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Three-Beam Diffraction in Finite Imperfect Crystals. An Analysis of Coherent Wavefield Interactions Using Statistical Dynamical Theory

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Abstract

By applying the concepts of statistical dynamical theory and the boundary-value Green-function technique, an analytical expression in the form of a series expansion has been obtained for the coherent contribution to the integrated power in a finite crystal geometry. The solution suggests that three-beam interference is likely to cause observable perturbations also in crystals with a high degree of imperfection. Owing to a first-order dependence on the invariant triplet phase sum, profile reversal may occur in cases where this sum is close to 90°. The result may serve as an experimental check on statistical dynamical parameters.

1. Introduction

In a previous paper, we have investigated the influence of scattering geometry on the three-beam profiles for finite *perfect* crystals (Thorkildsen & Larsen, 1998), hereafter denoted TL-I. Such crystals represent idealized probing systems because the scattering is purely coherent – and it is the coherent wave fields that carry the invariant phase information.

However, for real structural studies, the crystals undergoing investigations are generally *not* perfect – *i.e.* they possess distortions of various kinds. Among the crystallographic community, the concept of 'mosaicity' (Darwin, 1922) has become popular in describing crystal imperfections – leading, in the two-beam case, to the division of multiple-scattering effects into primary and secondary extinction. Throughout the last decades, however, a lot of effort has been put into developing and improving the theories in order to obtain an overall unified description of extinction in a crystal of any degree of imperfection (Kato, 1976*a*, 1980*a* 1982; Al Haddad & Becker, 1988; Guigay & Chukhovskii, 1992; Becker & Al Haddad, 1992; Davis, 1994).

In the three-beam case, little work has been performed in order to establish/implement mathematical theories assessing the influence of crystal imperfection on for instance the ψ-scan profiles. Nevertheless, numerous experimental studies (Post, 1977; Chapman *et al.*, 1981; Chang, 1982; Thorkildsen & Mo, 1982, 1983; Hauback & Mo, 1988; Hümmer *et al.*, 1991; Weckert *et al.*, 1993) have shown that phase information indeed can be extracted from 'mosaic' crystals too.

Thorkildsen (Hauback *et al.*, 1990) adopted a procedure similar to that of Moon & Shull (1964) and solved the Zachariasen–Hamilton equations for the three-beam case in a finite model crystal, thereby obtaining a correction formalism for multiple-diffraction effects to be used in crystal structure analysis. Being based on intensity coupling equations, this accounts for the pure incoherent scattering only and thus cannot describe the characteristic asymmetry of the ψ-scan profiles. The mosaic crystal concept is implemented into the theory by assigning a distribution function, $W_\alpha(\Delta)$, for the excitation error† (Thorkildsen, 1983). $W_\alpha(\Delta)$, which may be taken as a Lorentzian or a Gaussian, thus describes the smearing effects due to crystal imperfection. However, using this approach, it has not been possible to assign *a single* distribution function that simultaneously accounts for the magnitude of the intensity perturbation for a large set of three-beam situations.

An interesting development is due to Kohn and co-workers (Kohn, 1988; Kohn & Samoilova, 1992). Here, three-beam diffraction concepts are used to obtain a depth profile of the crystal distortions. Crystal imperfections were introduced by assuming a homogeneous strain along the surface. Using an adequate mathematical model for the lattice phase factor (Afanasev & Kohn, 1971), the possibility of probing/measuring the deformation at a given depth of a crystal‡ was demonstrated by computer simulations. In this work, a weak secondary lattice node was excited within the angular region of total reflection for the primary diffracted beam.

Another approach, advocating the concept of the so-called 'virtual Bragg scattering' is due to Shen (1986). Here, a perturbative approach is devised but no attempt to describe the crystal imperfection is made.

In the present work, we will adopt the concepts of the statistical dynamical theory in describing the crystal

† Owing to the misalignment of the perfect mosaic blocks, we cannot associate a single excitation error with a reciprocal-lattice point. Δ is a measure of the deviation from the mean excitation error. The function $W_\alpha(\Delta)$ may also incorporate the effect of finite mosaic block sizes.

‡ Bragg–Bragg scattering geometry from a semi-infinite crystal plate is assumed.

imperfections. We will confine ourselves to a case where the approximations inherent in the Takagi–Taupin theory (Takagi, 1962, 1969; Taupin, 1964) are valid and we neglect effects arising from polarization and photoelectric absorption (Larsen & Thorkildsen, 1998). We consider the interaction due to the *coherent* wave fields alone. Additional contributions from incoherent scattering events are thus not taken into account. The present treatment should however warrant an analysis of the relative dynamical perturbations in ψ-scan profiles from imperfect crystals.

