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Three-Beam Diffraction in Finite Perfect Crystals. I. Exploring Laue–Laue and Bragg–Laue Scattering Using a Series-Expansion Approach for the Solution of the Takagi–Taupin Equations

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Abstract

Using the boundary-value Green-function technique, analytical expressions in the form of finite series expansions are obtained for the relative change in the integrated power of the primary reflection due to the gradual excitation of a secondary reciprocal-lattice point on the Ewald sphere. Solutions are found for both a Laue–Laue and a Bragg–Laue case in finite shaped crystals confined by the scattering vectors. When the crystal sizes do not exceed the *Pendellösung* length of the involved reflections, the ψ profiles exhibit the same qualitative features in the two cases. The solutions do however indicate a strong dependence on the outer crystal dimensions – which add a geometrical aspect to the interpretation of the *Aufhellung* and *Umweganregung* concepts.

1. Introduction

During the last 10 years, the development of high-brilliance synchrotron sources has eased the possibilities of using three-beam diffraction as a tool to experimentally determine invariant triplet phase sums. For a general review on the topic, lead articles by Chang (1992) and Weckert & Hümmer (1997) should be consulted.

The idea of using three-beam diffraction to extract phase information originates from Lipscomb (1949). Throughout the years, different aspects concerning multiple-beam diffraction and the phase problem have been explored. Major theoretical advances have been achieved by several authors (Ewald & Héno, 1968; Colella, 1974; Hümmer & Billy, 1982; Høier & Marthinsen, 1983; Chang, 1984; Juretschke, 1984; Hümmer & Billy, 1986; Weckert & Hümmer, 1990). The theoretical basis common to all these works is the fundamental equation of the plane-wave dynamical theory (von Laue, 1932; Pinsker, 1978). Here, the description of the scattering process is transformed to an eigenvalue problem in the *Anpassung*, δ, leading to the concept of dispersion surfaces. Solutions are usually found using numerical methods and boundary conditions for semi-infinite crystal plates. Many-beam effects then can be associated with perturbations of the ordinary two-beam dispersion surface sheets.

Thorkildsen (1987) proposed an alternative approach using the Takagi–Taupin equations (Takagi, 1962). This was found to be particularly suited for describing three-beam diffraction in finite crystals. Analytical solutions were obtained for the wave fields D_o, D_h and D_g by applying the Laplace transformation technique. The calculations were performed using a parallelepiped model crystal spanned by the scattering vectors involved. In the following, we pursue these ideas. Instead of searching for closed solutions for the fields, we use a series-expansion technique that involves smallness parameters inversely proportional to the extinction length of the reflections (Thorkildsen, 1990). This approach, which actually counts the scattering events to a given order along the possible optical routes, generates analytical results in cases where other mathematical treatments are not applicable. It might even be possible to extend this technique to crystals having a more general outer shape than what is considered here. In this paper, the concepts of the approach are presented. The influence of absorption, resonant scattering, polarization and crystal imperfection will be considered separately in forthcoming papers.

2. Field equations

Let us start by considering a linear dielectric medium with no free charges or currents in the matter. We further presume that the medium should have the same magnetic properties as vacuum. By combining Maxwell's equations,† we obtain the following wave equation for the electric displacement field, **D**:

$$\nabla \times \nabla \times [1 - \chi_e(\mathbf{r})]\mathbf{D} = -(1/c^2)(\partial^2\mathbf{D}/\partial t^2), \quad (1)$$

where we have used von Laue's approximation for the electric field:

$$\mathbf{E} \approx (1/\varepsilon_0)[1 - \chi_e(\mathbf{r})]\mathbf{D}.$$

† SI units are used throughout this paper.

ε_0 is the permittivity of free space, $\chi_e(\mathbf{r})$ is the electric susceptibility, which generally is a complex quantity, and may be expanded into a Fourier series:

$$\chi_e(\mathbf{r}) = \sum_h \chi_h \exp(-2\pi i \mathbf{h} \cdot \mathbf{r}). \tag{2}$$

Using the field expansion

$$\mathbf{D} = \sum_g \mathbf{D}_g(\mathbf{r}) \exp[2\pi i(\nu t - \mathbf{k}_g \cdot \mathbf{r})] \tag{3}$$

and assuming a gentle spatial variation of $\mathbf{D}$, we obtain to first order in χ_e the so-called Takagi–Taupin equations (Takagi, 1962, 1969; Taupin, 1964; Authier *et al.*, 1968; Kato, 1973) for a perfect crystal:

$$(i/\pi)(\mathbf{k}_p \cdot \nabla)\mathbf{D}_p = \{K^2(1+\chi_o) - \mathbf{k}_p^2\}\mathbf{D}_p + \sum_{g \neq p} \chi_{p-g}\{\mathbf{k}_p^2 \mathbf{D}_g - (\mathbf{k}_p \cdot \mathbf{D}_g)\mathbf{k}_p\}. \tag{4}$$

$K = 1/\lambda$ is the wave number of the incoming vacuum wave, $\mathbf{K}_o$, and $\mathbf{k}_p = \mathbf{k}_o + \mathbf{p}$ is a crystal wavevector, $\mathbf{p}$ being a reciprocal-lattice vector.

Since $\mathbf{D}_p$ is perpendicular to $\mathbf{k}_p$, it may be expanded into two vector components:

$$\mathbf{D}_p(\mathbf{r}) = \sum_{\nu=1}^{2} D_p^\nu(\mathbf{r})\, \hat{\mathbf{e}}_p^\nu.$$

$\hat{\mathbf{e}}_p^1 = \hat{\boldsymbol{\sigma}}_p$ and $\hat{\mathbf{e}}_p^2 = \hat{\boldsymbol{\pi}}_p$ denote the unit polarization vectors associated with the wave $\mathbf{k}_p$.

Using this relation and defining the mean wavevector inside the crystal, $k = K(1 + \frac{1}{2}\chi_o)$, one can write the component form of (4) as

$$(\mathbf{k}_p \cdot \nabla)D_p^\mu = 2\pi i K(\|\mathbf{k}_p\| - k)D_p^\mu - \pi i K^2 \times \sum_{g \neq p} \sum_\nu \chi_{p-g}\{\hat{\mathbf{e}}_p^\mu \cdot \hat{\mathbf{e}}_g^\nu\} D_g^\nu,$$

where the notation $\|\mathbf{k}_p\|$ represents the *norm* (or length) of the vector $\mathbf{k}_p$. We define the *deviation parameter* (Authier *et al.*, 1968) as

$$\beta_p = \|\mathbf{k}_p\| - k = \alpha_p - K\delta\gamma_p - \tfrac{1}{2}K\chi_o, \tag{5}$$

where $\alpha_p = \|\mathbf{K}_p\| - K$ is the excitation error associated with reflection p. δ, the *Anpassung*, is treated in the kinematical limit by putting $\delta = -\chi_o/2\gamma_o$. This corresponds to $\beta_o = \alpha_o = 0$ (Authier, 1996). γ_p is a direction cosine, expressed by $\gamma_p = \hat{\mathbf{n}} \cdot \hat{\mathbf{s}}_p$, where $\hat{\mathbf{n}}$ is the unit normal vector to the entrance surface directed into the crystal and $\hat{\mathbf{s}}_p$ is a unit vector along $\mathbf{K}_p = \mathbf{K}_o + \mathbf{p}$.

