\
&+ \mathbf{p}^{(3)}_3|\eta_{\mathbf{hg}}|^2[g^{(3)}_{3,\Re}(u, v_{\mathbf{g}}) - ig^{(3)}_{3,\Im}(u, v_{\mathbf{g}})]\}. \qquad (28)
\end{aligned}$$

The corresponding diffracted intensity is given by

$$I_{\mathbf{h}} = (c/2\varepsilon_0)\mathbf{D}_{\mathbf{h}}(\mathbf{r})\cdot\mathbf{D}^*_{\mathbf{h}}(\mathbf{r}),$$

and we obtain:

$$\begin{aligned}
I_{\mathbf{h}} = &I_0(1/r^2)(r_e t/V_c)^2|F_{\mathbf{h}}|^2\delta^2(\Delta k_{\mathbf{h}x})\delta^2(\Delta k_{\mathbf{h}y})\{(\mathbf{p}^{(1)}\cdot\mathbf{p}^{(1)})h^{(0)}(u)\\
&+ 2(\mathbf{p}^{(1)}\cdot\mathbf{p}^{(3)}_1)|\eta_{\mathbf{oh}}|^2 h^{(2)}_1(u) - 2(\mathbf{p}^{(1)}\cdot\mathbf{p}^{(2)})\\
&\times\Gamma[h^{(1)}_1(u, v_{\mathbf{g}})\cos\Phi_\Sigma + h^{(1)}_2(u, v_{\mathbf{g}})\sin\Phi_\Sigma]\\
&+ 2(\mathbf{p}^{(1)}\cdot\mathbf{p}^{(3)}_2)|\eta_{\mathbf{og}}|^2 h^{(2)}_2(u, v_{\mathbf{g}}) + 2(\mathbf{p}^{(1)}\cdot\mathbf{p}^{(3)}_3)|\eta_{\mathbf{hg}}|^2 h^{(2)}_3(u, v_{\mathbf{g}})\\
&+ (\mathbf{p}^{(2)}\cdot\mathbf{p}^{(2)})\Gamma^2 h^{(2)}_4(u, v_{\mathbf{g}})\}. \qquad (29)
\end{aligned}$$

I_0 is the intensity of the incoming X-ray beam. The h functions are summarized in Appendix A. The integrated power is then calculated from (Warren, 1969; Becker & Coppens, 1974):

$$\mathcal{P}_{\mathbf{h}} = \frac{r^2\lambda^3}{\sin 2\theta_{oh}}\int \mathrm{d}^3\Delta k_{\mathbf{h}}\; I_{\mathbf{h}}.$$

θ_{oh} is the Bragg angle of the primary reflection and $1/\sin 2\theta_{oh}$ the Lorentz factor. Special care must be taken in handling the integrations involving $\delta^2(\Delta k_{\mathbf{h}x})$ and $\delta^2(\Delta k_{\mathbf{h}y})$. These δ functions arise from limiting processes of the type

$$\delta(\Delta k_{\mathbf{h}x}) = \lim_{L_x\to\infty} L_x\frac{\sin(\pi\Delta k_{\mathbf{h}x}L_x)}{\pi\Delta k_{\mathbf{h}x}L_x},$$

where L_x (L_y) is a lateral dimension of the crystal. In fact, for a semi-infinite crystal, $L_x\times L_y$ corresponds to the illuminated area, A, of the crystal surface. L_x (L_y) should thus be regarded as a finite quantity, although still large enough to warrant the use of a δ function in $\mathcal{S}$. It follows that:

$$\int_{-\infty}^{\infty}\mathrm{d}\Delta k_{\mathbf{h}x}\,\delta^2(\Delta k_{\mathbf{h}x}) = L_x^2(1/\pi L_x)\int_{-\infty}^{\infty}[(\sin^2 x)/x^2]\,\mathrm{d}x = L_x,$$

giving

$$\int_{-\infty}^{\infty}\mathrm{d}\Delta k_{\mathbf{h}x}\,\delta^2(\Delta k_{\mathbf{h}x})\int_{-\infty}^{\infty}\mathrm{d}\Delta k_{\mathbf{h}y}\,\delta^2(\Delta k_{\mathbf{h}y}) = L_xL_y = A.$$

We then obtain our final expression for the integrated primary diffracted power in the vicinity of a three-beam point:[4]

[3] We have applied the result $g^{(3)}_{1,\Im}(u) \equiv 0$.