2. Elements of statistical dynamical theory

In 1980, Kato published the so-called statistical dynamical theory (Kato, 1980*a*,*b*,*c*) – an attempt to unify the theories describing primary and secondary extinction by treating crystal imperfections as 'statistical features' initiating incoherent scattering. The same author later (Kato, 1991, 1994) put forward a rigorous mathematical foundation for the theory. Several other authors (Al Haddad & Becker, 1988; Becker & Al Haddad, 1989; Guigay, 1989; Becker & Al Haddad, 1990, 1992; Guigay & Chukhovskii, 1992) have discussed and modified different aspects concerning Kato's original work.

The Takagi–Taupin equations for the three-beam case in an imperfect crystal may be written in the following manner (Larsen, 1997):

$$\begin{aligned}\partial\tilde{D}_o/\partial s_o &= i\varphi_{oh}\kappa_{oh}\tilde{D}_h + i\varphi_{og}\kappa_{og}\tilde{D}_g\\ \partial\tilde{D}_h/\partial s_h &= i\varphi_{ho}\kappa_{ho}\tilde{D}_o + i\varphi_{hg}\kappa_{hg}\tilde{D}_g\\ \partial\tilde{D}_g/\partial s_g &= i\varphi_{go}\kappa_{go}\tilde{D}_o + i\varphi_{gh}\kappa_{gh}\tilde{D}_h.\end{aligned} \tag{1}$$

The coupling parameters (neglecting polarization) are given by

$$\kappa_{pq} = (r_e\lambda/V_c)F_{p-q} = (1/\Lambda_{pq}), \tag{2}$$

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

The lattice phase factor has the following form:

$$\varphi_{pq} = \varphi_{pq}(\mathbf{r}) = \exp[-2\pi i(\mathbf{q}-\mathbf{p})\cdot\mathbf{u}(\mathbf{r})], \tag{3}$$

where $\mathbf{u}(\mathbf{r})$ is the displacement field at a position $\mathbf{r}$.

The amplitude transformation for the electrical displacement is given by $D_p = \tilde{D}_p\exp(2\pi i\sum_q \beta_q s_q)$ (Thorkildsen, 1990; Thorkildsen & Larsen, 1998). β_p is the deviation parameter:

$$\beta_p = \|\mathbf{k}_p\| - k = \alpha_p - K\delta\gamma_p - \tfrac{1}{2}K\chi_o. \tag{4}$$

$K = 1/\lambda$ is the wave number of the incoming vacuum wave, $\mathbf{K}_o$. $\mathbf{k}_p$ is a crystal wavevector and $\|\mathbf{k}_p\|$ indicates its norm. $\mathbf{p}$ is a reciprocal-lattice vector. $k = K(1+\frac{1}{2}\chi_o)$ is the mean wavevector within the crystal. $\alpha_p = \|\mathbf{K}_p\| - K$ is the excitation error associated with reflection p, whereas δ, the *Anpassung*, is treated in the kinematical limit by putting $\delta = -\chi_o/2\gamma_o$. Thus, $\beta_o = \alpha_o = 0$ (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 a unit normal vector to the entrance surface, directed into the crystal. $\hat{\mathbf{s}}_p$ is a unit vector parallel to $\mathbf{K}_p = \mathbf{K}_o + \mathbf{p}$ and s_p is a positional coordinate measured along $\hat{\mathbf{s}}_p$.

It is convenient to separate the contributions to the electrical displacement into two parts: $\langle\tilde{D}_p\rangle$, which represents the ensemble average; and $\delta\tilde{D}_p$, representing the 'fluctuating' part. *I.e.*

$$\tilde{D}_p = \langle\tilde{D}_p\rangle + \delta\tilde{D}_p \tag{5}$$

with $p \in \{o, h, g\}$ and $\langle\delta\tilde{D}_p\rangle = 0$. The lattice phase factor may be split up in a similar way:

$$\varphi_{pq} = \langle\varphi_{pq}\rangle + \delta\varphi_{pq}. \tag{6}$$

The 'intensity' is then defined ($\stackrel{\text{def}}{=}$) by

$$I_p \stackrel{\text{def}}{=} \langle|\tilde{D}_p|^2\rangle = |\langle\tilde{D}_p\rangle|^2 + \langle|\delta\tilde{D}_p|^2\rangle = I_p^c + I_p^i, \tag{7}$$

where I_p^c and I_p^i are the *coherent* and *incoherent* intensities, respectively.

The incoherent intensity may be built either from incoherent scattering contributions in the vicinity of the entrance surface or partly from coherent contributions where phase correlation is subsequently lost in scattering events following the propagation of the beams within the crystal. Following Al Haddad & Becker (1988), Becker & Al Haddad (1992) and Kato (1980*c*), the incoherent part may again be split into a finite number of different terms (n) according to:

$$I_p^i = I_p^{i,0} + \sum_n I_p^{i,n}, \tag{8}$$

where $I_p^{i,0}$ is the purely incoherent contribution and $I_p^{i,n}$ is the *mixed* coherent and incoherent contributions ($n \neq 0$).