Finally, we introduce the definition

$$\kappa_{pq}^{\mu\nu} = -\pi K \chi_{p-q}\{\hat{\mathbf{e}}_p^\mu \cdot \hat{\mathbf{e}}_q^\nu\} = (r_e\lambda/V_c)F_{p-q}\{\hat{\mathbf{e}}_p^\mu \cdot \hat{\mathbf{e}}_q^\nu\}, \tag{6}$$

where r_e is the classical electron radius, V_c the unit-cell volume and F_{p-q} the structure factor associated with the reflection $p - q$.

The Takagi–Taupin equations may then be written

$$\partial D_p^\mu / \partial s_p = 2\pi i \beta_p D_p^\mu + i \sum_{q \neq p} \sum_\nu \kappa_{pq}^{\mu\nu} D_q^\nu, \tag{7}$$

s_p being a positional coordinate along $\hat{\mathbf{s}}_p$.

Redefining the phases of the field amplitudes by performing a unitary transformation,

$$D_p^\mu = \tilde{D}_p^\mu \exp\left(2\pi i \sum_q \beta_q s_q\right), \tag{8}$$

we remove the first term on the right-hand side of (7) and thus obtain the equations in their final general form:

$$\partial \tilde{D}_p^\mu / \partial s_p = i \sum_{q \neq p} \sum_\nu \kappa_{pq}^{\mu\nu} \tilde{D}_q^\nu. \tag{9}$$

The transformation (8) corresponds to using the Lorentz point as the excitation point for the fields (Thorkildsen & Larsen, 1997). $\tilde{D}_p^\mu$ are thus the solutions for the exact three-beam position.

In order to simplify the mathematical expressions, we neglect for the time being any interactions between different polarization states of the vector amplitudes. Effects of such interactions will be considered separately in a following paper.

In the case of three-beam interactions, $p, q \in \{o, h, g\}$, the set of equations (9) may be written

$$\begin{aligned} \partial \tilde{D}_o / \partial s_o &= i\kappa_{oh}\tilde{D}_h + i\kappa_{og}\tilde{D}_g \\ \partial \tilde{D}_h / \partial s_h &= i\kappa_{ho}\tilde{D}_o + i\kappa_{hg}\tilde{D}_g \\ \partial \tilde{D}_g / \partial s_g &= i\kappa_{go}\tilde{D}_o + i\kappa_{gh}\tilde{D}_h. \end{aligned} \tag{10}$$

3. Principle of solution

The field amplitudes are now formally obtained by integration of the above equations. It is convenient to introduce a linear operator, $\mathcal{L}_{pq}$:

$$\mathcal{L}_{pq}\tilde{D}_q = i\kappa_{pq} \int_{s_p^b}^{s_p} \mathrm{d}s_p' \, \tilde{D}_q. \tag{11}$$

The appearance of the operator $\mathcal{L}_{pq}$ implies a scattering event $\hat{\mathbf{s}}_q \rightarrow \hat{\mathbf{s}}_p$. Thus, scattering of the beam from $\hat{\mathbf{s}}_g$ to $\hat{\mathbf{s}}_h$ is governed by

$$\mathcal{L}_{hg}\tilde{D}_g(s_o, s_h, s_g) = i\kappa_{hg} \int_{s_h^b}^{s_h} \mathrm{d}s_h' \, \tilde{D}_g(s_o, s_h', s_g). \tag{12}$$

Later, the appearance of a member of the set $\{\kappa_{pq}\}$ will label a scattering process.

We may thus write

$$\begin{aligned} \tilde{D}_o &= \tilde{D}_o^{(b)} + \mathcal{L}_{oh}\tilde{D}_h + \mathcal{L}_{og}\tilde{D}_g \\ \tilde{D}_h &= \tilde{D}_h^{(b)} + \mathcal{L}_{ho}\tilde{D}_o + \mathcal{L}_{hg}\tilde{D}_g \\ \tilde{D}_g &= \tilde{D}_g^{(b)} + \mathcal{L}_{go}\tilde{D}_o + \mathcal{L}_{gh}\tilde{D}_h. \end{aligned} \tag{13}$$

Here, b indicates a boundary point. In order to obtain the boundary-value Green functions for the wave fields, we apply a point source of strength $D_o^{(e)}$, associated with the incident direction, $\mathbf{K}_o$; and formulate the boundary conditions for the transformed fields by

$$\begin{aligned}\tilde{D}_o^{(b)} &= \tilde{D}_o[s_o^b(s_h, s_g), s_h, s_g] = D_o^{(e)}\delta(s_h)\delta(s_g)\\ \tilde{D}_h^{(b)} &= \tilde{D}_h[s_o, s_h^b(s_o, s_g), s_g] = 0\\ \tilde{D}_g^{(b)} &= \tilde{D}_g[s_o, s_h, s_g^b(s_o, s_h)] = 0,\end{aligned} \tag{14}$$

where $\delta(\,)$ denotes the Dirac delta function.

In order to keep track of the number of scattering events, we associate a counting variable, ε, with the smallness parameters, κ_{pq}, of equation (11). We then seek solutions for the fields in the form of series expansions in ε (*cf.* Kato, 1976; Becker, 1977):

$$\begin{aligned}\tilde{D}_o(s_o, s_h, s_g) &= D_o^{(e)} \sum_{n=0}^{\infty} \varepsilon^n d_o^{(n)}(s_o, s_h, s_g)\\ \tilde{D}_h(s_o, s_h, s_g) &= D_o^{(e)} \sum_{n=1}^{\infty} \varepsilon^n d_h^{(n)}(s_o, s_h, s_g)\\ \tilde{D}_g(s_o, s_h, s_g) &= D_o^{(e)} \sum_{n=1}^{\infty} \varepsilon^n d_g^{(n)}(s_o, s_h, s_g).\end{aligned} \tag{15}$$

In the final expressions, we let $\varepsilon \to 1$.