[4] The variable $2v_{\mathbf{g}}$ corresponds to ξ_g of Thorkildsen *et al.* (2001).

$$P_{\mathbf{h}} = P_{\mathbf{h}}^0 \Bigg\{ 1 - \tfrac{1}{3}\frac{\mathbf{p}^{(1)}\cdot\mathbf{p}_1^{(3)}}{\mathbf{p}^{(1)}\cdot\mathbf{p}^{(1)}}|\eta_{\mathbf{oh}}|^2 - 2\frac{\mathbf{p}^{(1)}\cdot\mathbf{p}^{(2)}}{\mathbf{p}^{(1)}\cdot\mathbf{p}^{(1)}}\Gamma[f_2(2v_{\mathbf{g}})\cos\Phi_\Sigma + f_1(2v_{\mathbf{g}})\sin\Phi_\Sigma] - 2\left[\frac{\mathbf{p}^{(1)}\cdot\mathbf{p}_2^{(3)}}{\mathbf{p}^{(1)}\cdot\mathbf{p}^{(1)}}|\eta_{\mathbf{og}}|^2 + \frac{\mathbf{p}^{(1)}\cdot\mathbf{p}_3^{(3)}}{\mathbf{p}^{(1)}\cdot\mathbf{p}^{(1)}}|\eta_{\mathbf{hg}}|^2\right]f_3(2v_{\mathbf{g}}) + 2\frac{\mathbf{p}^{(2)}\cdot\mathbf{p}^{(2)}}{\mathbf{p}^{(1)}\cdot\mathbf{p}^{(1)}}\Gamma^2 f_3(2v_{\mathbf{g}})\Bigg\}. \quad (30)$$

$\mathcal{P}_{\mathbf{h}}^0$ is the kinematical integrated power. The f functions are defined by:

$$f_1(x) = (1-\cos x)/x^2,$$
$$f_2(x) = (x-\sin x)/x^2,$$
$$f_3(x) = (x-\sin x)/x^3.$$

We observe that equation (30) corresponds to equation (6) of Thorkildsen *et al.* (2001), the main result from the Takagi–Taupin approach. Furthermore, it also corresponds to equation (11) of Shen (2000) or equation (12) of Shen & Huang (2001), the 'thin-crystal' result from the EDWA approach. The term in (30) proportional to $|\eta_{\mathbf{oh}}|^2$ is the first-order correction of the integrated power owing to primary extinction. The term involving $\cos\Phi_\Sigma$ and $\sin\Phi_\Sigma$ carries the triplet phase information in a combination with the profile shape functions f_1 and f_2. One observes that the sine term, which is missing in the original formulation by Shen, enters the calculation through the coupling to the imaginary part of $g^{(2)}$ originating from the δ-function term in (27). The terms proportional to $|\eta_{\mathbf{og}}|^2$ and $|\eta_{\mathbf{hg}}|^2$ contribute to *Aufhellung*, while the one proportional to Γ^2 gives rise to phase-independent *Umweganregung*.

Fig. 1 shows results for the relative change in integrated power, $(\mathcal{P}_{\mathbf{h}} - \mathcal{P}_{\mathbf{h}}^0)/\mathcal{P}_{\mathbf{h}}^0$, for the $2\bar{2}0/0\bar{2}2/20\bar{2}$ three-beam case in silicon, comparing equation (11) of Shen (2000) with equation (30) of the present work. Parameters and units are specified in Thorkildsen *et al.* (2001) with the addition of $A\tau = 0.1252t$, $A\eta_G = -0.2443t\Delta\psi$ and $\delta = \Phi_\Sigma$, necessary to handle Shen's equation. The incoming beam is σ-polarized with $\mathbf{D}_0$ normal to the plane spanned by $\hat{\mathbf{s}}_{\mathbf{o}}$ and $\hat{\mathbf{s}}_{\mathbf{g}}$, the polarization state specified by Shen. The minor differences appearing in the figure are due to the presence of the *Aufhellung* terms in (30). Three-beam profiles calculated from (30) are indistinguishable from what is obtained from the Takagi theory, *cf.* equation (12) of Thorkildsen *et al.* (2001). In that work, a comparison is also made with calculations from the full dynamical theory, indicating the actual range of applicability of series-expansion approaches.