The integrated power is similarly decomposed according to

$$\mathcal{P}_p = \mathcal{P}_p^c + \mathcal{P}_p^i, \tag{9}$$

in which the incoherent integrated power may be partitioned:

$$\mathcal{P}_p^i = \mathcal{P}_p^{i,0} + \mathcal{M}_p, \tag{10}$$

where $\mathcal{M}_p = \sum_n \mathcal{M}_p^n$ corresponds to the mixed terms.

In analogy with Kato (1980*b*), we introduce a generalized *long-range-order* parameter, E_{pq} (often called 'the static Debye–Waller factor'):

$$E_{pq} = \langle\varphi_{pq}\rangle = (1/v)\int_v \mathrm{d}\mathbf{r}\,\varphi_{pq}(\mathbf{r}), \tag{11}$$

v being the crystal volume. According to Becker & Al

Haddad (1989), equation (11) may be written as

$$E_{pq} = \int \mathrm{d}\mathbf{u}\, p(\mathbf{u}) \exp[-2\pi i(\mathbf{q} - \mathbf{p}) \cdot \mathbf{u}], \qquad (12)$$

where $p(\mathbf{u})$ is the probability density function† of the displacement field, $\mathbf{u}$. Generally, we have $E_{pq} \leq 1$. The value $E_{pq} = 1$ corresponds to the perfect-crystal case, while the limit $E_{pq} \to 0$ represents the 'ideal imperfect' crystal.

It is appropriate also to introduce a spatial correlation function of the phases, $f_{pq,rs}(\mathbf{t})$, which is assumed to be a real function (Becker & Al Haddad, 1989):

$$f_{pq,rs}(\mathbf{t}) \overset{\text{def}}{=} (1/v) \int_v \mathrm{d}\mathbf{r}\, \varphi_{pq}(\mathbf{r}) \varphi^*_{rs}(\mathbf{r} + \mathbf{t})$$
$$= \langle \varphi_{pq}(\mathbf{r}) \varphi^*_{rs}(\mathbf{r} + \mathbf{t}) \rangle, \qquad (13)$$

where $p, q, r, s \in \{o, g, h\}$. If $t = |\mathbf{t}|$ is small compared with the crystal dimensions, this may be written (Becker & Al Haddad, 1989, 1990)

$$f_{pq,rs}(\mathbf{t}) \approx E_{pq}E_{rs} + \langle \delta\varphi_{pq}(\mathbf{r}) \delta\varphi^*_{rs}(\mathbf{r} + \mathbf{t}) \rangle$$
$$\overset{\text{def}}{=} E_{pq}E_{rs} + (1 - E_{pq}E_{rs}) g_{pq,rs}(\mathbf{t}). \qquad (14)$$

$g_{pq,rs}(\mathbf{t})$, the intrinsic correlation function (Kato, 1980*a*) satisfies $g_{pq,rs}(0) = 1$.

A statistical hypothesis is then commonly introduced, stating that the diffracted power does not depend on the details of the displacement field $\mathbf{u}(\mathbf{r})$ but only on its statistical properties. This implies the existence of an ensemble of (hypothetical) crystals having *different* displacement fields but the same distribution function, $p(\mathbf{u})$, and intrinsic correlation function, $g_{pq,rs}(\mathbf{t})$. If such an assumption is valid, it is possible to describe the intensities of the transmitted and diffracted wave fields as an average over all crystals in the ensemble. $\varphi_{pq}(\mathbf{r})$ is thus classified as an ergodic and homogeneous stochastic process (Ross, 1993).

† Usually taken as a Gaussian.

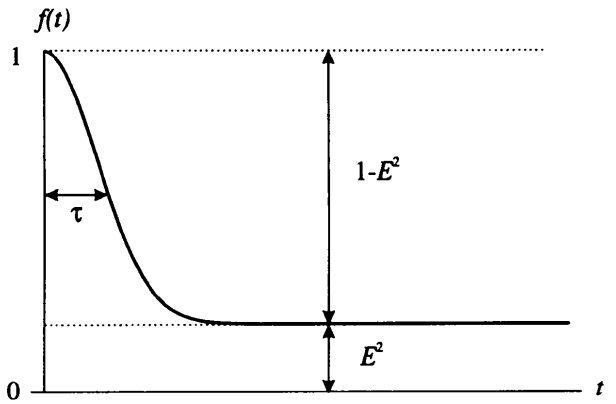

Fig. 1. Schematic behaviour of the correlation function. Short-range and long-range parameters. After Kato (1982).

Finally, assuming spatial homogeneity, $g_{pq,rs}(\mathbf{t}) \to g_{pq,rs}(t)$ and $p(\mathbf{u}) \to p(u)$, it is convenient to introduce the short-range parameter, $\tau_{pq,rs}$, defined by

$$\tau_{pq,rs} = \int_0^\infty \mathrm{d}t\, g_{pq,rs}(t). \qquad (15)$$

τ represents the distance along which different optical routes lose their mutual phase coherence. The relation between τ, E and $f(t)$ is schematically illustrated in Fig. 1. According to Becker & Al Haddad (1989), the decomposition scheme of equation (14) is possible for crystals with a high degree of perfection. In that case, the correlation length $\tau_{pq,rs}$ becomes a constant, τ.