Combining the sets (13) and (15), we obtain, by equating equal powers of the counting variable, ε, the following recurrence relations (for $n \geq 1$):

$$\begin{aligned}d_o^{(n)} &= \mathcal{L}_{oh} d_h^{(n-1)} + \mathcal{L}_{og} d_g^{(n-1)}\\ d_h^{(n)} &= \mathcal{L}_{ho} d_o^{(n-1)} + \mathcal{L}_{hg} d_g^{(n-1)}\\ d_g^{(n)} &= \mathcal{L}_{go} d_o^{(n-1)} + \mathcal{L}_{gh} d_h^{(n-1)}.\end{aligned} \tag{16}$$

This, together with the boundary conditions for $\{d_p\}$,

$$d_o^{(0)} = \delta(s_h)\delta(s_g) \tag{17}$$

$$d_h^{(0)} = d_g^{(0)} = 0, \tag{18}$$

enable us to perform the calculations of the amplitudes in principle to any desired order. Explicitly, we have for the $\{d_h^{(n)}\}$:

$$d_h^{(1)} = \mathcal{L}_{ho}\, d_o^{(0)} \tag{19}$$

$$d_h^{(2)} = \mathcal{L}_{hg}\, \mathcal{L}_{go}\, d_o^{(0)} \tag{20}$$

$$\begin{aligned}d_h^{(3)} = {} & \mathcal{L}_{hg}\, \mathcal{L}_{gh}\, \mathcal{L}_{ho}\, d_o^{(0)} + \mathcal{L}_{ho}\, \mathcal{L}_{og}\, \mathcal{L}_{go}\, d_o^{(0)}\\ & + \mathcal{L}_{ho}\, \mathcal{L}_{oh}\, \mathcal{L}_{ho}\, d_o^{(0)}.\end{aligned} \tag{21}$$

This buildup of the field corresponds to Kato's optical zigzag routes representing multiple reflections (Kato, 1976).

The number of subterms, N, constituting $d_h^{(n)}$ is given by the recurrence relation $N(n) = N(n-1) + 2N(n-2)$, where $N(1) = N(2) = 1$. *I.e.* $N(6) = 21$, $N(10) = 341$ and $N(20) = 349\,525$. It is thus hardly worthwhile to go beyond the tenth order in the expansion – which sets a practical limit to this method.

The solution for $\tilde{D}_h$ gives the value of the transformed amplitude of the primary diffracted beam at the point (s_o, s_h, s_g) due to a point source at the origin. The wave amplitude of this beam at a point P, with coordinates $(s_o(P), s_h(P), s_g(P))$, due to a source at S with coordinates $(s_o(S), s_h(S), s_g(S))$ is given by†

$$\begin{aligned}D_h(P \leftarrow S) = {} & J\tilde{D}_h(\Delta_o, \Delta_h, \Delta_g)\exp(2\pi i\alpha_h\Delta_h)\\ & \times \exp(2\pi i\alpha_g\Delta_g)\theta(\Delta_o)\theta(\Delta_h)\theta(\Delta_g).\end{aligned} \tag{22}$$

Here, J is a geometrical factor, which enters through the transformation of the source Dirac field density function, using a representation in coordinates normal to the incident direction to the form $\delta(s_h)\delta(s_g)$ used in (14). We have also defined

$$\Delta_q = s_q(P) - s_q(S). \tag{23}$$

$\theta(\Delta_q)$ denotes the Heaviside unit step function, as the wave fields are zero outside the pyramid defined by the unit vectors $\hat{\mathbf{s}}_o$, $\hat{\mathbf{s}}_h$ and $\hat{\mathbf{s}}_g$, with the top located at S.

The real ($\Re$) and imaginary ($\Im$) components of the fields at an exit point, P, due to the incoming plane wave are obtained by superimposing the amplitudes from all source points, S, on the entrance surface, which contribute to the field at P (Becker, 1977; Becker & Dunstetter, 1984; Bremer & Thorkildsen, 1986). *I.e.*

$$\Re D_h(P) = \int \mathrm{d}\mathbf{S} \cdot \hat{\mathbf{s}}_o\, \Re D_h(P \leftarrow S), \tag{24}$$

with a similar equation for the imaginary part of the amplitude. The intensity of the primary diffracted beam at the point P on the exit surface is given by

$$I_h(P) = \frac{c}{2\varepsilon_0}|D_h(P)|^2 = \frac{c}{2\varepsilon_0}\{[\Re D_h(P)]^2 + [\Im D_h(P)]^2\}. \tag{25}$$

The power of the primary diffracted beam is then obtained by summing the contributions due to all points on the exit surface:

$$P_h = \int \mathrm{d}\mathbf{P} \cdot \hat{\mathbf{s}}_h\, I_h. \tag{26}$$

For calculation of the integrated power, it has been shown (Høier & Marthinsen, 1983) that the excitation errors, α_h and α_g, depend on two external divergence angles, ε_1 and ε_2.‡ Neglecting any dependence of the vertical divergence ε_2 and assuming that α_g varies negligibly with ε_1, we have to consider

$$\alpha_h = (\sin 2\theta_{oh}/\lambda)\varepsilon_1. \tag{27}$$

† Absorption and refraction are here neglected, *i.e.* $\chi_o \to 0 \Rightarrow \beta_p \to \alpha_p$.

‡ Which give the position of the incident wavevector relative to the Laue point.

The integrated power thus becomes

$$\mathcal{P}_h = \int_{-\infty}^{\infty} \mathrm{d}\varepsilon_1 \, P_h[\alpha_h(\varepsilon_1), \alpha_g]. \tag{28}$$

4. Crystal geometries

In this work, we have chosen two finite crystal shapes initiating different types of scattering geometry. As depicted in Figs. 1 and 2, the crystals are all confined by the unit scattering vectors $\{\hat{\mathbf{s}}_o, \hat{\mathbf{s}}_h, \hat{\mathbf{s}}_g\}$. This is in order to simplify the mathematical treatment, *i.e.* the integration set-up in equations (24) and (26) and the treatment of the boundary conditions for the fields. The model crystal shapes are chosen such that the outer surfaces are parallel to the characteristic planes associated with the equations (Sommerfeld, 1949; Sneddon, 1957; Myint-U, 1980).

Crystal dimensions are denoted l_o, l_h, l_g. Fig. 1 represents a Laue–Laue case, whereas Fig. 2 represents a Bragg–Laue case.

The volume, V_s^0, of the cell spanned by the set of unit vectors $\{\hat{\mathbf{s}}_o, \hat{\mathbf{s}}_h, \hat{\mathbf{s}}_g\}$ is given by

$$V_s^0 = \{1 - \cos^2 2\theta_{oh} - \cos^2 2\theta_{og} - \cos^2 2\theta_{hg} + 2\cos 2\theta_{oh} \cos 2\theta_{og} \cos 2\theta_{hg}\}^{1/2},$$

where $2\theta_{pq} \angle (\hat{\mathbf{s}}_p, \hat{\mathbf{s}}_q)$.