3. Concluding remarks

The current work shows that a modified version of the original approach by Shen to three-beam diffraction in a perfect crystal is equivalent to the Takagi–Taupin approach when applied to a semi-infinite crystal slab. The formal structure of the present approach is similar to what is found when quantum field theoretical methods are applied to dynamical diffraction theories (Ohtsuki & Yanagawa, 1966*a*,*b*). Besides, a generalization to more than three beams, which is not obvious within the Takagi–Taupin approach, may be implemented. It is an open question whether the present procedure for calculation might be carried out for a finite perfect crystal for instance in the shape of a sphere where $\mathcal{S}(\Delta\mathbf{k}) = \mathcal{S}(|\Delta\mathbf{k}|)$. This would eliminate the time-consuming analysis needed to reveal the coupled surface integrations that appear when the point-source method (Becker, 1977) is used to analyze the Takagi–Taupin equations.

The self-consistent dynamical balance of wave amplitudes, as it appears within the fundamental theory, is broken when perturbative approximation schemes are applied. One should therefore expect significant deviations when results obtained by series-expansion methods are compared with full simulations based on the fundamental theory in cases where the characteristic dimension of the diffracting domain exceeds one extinction length.

APPENDIX *A*
Function definitions

The g functions are given by:

$$g_{\Re}^{(2)}(u, v_{\mathbf{g}}) = \frac{(u+2v_{\mathbf{g}})\sin u - u\sin(u+2v_{\mathbf{g}})}{4uv_{\mathbf{g}}(u+v_{\mathbf{g}})},$$
$$g_{\Im}^{(2)}(u, v_{\mathbf{g}}) = \frac{\sin v_{\mathbf{g}}\sin(u+v_{\mathbf{g}})}{2v_{\mathbf{g}}(u+v_{\mathbf{g}})},$$
$$g_{1,\Re}^{(3)}(u) = \frac{u\cos u - \sin u}{2u^3},$$
$$g_{1,\Im}^{(3)}(u) = 0,$$
$$g_{2,\Re}^{(3)}(u, v_{\mathbf{g}}) = [-2uv_{\mathbf{g}}(u+v_{\mathbf{g}})\cos u - (u^2 - 2v_{\mathbf{g}}^2)\sin u + u^2\sin(u+2v_{\mathbf{g}})]/[8u^2v_{\mathbf{g}}^2(u+v_{\mathbf{g}})],$$
$$g_{2,\Im}^{(3)}(u, v_{\mathbf{g}}) = \frac{v_{\mathbf{g}}(u+v_{\mathbf{g}})\sin u - u\sin v_{\mathbf{g}}\sin(u+v_{\mathbf{g}})}{4uv_{\mathbf{g}}^2(u+v_{\mathbf{g}})},$$
$$g_{3,\Re}^{(3)}(u, v_{\mathbf{g}}) = [2uv_{\mathbf{g}}(u+v_{\mathbf{g}})\cos u - (u^2 + 4uv_{\mathbf{g}} + 2v_{\mathbf{g}}^2)\sin u + u^2\sin(u+2v_{\mathbf{g}})]/[8u^2v_{\mathbf{g}}(u+v_{\mathbf{g}})^2],$$
$$g_{3,\Im}^{(3)}(u, v_{\mathbf{g}}) = \frac{v_{\mathbf{g}}(u+v_{\mathbf{g}})\sin u - u\sin v_{\mathbf{g}}\sin(u+v_{\mathbf{g}})}{4uv_{\mathbf{g}}(u+v_{\mathbf{g}})^2}.$$

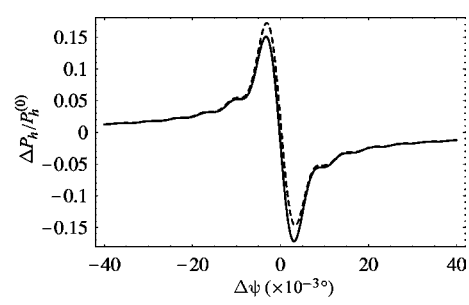

Figure 1
Three-beam profiles for the symmetrical transmission case $2\bar{2}0/0\bar{2}2/20\bar{2}$ in silicon. Plate thickness $t = 2$ µm. The solid line represents equation (30) of the present work, the dashed line equation (11) of Shen (2000), *cf.* also Fig. 4 of Thorkildsen *et al.* (2001).