3. Propagation equations for the coherent waves in the three-beam case

From equations (6) and (11), we may straightforwardly write the Takagi–Taupin equations for the coherent part of the diffracted wave fields in the three-beam case:

$$\partial\langle\tilde{D}_o\rangle/\partial s_o = i\kappa_{oh}E_{oh}\langle\tilde{D}_h\rangle + i\kappa_{oh}\langle\delta\varphi_{oh}\tilde{D}_h\rangle$$
$$+ i\kappa_{og}E_{og}\langle\tilde{D}_g\rangle + i\kappa_{og}\langle\delta\varphi_{og}\tilde{D}_g\rangle \qquad (16)$$
$$\partial\langle\tilde{D}_h\rangle/\partial s_h = i\kappa_{ho}E_{ho}\langle\tilde{D}_o\rangle + i\kappa_{ho}\langle\delta\varphi_{ho}\tilde{D}_o\rangle$$
$$+ i\kappa_{hg}E_{hg}\langle\tilde{D}_g\rangle + i\kappa_{hg}\langle\delta\varphi_{hg}\tilde{D}_g\rangle \qquad (17)$$
$$\partial\langle\tilde{D}_g\rangle/\partial s_g = i\kappa_{go}E_{go}\langle\tilde{D}_o\rangle + i\kappa_{go}\langle\delta\varphi_{go}\tilde{D}_o\rangle$$
$$+ i\kappa_{gh}E_{gh}\langle\tilde{D}_h\rangle + i\kappa_{gh}\langle\delta\varphi_{gh}\tilde{D}_h\rangle. \qquad (18)$$

The boundary-value Green functions for the set of coupled first-order partial differential equations may be obtained by integration. *E.g.* for $\langle\tilde{D}_o\rangle$:

$$\langle\tilde{D}_o(s_o, s_h, s_g)\rangle$$
$$= \delta(s_h)\delta(s_g) + i\kappa_{oh}E_{oh} \int_{s_o^b(s_h,s_g)}^{s_o} \mathrm{d}s'_o\, \langle\tilde{D}_h(s'_o, s_h, s_g)\rangle$$
$$+ i\kappa_{oh} \int_{s_o^b(s_h,s_g)}^{s_o} \mathrm{d}s'_o\, \langle\delta\varphi_{oh}(s'_o, s_h, s_g)\tilde{D}_h(s'_o, s_h, s_g)\rangle$$
$$+ i\kappa_{og}E_{og} \int_{s_o^b(s_h,s_g)}^{s_o} \mathrm{d}s'_o\, \langle\tilde{D}_g(s'_o, s_h, s_g)\rangle$$
$$+ i\kappa_{og} \int_{s_o^b(s_h,s_g)}^{s_o} \mathrm{d}s'_o\, \langle\delta\varphi_{og}(s'_o, s_h, s_g)\tilde{D}_g(s'_o, s_h, s_g)\rangle. \qquad (19)$$

s_p^b denotes a boundary point. Analogous expressions are obtained for $\langle\tilde{D}_h(s_o, s_h, s_g)\rangle$ and $\langle\tilde{D}_g(s_o, s_h, s_g)\rangle$. We have applied a point source of unit strength at the entrance surface associated with the incident-beam direction, $\mathbf{K}_o$.

Thus,

$$\begin{aligned}\langle \tilde{D}_o(s_o^b(s_h, s_g), s_h, s_g)\rangle &= \delta(s_h)\delta(s_g)\\ \langle \tilde{D}_h(s_o, s_h^b(s_o, s_g), s_g)\rangle &= 0\\ \langle \tilde{D}_g(s_o, s_h, s_g^b(s_o, s_h))\rangle &= 0.\end{aligned} \tag{20}$$

Using (19) and the corresponding integral expressions for $\langle \tilde{D}_h(s_o, s_h, s_g)\rangle$ and $\langle \tilde{D}_g(s_o, s_h, s_g)\rangle$ in equations (16)–(18), we obtain after a lengthy but straightforward calculation (Larsen, 1997) for the term $i\kappa_{oh}\langle \delta\varphi_{oh}\tilde{D}_h\rangle$:

$$\begin{aligned}&i\kappa_{oh}\langle \delta\varphi_{oh}\tilde{D}_h\rangle\\ &= -\kappa_{oh}\kappa_{ho}E_{ho}\int_{s_h^b(s_o,s_g)}^{s_h} \mathrm{d}s_h' \,\langle \delta\varphi_{oh}(s_o, s_h, s_g)\tilde{D}_o(s_o, s_h', s_g)\rangle\\ &\quad -\kappa_{oh}\kappa_{ho}\int_{s_h^b(s_o,s_g)}^{s_h} \mathrm{d}s_h' \,\langle \delta\varphi_{oh}(s_o, s_h, s_g)\delta\varphi_{ho}(s_o, s_h', s_g)\\ &\quad \times \tilde{D}_o(s_o, s_h', s_g)\rangle\\ &\quad -\kappa_{oh}\kappa_{hg}E_{hg}\int_{s_h^b(s_o,s_g)}^{s_h} \mathrm{d}s_h' \,\langle \delta\varphi_{oh}(s_o, s_h, s_g)\tilde{D}_g(s_o, s_h', s_g)\rangle\\ &\quad -\kappa_{oh}\kappa_{hg}\int_{s_h^b(s_o,s_g)}^{s_h} \mathrm{d}s_h' \,\langle \delta\varphi_{oh}(s_o, s_h, s_g)\delta\varphi_{hg}(s_o, s_h', s_g)\\ &\quad \times \tilde{D}_g(s_o, s_h', s_g)\rangle.\end{aligned} \tag{21}$$

In order to get a set of equations that is easier to handle, some approximations are called for. The values of $\{\tilde{D}_p(s_o, s_h, s_g)\}$ are determined by the scattering events along the successive positions (s_o', s_h', s_g') along the optical route through the crystal. Following Becker & Al Haddad (1992), phase coherence is assumed to be lost outside nearest-neighbour scattering points. This approximation allows us to neglect the correlation between the fields and the phase fluctuations, hence we omit all terms proportional to E_{pq} in (21):

$$\begin{aligned}&i\kappa_{oh}\langle \delta\varphi_{oh}\tilde{D}_h\rangle\\ &\approx -\kappa_{oh}\kappa_{ho}\langle \tilde{D}_o\rangle(1 - E_{oh}E_{ho})\int_{s_h^b(s_o,s_g)}^{s_h} \mathrm{d}s_h' \, g_{oh,ho}(s_h')\\ &\quad -\kappa_{oh}\kappa_{hg}\langle \tilde{D}_g\rangle(1 - E_{oh}E_{hg})\int_{s_h^b(s_o,s_g)}^{s_h} \mathrm{d}s_h' \, g_{oh,hg}(s_h').\end{aligned} \tag{22}$$

We further assume $\{\langle \tilde{D}_p\rangle\}$ to have negligible variations over a distance $\tau_{pq,rs}$ and perform the integrations in equation (22) using the assumption $(s_p - s_p^b) > \tau_{pq,rs}$, *cf.* Guigay & Chukhovskii (1992) for a more thorough discussion of this approximation. By applying the definition of $\tau_{pq,rs}$, *cf.* (15), we may then write:

$$\begin{aligned}i\kappa_{oh}\langle \delta\varphi_{oh}\tilde{D}_h\rangle \approx\ &-\kappa_{oh}\kappa_{ho}\langle \tilde{D}_o\rangle(1 - E_{oh}E_{ho})\tau_{oh,ho}\\ &-\kappa_{oh}\kappa_{hg}\langle \tilde{D}_g\rangle(1 - E_{oh}E_{hg})\tau_{oh,hg}.\end{aligned} \tag{23}$$

Analogous expressions may be obtained for the other members of the set $\{i\kappa_{pq}^b\langle \delta\varphi_{pq}\tilde{D}_q\rangle\}$. In order to further simplify the mathematical treatment, we suppose that the long- and short-range parameters may be treated as isotropic quantities with respect to the involved reflections. *I.e.* $E_{pq} \to E$ and $\tau_{pq,rs} \to \tau$. By inserting these expressions into equations (16)–(18), we obtain the statistical dynamical formulation of the Takagi–Taupin equations, which, within the given approximations are:

$$\begin{aligned}\partial\langle \tilde{D}_o\rangle/\partial s_o = {}& i\kappa_{oh}E\langle \tilde{D}_h\rangle - (1 - E^2)\tau(\kappa_{oh}\kappa_{ho}\langle \tilde{D}_o\rangle\\ &+ \kappa_{oh}\kappa_{hg}\langle \tilde{D}_g\rangle) + i\kappa_{og}E\langle \tilde{D}_g\rangle - (1 - E^2)\\ &\times \tau(\kappa_{og}\kappa_{go}\langle \tilde{D}_o\rangle + \kappa_{og}\kappa_{gh}\langle \tilde{D}_h\rangle)\end{aligned} \tag{24}$$

$$\begin{aligned}\partial\langle \tilde{D}_h\rangle/\partial s_h = {}& i\kappa_{ho}E\langle \tilde{D}_o\rangle - (1 - E^2)\tau(\kappa_{ho}\kappa_{oh}\langle \tilde{D}_h\rangle\\ &+ \kappa_{ho}\kappa_{og}\langle \tilde{D}_g\rangle) + i\kappa_{hg}E\langle \tilde{D}_g\rangle - (1 - E^2)\\ &\times \tau(\kappa_{hg}\kappa_{gh}\langle \tilde{D}_h\rangle + \kappa_{hg}\kappa_{go}\langle \tilde{D}_o\rangle)\end{aligned} \tag{25}$$