The geometrical factor, J, *cf.* (22), is in both cases

$$J = 1/V_s^0.$$

For the Laue–Laue case, we have:

$$\begin{aligned} \Delta_o &= s_o(P) \\ \Delta_h &= l_h - s_h(S) \\ \Delta_g &= s_g(P) - s_g(S). \end{aligned} \tag{29}$$

The wave field at P due to an incoming plane wave thus becomes

$$D_h(P) = V_s^0 \int_0^{l_h} \mathrm{d}\Delta_h \int_0^{s_g(P)} \mathrm{d}\Delta_g \, D_h(P \leftarrow S)$$

and the power at the exit surface is

$$P_h = V_s^0 \int_0^{l_o} \mathrm{d}s_o(P) \int_0^{l_g} \mathrm{d}s_g(P) \, I_h(P).$$

In the Bragg–Laue case, it is convenient to use a coordinate system, $\{\hat{\mathbf{r}}_o, \hat{\mathbf{r}}_1, \hat{\mathbf{r}}_2\}$, where $\hat{\mathbf{r}}_1$ is spanned by $\hat{\mathbf{s}}_o$ and $\hat{\mathbf{s}}_h$. The relations between the coordinates are given by

$$\begin{aligned} s_o &= r_o + r_1/2\cos\theta_{oh} \\ s_h &= r_1/2\cos\theta_{oh} \\ s_g &= r_2 \end{aligned}$$

and consequently

$$\begin{aligned} \Delta_o = \Delta_h &= (1/2\cos\theta_{oh})\{r_1(P) - r_1(S)\} \\ &\equiv (1/2\cos\theta_{oh})\Delta_1 \\ \Delta_g &= r_2(P) - r_2(S) \equiv \Delta_2. \end{aligned} \tag{30}$$

The wave field at P due to an incoming plane wave is written

$$D_h(P) = (V_s^0/2\cos\theta_{oh}) \int_0^{r_1(P)} \mathrm{d}\Delta_1 \int_0^{r_2(P)} \mathrm{d}\Delta_2 \, D_h(P \leftarrow S)$$

and the power at the exit surface becomes

$$P_h = (V_s^0/2\cos\theta_{oh}) \int_0^{2l_o\cos\theta_{oh}} \mathrm{d}r_1(P) \int_0^{l_g} \mathrm{d}r_2(P) \, I_h(P).$$

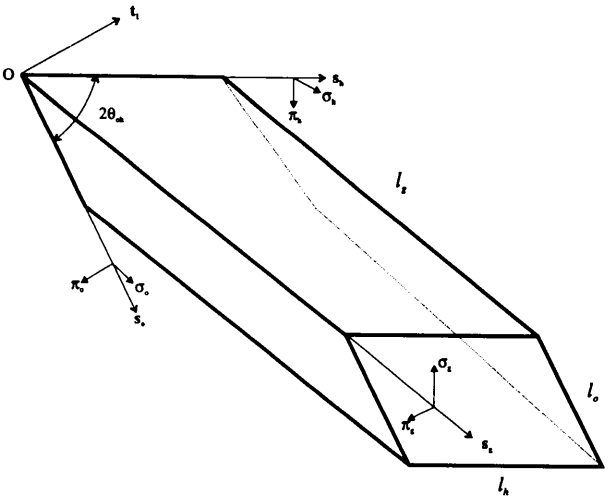

Fig. 1. Scattering, crystal and polarization geometry for the Laue–Laue case.

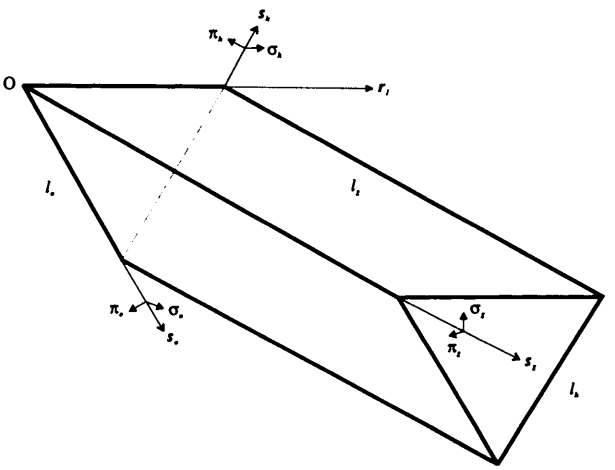

Fig. 2. Scattering, crystal and polarization geometry for the Bragg–Laue case.

5. Results

Using (11), (16), (17) and (18), we obtain the following results to the sixth order of perturbation (all expressions are to be multiplied by the factor $i\kappa_{ho}$).

Laue–Laue case:

$$
\begin{aligned}
d_h^{(1)} &= \delta(s_g)\\
d_h^{(2)} &= \Gamma(i\cos\varphi_\Sigma - \sin\varphi_\Sigma)\\
d_h^{(3)} &= -\Delta_o\Delta_h u\delta(s_g) - \Delta_o v - \Delta_h w\\
d_h^{(4)} &= \Gamma\{[\Delta_h\Delta_o u + \Delta_g\Delta_o v + \Delta_g\Delta_h w]\sin\varphi_\Sigma\\
&\quad - i[3\Delta_h\Delta_o u + \Delta_g\Delta_o v + \Delta_g\Delta_h w]\cos\varphi_\Sigma\}\\
d_h^{(5)} &= \tfrac{1}{4}\Delta_h^2\Delta_o^2u^2\delta(s_g) + \Delta_h\Delta_o^2uv + \tfrac{1}{2}\Delta_g\Delta_o^2v^2\\
&\quad + \Delta_h^2\Delta_o uw + 3\Delta_g\Delta_h\Delta_o vw + \tfrac{1}{2}\Delta_g\Delta_h^2w^2\\
&\quad + \Delta_g\Delta_h\Delta_o vw\cos 2\varphi_\Sigma + i\Delta_g\Delta_h\Delta_o vw\sin 2\varphi_\Sigma\\
d_h^{(6)} &= -\Gamma\{[\tfrac{1}{4}\Delta_h^2\Delta_o^2u^2 + \Delta_g\Delta_h\Delta_o^2uv + \tfrac{1}{4}\Delta_g^2\Delta_o^2v^2\\
&\quad + \Delta_g\Delta_h^2\Delta_o uw + \Delta_g^2\Delta_h\Delta_o vw + \tfrac{1}{4}\Delta_g^2\Delta_h^2w^2]\sin\varphi_\Sigma\\
&\quad - i[\tfrac{5}{4}\Delta_h^2\Delta_o^2u^2 + 3\Delta_g\Delta_h\Delta_o^2uv + \tfrac{1}{4}\Delta_g^2\Delta_o^2v^2\\
&\quad + 3\Delta_g\Delta_h^2\Delta_o uw + \Delta_g^2\Delta_h\Delta_o vw\\
&\quad + \tfrac{1}{4}\Delta_g^2\Delta_h^2w^2]\cos\varphi_\Sigma\}.
\end{aligned}
$$