The h functions are defined according to:

$$\begin{aligned}
h^{(0)}(u) &= [g^{(1)}(u)]^2,\\
h_1^{(1)}(u, v_{\mathbf{g}}) &= g^{(1)}(u) g_{\Re}^{(2)}(u, v_{\mathbf{g}}),\\
h_2^{(1)}(u, v_{\mathbf{g}}) &= g^{(1)}(u) g_{\Im}^{(2)}(u, v_{\mathbf{g}}),\\
h_1^{(2)}(u) &= g^{(1)}(u) g_{1,\Re}^{(3)}(u),\\
h_2^{(2)}(u, v_{\mathbf{g}}) &= g^{(1)}(u) g_{2,\Re}^{(3)}(u, v_{\mathbf{g}}),\\
h_3^{(2)}(u, v_{\mathbf{g}}) &= g^{(1)}(u) g_{3,\Re}^{(3)}(u, v_{\mathbf{g}}),\\
h_4^{(2)}(u, v_{\mathbf{g}}) &= [g_{\Re}^{(2)}(u, v_{\mathbf{g}})]^2 + [g_{\Im}^{(2)}(u, v_{\mathbf{g}})]^2
\end{aligned}$$

and are explicitly expressed by:

$$\begin{aligned}
h^{(0)}(u) &= (\sin u/u)^2,\\
h_1^{(1)}(u, v_{\mathbf{g}}) &= \frac{\sin u[(u + 2v_{\mathbf{g}}) \sin u - u \sin(u + 2v_{\mathbf{g}})]}{4u^2 v_{\mathbf{g}}(u + v_{\mathbf{g}})},\\
h_2^{(1)}(u, v_{\mathbf{g}}) &= \frac{\sin u \sin v_{\mathbf{g}} \sin(u + v_{\mathbf{g}})}{2u v_{\mathbf{g}}(u + v_{\mathbf{g}})},\\
h_1^{(2)}(u) &= \frac{\sin u[u \cos u - \sin u]}{2u^4},\\
h_2^{(2)}(u, v_{\mathbf{g}}) &= \{\sin u[-2u v_{\mathbf{g}}(u + v_{\mathbf{g}}) \cos u - (u^2 - 2v_{\mathbf{g}}^2) \sin u\\
&\quad + u^2 \sin(u + 2v_{\mathbf{g}})]\}/[8u^3 v_{\mathbf{g}}^2 (u + v_{\mathbf{g}})],\\
h_3^{(2)}(u, v_{\mathbf{g}}) &= \{\sin u[2u v_{\mathbf{g}}(u + v_{\mathbf{g}}) \cos u - (u^2 + 4u v_{\mathbf{g}} + 2v_{\mathbf{g}}^2) \sin u\\
&\quad + u^2 \sin(u + 2v_{\mathbf{g}})]\}/[8u^3 v_{\mathbf{g}} (u + v_{\mathbf{g}})^2],\\
h_4^{(2)}(u, v_{\mathbf{g}}) &= \{u^2 + u v_{\mathbf{g}} + v_{\mathbf{g}}^2 - (u + v_{\mathbf{g}})(v_{\mathbf{g}} \cos 2u + u \cos 2v_{\mathbf{g}})\\
&\quad + u v_{\mathbf{g}} \cos[2(u + v_{\mathbf{g}})]\}/[8u^2 v_{\mathbf{g}}^2 (u + v_{\mathbf{g}})^2].
\end{aligned}$$

These functions correspond to those defined in Thorkildsen *et al.* (2001). Notice the two errors occurring in that paper: In equation (5), the last term should read $\zeta^2 \Gamma^2 h_4^{(2)}(\alpha, \beta)$ and in the definition of $h_1^{(1)}$, $\sin[(3\alpha/2)(\alpha - 2\beta)]$ should be replaced by $\sin[(3/2)(\alpha - 2\beta)]$.

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