$$\begin{aligned}\partial\langle \tilde{D}_g\rangle/\partial s_g = {}& i\kappa_{go}E\langle \tilde{D}_o\rangle - (1 - E^2)\tau(\kappa_{go}\kappa_{og}\langle \tilde{D}_g\rangle\\ &+ \kappa_{go}\kappa_{oh}\langle \tilde{D}_h\rangle) + i\kappa_{gh}E\langle \tilde{D}_h\rangle - (1 - E^2)\\ &\times \tau(\kappa_{gh}\kappa_{hg}\langle \tilde{D}_g\rangle + \kappa_{gh}\kappa_{ho}\langle \tilde{D}_o\rangle).\end{aligned} \tag{26}$$

The condition of validity of equations (24)–(26) becomes (Becker & Al Haddad, 1992)

$$\tau \ll \Lambda_{pq}/E. \tag{27}$$

Equations (24)–(26) may more conveniently be written in the following way:

$$\begin{aligned}\partial\tilde{D}_o/\partial s_o = {}& (i\kappa_{oh}E - \kappa_{og}\kappa_{gh}\mu_e)\tilde{D}_h + (i\kappa_{og}E - \kappa_{oh}\kappa_{hg}\mu_e)\tilde{D}_g\\ &- (\kappa_{oh}\kappa_{ho} + \kappa_{og}\kappa_{go})\mu_e\tilde{D}_o\end{aligned} \tag{28}$$

$$\begin{aligned}\partial\tilde{D}_h/\partial s_h = {}& (i\kappa_{ho}E - \kappa_{hg}\kappa_{go}\mu_e)\tilde{D}_o + (i\kappa_{hg}E - \kappa_{ho}\kappa_{og}\mu_e)\tilde{D}_g\\ &- (\kappa_{ho}\kappa_{oh} + \kappa_{hg}\kappa_{gh})\mu_e\tilde{D}_h\end{aligned} \tag{29}$$

$$\begin{aligned}\partial\tilde{D}_g/\partial s_g = {}& (i\kappa_{go}E - \kappa_{gh}\kappa_{ho}\mu_e)\tilde{D}_o + (i\kappa_{gh}E - \kappa_{go}\kappa_{oh}\mu_e)\tilde{D}_h\\ &- (\kappa_{go}\kappa_{og} + \kappa_{gh}\kappa_{hg})\mu_e\tilde{D}_g.\end{aligned} \tag{30}$$

From this stage, we omit the $\langle\rangle$ brackets – the displacement amplitudes are implicitly considered to be ensemble averaged quantities. We have also defined the 'effective absorption coefficient', μ_e, having the dimension of length:

$$\mu_e \stackrel{\mathrm{def}}{=} (1 - E^2)\tau. \tag{31}$$

We then introduce the following transformation for the displacement amplitudes:

$$\tilde{D}_r = \mathsf{D}_r \exp\left[-\mu_e \sum_p \sum_{q\neq p} \kappa_{pq}\kappa_{qp}s_p\right]. \tag{32}$$

Applying this to equations (28)–(30), we get:

$$\partial \mathsf{D}_o/\partial s_o = (i\kappa_{oh}E - \kappa_{og}\kappa_{gh}\mu_e)\mathsf{D}_h + (i\kappa_{og}E - \kappa_{oh}\kappa_{hg}\mu_e)\mathsf{D}_g \quad (33)$$

$$\partial \mathsf{D}_h/\partial s_h = (i\kappa_{ho}E - \kappa_{hg}\kappa_{go}\mu_e)\mathsf{D}_o + (i\kappa_{hg}E - \kappa_{ho}\kappa_{og}\mu_e)\mathsf{D}_g \quad (34)$$

$$\partial \mathsf{D}_g/\partial s_g = (i\kappa_{go}E - \kappa_{gh}\kappa_{ho}\mu_e)\mathsf{D}_o + (i\kappa_{gh}E - \kappa_{go}\kappa_{oh}\mu_e)\mathsf{D}_h. \quad (35)$$