Bragg–Laue case:

$$
\begin{aligned}
d_h^{(1)} &= \delta(s_g)\\
d_h^{(2)} &= \Gamma(i\cos\varphi_\Sigma - \sin\varphi_\Sigma)\\
d_h^{(3)} &= -\tfrac{1}{2}\Delta_h^2u\delta(s_g) - \Delta_h v - \Delta_h w\\
d_h^{(4)} &= \Gamma\{[\tfrac{1}{2}\Delta_h^2u + \Delta_h\Delta_g v + \Delta_h\Delta_g w]\sin\varphi_\Sigma\\
&\quad - i[\tfrac{3}{2}\Delta_h^2u + \Delta_h\Delta_g v + \Delta_h\Delta_g w]\cos\varphi_\Sigma\}\\
d_h^{(5)} &= \tfrac{1}{12}\Delta_h^4u^2\delta(s_g) + \tfrac{1}{2}\Delta_h^3uv + \tfrac{1}{2}\Delta_h^2\Delta_g v^2\\
&\quad + \tfrac{1}{2}\Delta_h^3uw + 2\Delta_h^2\Delta_g vw + \tfrac{1}{2}\Delta_h^2\Delta_g w^2\\
&\quad + \tfrac{1}{2}\Delta_h^2\Delta_g vw\cos 2\varphi_\Sigma + \tfrac{i}{2}\Delta_h^2\Delta_g vw\sin 2\varphi_\Sigma\\
d_h^{(6)} &= -\Gamma\{[\tfrac{1}{12}\Delta_h^4u^2 + \tfrac{1}{2}\Delta_h^3\Delta_g uv + \tfrac{1}{4}\Delta_h^2\Delta_g^2v^2\\
&\quad + \tfrac{1}{2}\Delta_h^3\Delta_g uw + \tfrac{3}{4}\Delta_h^2\Delta_g^2vw + \tfrac{1}{4}\Delta_h^2\Delta_g^2w^2]\sin\varphi_\Sigma\\
&\quad - i[\tfrac{5}{12}\Delta_h^4u^2 + \tfrac{3}{2}\Delta_h^3\Delta_g uv + \tfrac{1}{4}\Delta_h^2\Delta_g^2v^2\\
&\quad + \tfrac{3}{2}\Delta_h^3\Delta_g uw + \tfrac{3}{4}\Delta_h^2\Delta_g^2vw + \tfrac{1}{4}\Delta_h^2\Delta_g^2w^2]\cos\varphi_\Sigma\}.
\end{aligned}
$$

For the Laue–Laue case, $\{\Delta_o, \Delta_h, \Delta_g\}$ are given by (29) and, for the Bragg–Laue case, $\{\Delta_o = \Delta_h, \Delta_g\}$ by (30).

In addition, we have introduced the following definitions:

$$u = \kappa_{oh}\kappa_{ho}, \quad v = \kappa_{og}\kappa_{go}, \quad w = \kappa_{hg}\kappa_{gh}, \tag{31}$$

$$\Gamma = |\kappa_{hg}||\kappa_{go}|/|\kappa_{ho}|,$$

$$\varphi_\Sigma = \varphi_{oh} + \varphi_{hg} + \varphi_{go}. \tag{32}$$

Our definition of φ_Σ is used owing to symmetry reasons and corresponds to the one given by Giacovazzo *et al.* (1992) for the phase of the triplet invariant. In experiments, the observable phase information will be given by $\varphi_{hg} + \varphi_{go} - \varphi_{ho}$ (Hümmer & Weckert, 1990). The difference between this expression and our definition will be apparent in cases of resonant scattering (Larsen & Thorkildsen, 1998). It should also be noted that the sign in the definition of φ_Σ, equation (32), has been reversed compared to the previous paper (Thorkildsen, 1987). The present definition is therefore in accordance with prevalent notations (Hümmer & Billy, 1982) when resonant scattering is neglected.

The phase angle φ_{pq} is defined through

$$\kappa_{pq} = |\kappa_{pq}|\exp(i\varphi_{pq}). \tag{33}$$

Neglecting absorption and anomalous scattering, we may write

$$\kappa_{qp} = |\kappa_{pq}|\exp(-i\varphi_{pq}).$$

Algorithms corresponding to equations (16), (22), (25) and (26) were implemented in *MATHEMATICA*.† The integration procedures were extensively coded in order to ease the algebraic manipulations. Finally, the following expressions for the power in the Laue–Laue and Bragg–Laue cases were obtained (for convenience, only the results to third order are presented here; the *MATHEMATICA* code for the expressions up to the sixth order is available from the authors upon request).

Laue–Laue case:

$$
\begin{aligned}
P_h &= I_o|\kappa_{ho}|^2v_Ll_h\{2f_1(\xi_h) - |\eta_{oh}|^2f_1(\xi_h)\\
&\quad - 4(|\eta_{hg}||\eta_{go}|/|\eta_{ho}|)[f_2(\xi_g)\cos\varphi_\Sigma\\
&\quad + f_1(\xi_g)\sin\varphi_\Sigma]f_1(\xi_h) - 2|\eta_{go}|^2f_1(\xi_g)f_1(\xi_h)\\
&\quad - 2|\eta_{hg}|^2[f_1(\xi_g)f_1(\xi_h) + f_2(\xi_g)f_4(\xi_h)]\\
&\quad + 4(|\eta_{hg}|^2|\eta_{go}|^2/|\eta_{ho}|^2)f_3(\xi_g)f_1(\xi_h)\}.
\end{aligned} \tag{34}
$$

Bragg–Laue case:

$$
\begin{aligned}
P_h &= 2I_o|\kappa_{ho}|^2v_Bl_o\{2f_3(\xi_h) - |\eta_{oh}|^2f_5(\xi_h)\\
&\quad - 4(|\eta_{hg}||\eta_{go}|/|\eta_{ho}|)[f_2(\xi_g)\cos\varphi_\Sigma\\
&\quad + f_1(\xi_g)\sin\varphi_\Sigma]f_3(\xi_h)\\
&\quad - 2(|\eta_{go}|^2 + |\eta_{hg}|^2)[f_1(\xi_g)f_6(\xi_h) - f_2(\xi_g)f_7(\xi_h)]\\
&\quad + 2(|\eta_{hg}|^2|\eta_{go}|^2/|\eta_{ho}|^2)f_3(\xi_g)f_3(\xi_h)\}.
\end{aligned} \tag{35}
$$

Here, $I_o = (c/2\varepsilon_0)|D_o^{(e)}|^2$ is the intensity of the incident beam, and v_L and v_B are the volumes of the Laue–Laue and Bragg–Laue crystals, respectively.

† *MATHEMATICA* is a trademark of Wolfram Research Inc., Champaign, IL 61820, USA.

We have also introduced the following dimensionless quantities:

$$|\eta_{pq}|^2 = |\kappa_{pq}|^2 l_p l_q$$
$$\xi_h = 2\pi\alpha_h l_h$$
$$\xi_g = 2\pi\alpha_g l_g.$$

The functions f_i are given by

$$f_1(u) = (1/u^2)(1 - \cos u)$$
$$f_2(u) = (1/u)[1 - (1/u)\sin u]$$
$$f_3(u) = (1/u^2)[1 - (1/u)\sin u]$$
$$f_4(u) = (1/u^2)[\sin u - (2/u)(1 - \cos u)]$$
$$f_5(u) = (1/3u^5)(-12u + u^3 - 12u\cos u + 24\sin u - 3u^2\sin u)$$
$$f_6(u) = (1/2u^4)(2 + u^2 - 2\cos u - 2u\sin u)$$
$$f_7(u) = (1/u^4)(-2u - u\cos u + 3\sin u). \quad (36)$$

The integrated power may now be calculated using (28), neglecting any variation of α_g with ε_1 – *cf.* (27). The

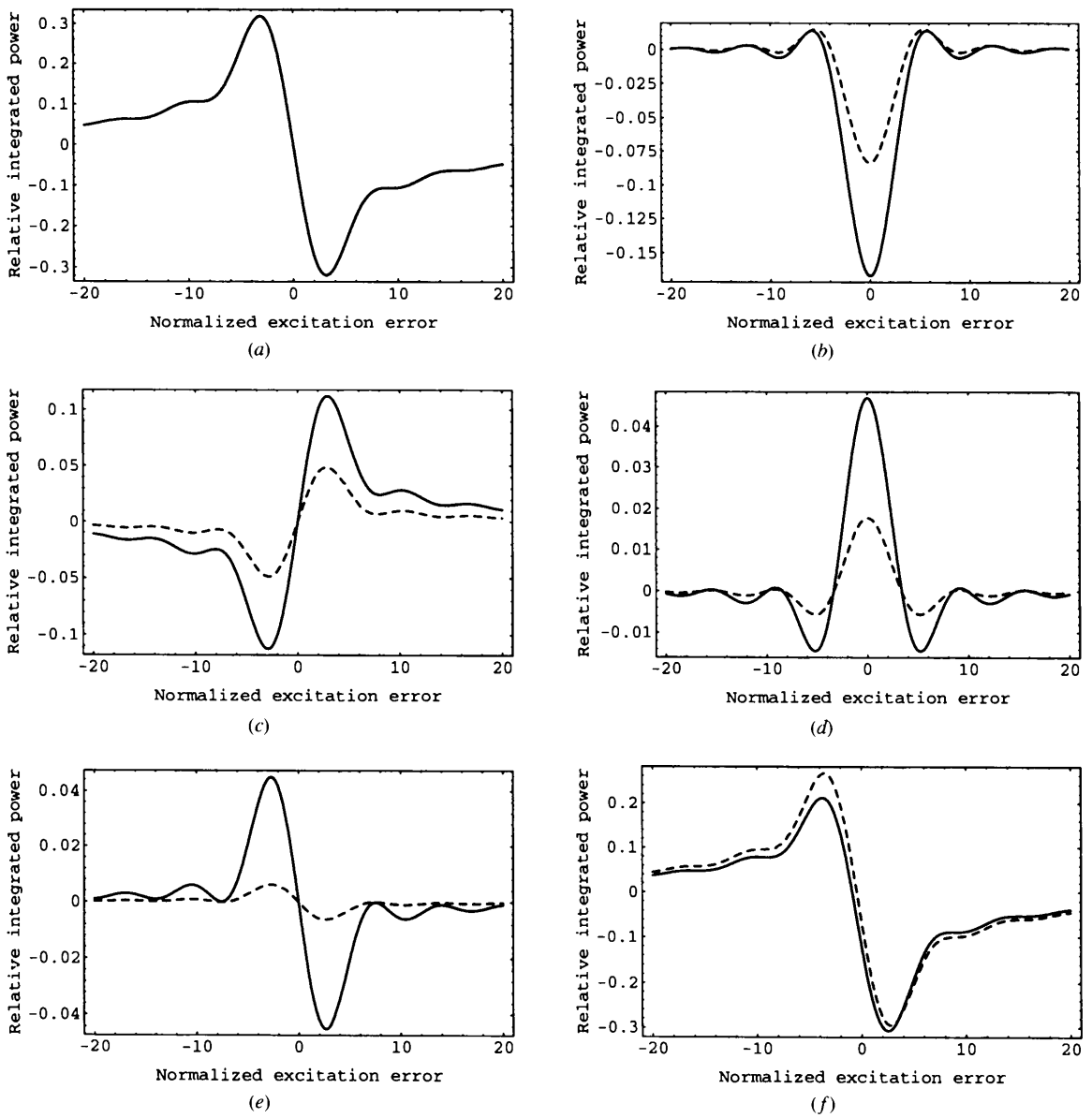

Fig. 3. Relative integrated power, $\Delta\mathcal{P}_h/\mathcal{P}_h^0$, as a function of the normalized excitation error, $\xi_g = 2\pi\alpha_g l_g$. Solid line: Laue–Laue case; dashed line: Bragg–Laue case. $|\eta_{oh}| = |\eta_{og}| = |\eta_{gh}| = 0.5$. $\varphi_\Sigma = 0$. (*a*) Second order, (*b*) third order, (*c*) fourth order, (*d*) fifth order, (*e*) sixth order. (*f*) Contribution added. Note the different scales on the ordinate axis.

terms proportional to $|\eta_{oh}|^2$ in equations (34) and (35) represent first-order correction of the diffracted power due to primary extinction. They are built from the scattering event $\kappa_{ho}\kappa_{oh}\kappa_{ho}$ in equation (21), thus involving multiple scattering among the $\hat{\mathbf{s}}_o$ and $\hat{\mathbf{s}}_h$ beams. In subsequent treatment, we neglect such terms. In order to obtain an expression for the relative change in the integrated power due to three-beam interaction, we introduce the kinematical two-beam integrated power:

$$\mathcal{P}_h^0 = I_o v|\kappa_{ho}|^2(\lambda/\sin 2\theta_{oh}). \tag{37}$$

To the third order of perturbation, we thus get:

Laue–Laue case:

$$\begin{aligned}\Delta\mathcal{P}_h(\xi_g)/\mathcal{P}_h^0 = &-2(|\eta_{hg}||\eta_{go}|/|\eta_{ho}|)\\ &\times [f_2(\xi_g)\cos\varphi_\Sigma + f_1(\xi_g)\sin\varphi_\Sigma]\\ &-(|\eta_{go}|^2 + |\eta_{hg}|^2)f_1(\xi_g)\\ &+2(|\eta_{hg}|^2|\eta_{go}|^2/|\eta_{ho}|^2)f_3(\xi_g).\end{aligned} \tag{38}$$

Bragg–Laue case:

$$\begin{aligned}\Delta\mathcal{P}_h(\xi_g)/\mathcal{P}_h^0 = &-2(|\eta_{hg}||\eta_{go}|/|\eta_{ho}|)[f_2(\xi_g)\cos\varphi_\Sigma\\ &+f_1(\xi_g)\sin\varphi_\Sigma] - \tfrac{2}{3}(|\eta_{go}|^2 + |\eta_{hg}|^2)f_1(\xi_g)\\ &+(|\eta_{hg}|^2|\eta_{go}|^2/|\eta_{ho}|^2)f_3(\xi_g)\end{aligned} \tag{39}$$

with $\xi_g = \xi_g(\psi) = 2\pi\alpha_g(\psi)l_g$. We note that the result for the Laue–Laue case is identical to the one found from the exact field expressions (Thorkildsen, 1987).