The equations are compactly written as

$$\begin{aligned}\partial \mathsf{D}_o/\partial s_o &= i\kappa_{oh}\Gamma_u^{(-)}\mathsf{D}_h + i\kappa_{og}\Gamma_v^{(+)}\mathsf{D}_g\\ \partial \mathsf{D}_h/\partial s_h &= i\kappa_{ho}\Gamma_u^{(+)}\mathsf{D}_o + i\kappa_{hg}\Gamma_w^{(-)}\mathsf{D}_g\\ \partial \mathsf{D}_g/\partial s_g &= i\kappa_{go}\Gamma_v^{(-)}\mathsf{D}_o + i\kappa_{gh}\Gamma_w^{(+)}\mathsf{D}_h\end{aligned} \quad (36)$$

with

$$\Gamma_m^{(+)} = E + i(T/m)\mu_e \quad (37)$$

$$\Gamma_m^{(-)} = E + i(T^*/m)\mu_e, \quad (38)$$

where $m \in \{u, v, w\}$,

$$\begin{aligned}u &= \kappa_{oh}\kappa_{ho}\\ v &= \kappa_{og}\kappa_{go}\\ w &= \kappa_{hg}\kappa_{gh},\end{aligned} \quad (39)$$

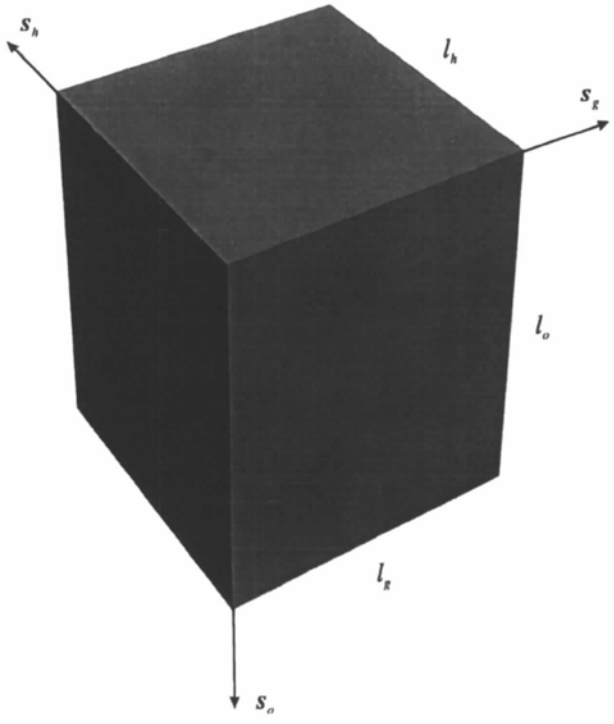

Fig. 2. Scattering and crystal geometry for the Laue–Laue case.

and, neglecting resonant scattering,

$$T = \kappa_{oh}\kappa_{hg}\kappa_{go} = |\kappa_{hg}||\kappa_{go}||\kappa_{oh}|\exp(i\phi_\Sigma), \quad (40)$$

ϕ_Σ being the invariant triplet phase sum:

$$\phi_\Sigma \stackrel{\text{def}}{=} \phi_{oh} + \phi_{hg} + \phi_{go}. \quad (41)$$

The field amplitudes are then formally obtained by integration.

In direct analogy with TL-I, it is convenient to introduce a set of linear operators $\mathcal{L}_{pq}^{m(\mathsf{s})}$, where $(\mathsf{s}) \in \{(+), (-)\}$:

$$\mathcal{L}_{pq}^{m(\mathsf{s})}\mathsf{D}_q(s_o, s_h, s_g) = i\kappa_{pq}\Gamma_m^{(\mathsf{s})}\int_{s_p^b}^{s_p} \mathrm{d}s_p'\, \mathsf{D}_q(s_p', \{s_q\}). \quad (42)$$

The solutions may therefore be expressed in the following form:

$$\begin{aligned}\mathsf{D}_o &= \mathsf{D}_o^{(b)} + \mathcal{L}_{oh}^{u(-)}\mathsf{D}_h + \mathcal{L}_{og}^{v(+)}\mathsf{D}_g\\ \mathsf{D}_h &= \mathsf{D}_h^{(b)} + \mathcal{L}_{ho}^{u(+)}\mathsf{D}_o + \mathcal{L}_{hg}^{w(-)}\mathsf{D}_g\\ \mathsf{D}_g &= \mathsf{D}_g^{(b)} + \mathcal{L}_{go}^{v(-)}\mathsf{D}_o + \mathcal{L}_{gh}^{w(+)}\mathsf{D}_h\end{aligned} \quad (43)$$

with boundary conditions identical to (20).

The set of equations was solved for a finite crystal Laue–Laue case, *cf.* Fig. 2, by the method presented in TL-I. That is, by obtaining a series expansion for the boundary-value Green functions and performing integrations term by term over the entrance and exit surfaces of the crystal. An extensive coding scheme was adopted for *MATHEMATICA*† (Thorkildsen, 1990; Larsen, 1997) in order to perform these operations and ease the subsequent algebraic manipulations. A detailed description for the present case can be found in Larsen (1997).

4. Results

The results presented below are in terms of *integrated power*, $\mathcal{P}_h$, where an additional integration over the deviation from the Bragg condition, $\Delta\theta_{oh}$, for the power, P_h, of the primary reflection has been carried out (Thorkildsen & Larsen, 1998).

$$\mathcal{P}_h = \int_{-\infty}^{\infty} \mathrm{d}\Delta\theta_{oh}\, P_h(\Delta\theta_{oh}). \quad (44)$$

Owing to the increasing complexity, the series expansion was terminated after the third order – thus yielding the important *Aufhellung* and *Umweganregung* terms.