6. Discussion

The solutions obtained for the different crystal and scattering geometries are confined to a limited range of $|\eta_{pq}|$ values, owing to the series-expansion cut-off. For the present development to the sixth order in the scattering power, the expressions are valid for $|\eta_{pq}| < 1$. Then, the systematic error due to the cut-off is less than 10^{-4}.

Fig. 3 shows how the perturbation terms add up to build the diffracted power. It should be noticed that the contribution to the second order† is the same in both the Laue–Laue and Bragg–Laue cases considered. From the figure, and from equations (38) and (39), we find that there are no qualitative differences between the Laue–Laue and Bragg–Laue reflection geometries – at least for crystal dimensions not exceeding the *Pendellösung* distance in the Laue–Laue case. This is illustrated in Fig. 4, where the relative change in the integrated power for four different crystal sizes‡ is depicted. From the

† In the series expansion.

‡ We hence consider the same reflection triplet, but vary l_o, l_h and l_g.

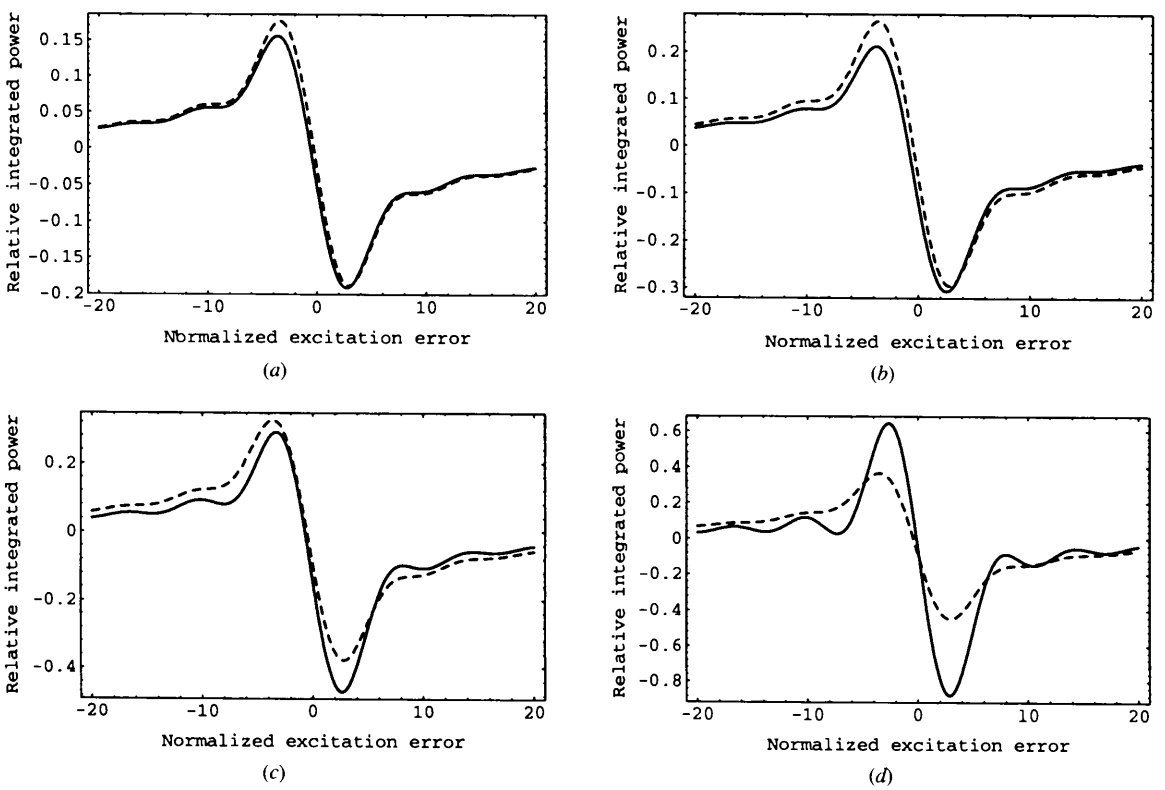

Fig. 4. Effect of increasing the crystal size. Solid line: Laue–Laue case; dashed line: Bragg–Laue case. $\varphi_\Sigma = 0$. (*a*) $|\eta_{oh}| = |\eta_{og}| = |\eta_{gh}| = 0.3$, (*b*) $|\eta_{oh}| = |\eta_{og}| = |\eta_{gh}| = 0.5$, (*c*) $|\eta_{oh}| = |\eta_{og}| = |\eta_{gh}| = 0.7$, (*d*) $|\eta_{oh}| = |\eta_{og}| = |\eta_{gh}| = 0.9$.

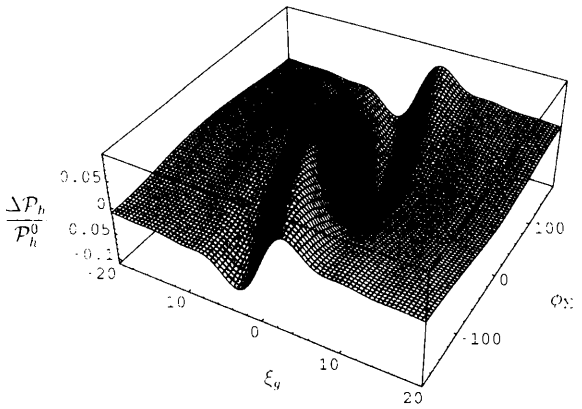

Fig. 5. Relative integrated power as a function of the invariant triplet phase sum and the normalized excitation error. Laue Laue case. $|\eta_{oh}| = |\eta_{og}| = |\eta_{gh}| = 0.1$.

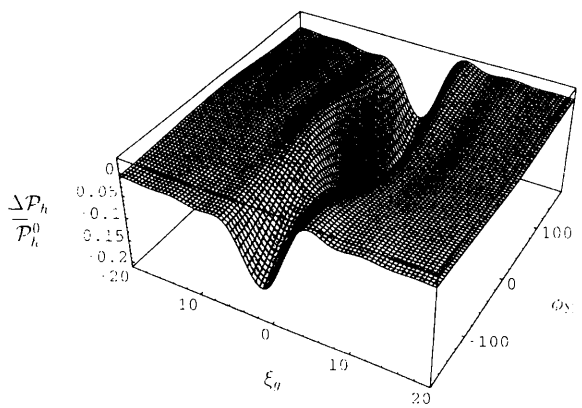

Fig. 6. Relative integrated power as a function of the invariant triplet phase sum and the normalized excitation error. Laue Laue case. $|\eta_{oh}| = 0.7$, $|\eta_{og}| = 0.1$, $|\eta_{gh}| = 0.7$; *i.e.* an *Aufhellung* situation.

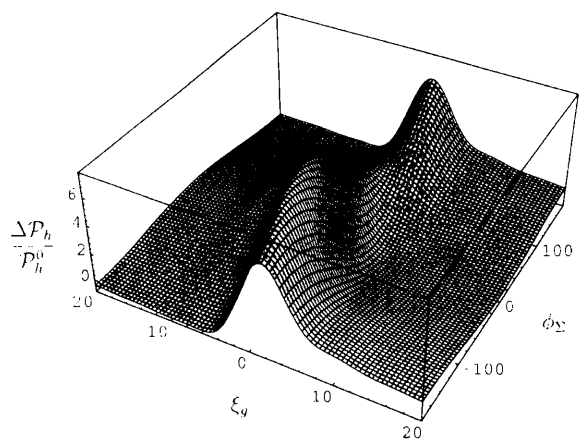

Fig. 7. Relative integrated power as a function of the invariant triplet phase sum and the normalized excitation error. Laue–Laue case. $|\eta_{oh}| = 0.1$, $|\eta_{og}| = |\eta_{gh}| = 0.7$; *i.e.* an *Umweganregung* situation.

curves, we do not see any significant differences for the two reflection geometries before $\{|\eta_{pq}|\}$ approach unity. Then, *Pendellösung* effects start to become important for the Laue transmission case, resulting in substantial deviations from the Bragg reflection case. This confirms previous findings (Hümmer *et al.*, 1990; Weckert & Hümmer, 1997). From (38) and (39), it is seen that the third-order terms in the perturbation-series solution are phase independent. They correspond to the scattering events $\kappa_{hg}\kappa_{gh}\kappa_{ho}$ and $\kappa_{ho}\kappa_{og}\kappa_{go}$ in equation (21) and consequently do not involve φ_Σ. *I.e.* for $|\eta_{pq}| < 1$ they represent the main contribution to the well known *Umweganregung* and *Aufhellung* effects. In order to keep such effects as small as possible, we should have $|\eta_{oh}|$, $|\eta_{hg}|$ and $|\eta_{og}|$ all of the same order of magnitude. This is shown in Fig. 5, where we have plotted the relative integrated power as a function of both the invariant phase sum and the deviation parameter, $\xi_g(\psi)$. We suppose that the magnitudes of the involved structure factors are the same and that *Aufhellung* (Fig. 6) and *Umweganregung* (Fig. 7) effects are generated by crystal shape anisotropy alone. These limitations, imposed on the $|\eta_{pq}|$ parameters, are in accordance with Weckert & Hümmer (1990) who, based on experimental experience, found that the ratio of polarization corrected structure factors, $|F_{hg}F_g|/|F_h^2|$, should stay within the range 2–6. However, based on our results, this implies that the crystal dimensions should be fairly isotropic. *I.e.* crystal geometry, in addition to structure-factor magnitudes, should also be considered when selecting suitable three-beam cases for investigation.

It seems to be an important point to stress that the use of the Takagi–Taupin equations shows that the η_{pq} parameters are the governing quantities regarding three-beam diffraction in finite perfect crystals. This implies that the well known *Umweganregung* and *Aufhellung* effects also have a geometrical aspect to be considered. Such effects arise even if the structure factors involved are of equal magnitudes, provided a sufficient crystal shape anisotropy is present.

References

Authier, A. (1996). *Proceedings, X-ray and Neutron Dynamical Diffraction. Theory and Applications*, edited by A. Authier, S. Lagomarsino & B. Tanner. *NATO ASI Series B: Physics*, Vol. 357, pp. 1–31. New York: Plenum Press.

Authier, A., Malgrange, C. & Tournarie, M. (1968). *Acta Cryst.* A**24**, 126–136.

Becker, P. (1977). *Acta Cryst.* A**33**, 667–671.

Becker, P. & Dunstetter, F. (1984). *Acta Cryst.* A**40**, 241–251.

Bremer, J. & Thorkildsen, G. (1986). *Acta Cryst.* A**42**, 191–197.

Chang, S.-L. (1984). *Multiple Diffraction of X-rays in Crystals*. Berlin: Springer Verlag.

Chang, S.-L. (1992). *Int. J. Mod. Phys.* B**6**, 2987–3020.

Colella, R. (1974). *Acta Cryst.* A**30**, 413–423.

Ewald, P. P. & Héno, Y. (1968). *Acta Cryst.* A**24**, 5–15.
Giacovazzo, C., Monaco, H. L., Viterbo, D., Scordari, F., Gilli, G., Zanotti, G. & Catti, M. (1992). *Fundamentals of Crystallography.* London: Oxford University Press.
Høier, R. & Marthinsen, K. (1983). *Acta Cryst.* A**39**, 854–860.
Hümmer, K. & Billy, H. (1982). *Acta Cryst.* A**38**, 841–848.
Hümmer, K. & Billy, H. (1986). *Acta Cryst.* A**42**, 127–133.
Hümmer, K. & Weckert, E. (1990). *Acta Cryst.* A**46**, 534–536.
Hümmer, K., Weckert, E. & Bondza, H. (1990). *Acta Cryst.* A**46**, 393–402.
Juretschke, H. J. (1984). *Acta Cryst.* A**40**, 379–389.
Kato, N. (1973). *Z. Naturforsch. Teil A*, **28**, 604–609.
Kato, N. (1976). *Acta Cryst.* A**32**, 453–457.
Larsen, H. B. & Thorkildsen, G. (1998). *Acta Cryst.* A**54**, 129–136.
Laue, M. von (1932). *Ergeb. Exakten Naturwiss.* **10**, 133–158.
Lipscomb, W. N. (1949). *Acta Cryst.* **2**, 193–194.
Myint-U, T. (1980). *Partial Differential Equations of Mathematical Physics.* New York: North Holland.
Pinsker, Z. G. (1978). *Dynamical Scattering of X-rays in Crystals.* Berlin: Springer-Verlag.
Sneddon, I. N. (1957). *Elements of Partial Differential Equations.* London: McGraw-Hill.
Sommerfeld, A. (1949). *Partial Differential Equations.* New York: Academic Press.
Takagi, S. (1962). *Acta Cryst.* **15**, 1311–1312.
Takagi, S. (1969). *J. Phys. Soc. Jpn*, **26**, 1239–1253.
Taupin, D. (1964). *Bull. Soc. Fr. Minéral. Cristallogr.* **87**, 469–511.
Thorkildsen, G. (1987). *Acta Cryst.* A**43**, 361–369.
Thorkildsen, G. (1990). *Working Papers from Rogaland University Centre*, **111**, 1–99.
Thorkildsen, G. & Larsen, H. B. (1997). *Two Beam Diffraction in Perfect Crystals Analyzed by Takagi's Equations.* Part 1. *Basic Principles and Methods of Solutions.* Technical Report 30, Stavanger College, Stavanger, Norway.
Weckert, E. & Hümmer, K. (1990). *Acta Cryst.* A**46**, 387–393.
Weckert, E. & Hümmer, K. (1997). *Acta Cryst.* A**53**, 108–143.