It is convenient to express the integrated power in the following way:

$$\mathcal{P}_h = I_o|\kappa_{ho}|^2\nu_L l_h(1/m_o) \times [1 - \exp(-m_o)]\{\wp^{(1)} + \wp^{(2)} + \wp^{(3)}\}. \quad (45)$$

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

Here, $I_o = (c/2\varepsilon_0)|D_0^{(e)}|^2$ is the intensity of the incident beam. v_L is the volume of the Laue–Laue crystal. $(1/m_o)[1 - \exp(-m_o)]$, with $m_o = \mu_e l_o(u + v)$, is a statistical dynamical pre-factor, which equals unity in the perfect-crystal case. l_o, l_h and l_g are the crystal dimensions as indicated in Fig. 2. $\{\wp^{(\cdot)}\}$ denotes the expansion terms, in which contributions to extinction have been left out. The third-order term is quite lengthy, thus, for convenience, only $\wp^{(1)}$ and $\wp^{(2)}$ will be presented here – a *MATHEMATICA*-coded expression for $\wp^{(3)}$ is available from the authors upon request.

$$\wp_h^{(1)} = j_1^c[E^2 + (|\eta_{hg}|^2|\eta_{go}|^2/|\eta_{ho}|^2)\mu_g^2 - 2E(|\eta_{hg}||\eta_{go}|/|\eta_{ho}|)\mu_g \sin\phi_\Sigma] \quad (46)$$

$$\begin{aligned}\wp_h^{(2)} = 2j_2^c\{&-E^3(|\eta_{hg}||\eta_{go}|/|\eta_{ho}|) \\ &\times [g_1^c(\xi_g)\cos\phi_\Sigma + g_2^c(\xi_g)\sin\phi_\Sigma] \\ &- E[(|\eta_{hg}|^3|\eta_{go}|/|\eta_{ho}|)\mu_g\mu_h \\ &+ (|\eta_{hg}||\eta_{go}|^3/|\eta_{ho}|)\mu_g\mu_o - |\eta_{hg}||\eta_{go}||\eta_{oh}|\mu_h\mu_o] \\ &\times [g_1^c(\xi_g)\cos\phi_\Sigma - g_2^c(\xi_g)\sin\phi_\Sigma] \\ &+ E^2[(|\eta_{hg}|^2|\eta_{go}|^2/|\eta_{ho}|^2)\mu_g - |\eta_{hg}|^2\mu_h \\ &- |\eta_{go}|^2\mu_o]g_2^c(\xi_g) - |\eta_{hg}|^2|\eta_{go}|^2\mu_g\mu_h\mu_o \\ &\times [g_2^c(\xi_g)\cos 2\phi_\Sigma + g_1^c(\xi_g)\sin 2\phi_\Sigma]\}, \end{aligned} \quad (47)$$

where $|\eta_{pq}| = |\kappa_{pq}|(l_p l_q)^{1/2}$ and $\mu_p = \mu_e/l_p$. The g^c functions are given by

$$\begin{aligned} g_1^c(\xi_g) = &[(-2m_g\xi_g + m_g^2\xi_g + \xi_g^3)\exp(m_g) \\ &+ (m_g^2 - \xi_g^2)\sin\xi_g + 2m_g\xi_g\cos\xi_g] \\ &\times [(m_g^2 + \xi_g^2)^2\exp(m_g)]^{-1} \end{aligned} \quad (48)$$

$$\begin{aligned} g_2^c(\xi_g) = &[(-m_g^2 + m_g^3 + \xi_g^2 + m_g\xi_g^2)\exp(m_g) \\ &+ (m_g^2 - \xi_g^2)\cos\xi_g - 2m_g\xi_g\sin\xi_g] \\ &\times [(m_g^2 + \xi_g^2)^2\exp(m_g)]^{-1}, \end{aligned} \quad (49)$$

where $\xi_g = 2\pi\alpha_g l_g$ and $m_g = \mu_e l_g(v + w)$.

The j^c parameters are:

$$\begin{aligned} j_1^c &= (\pi/m_h)\{1 + \exp(-2m_h) \\ &\quad - 2\exp(-m_h)(\cosh m_h - \sinh m_h)\} \\ j_2^c &= (\pi/m_h)\{(1/2m_h) + (1 + 1/2m_h)\exp(-2m_h) \\ &\quad + (2 + 1/m_h)(\sinh m_h - \cosh m_h)\exp(-m_h)\}, \end{aligned}$$

where $m_h = \mu_e l_h(u + w)$.

The above expressions approach the correct perfect-crystal values (Thorkildsen & Larsen, 1998) when $E \to 1$.

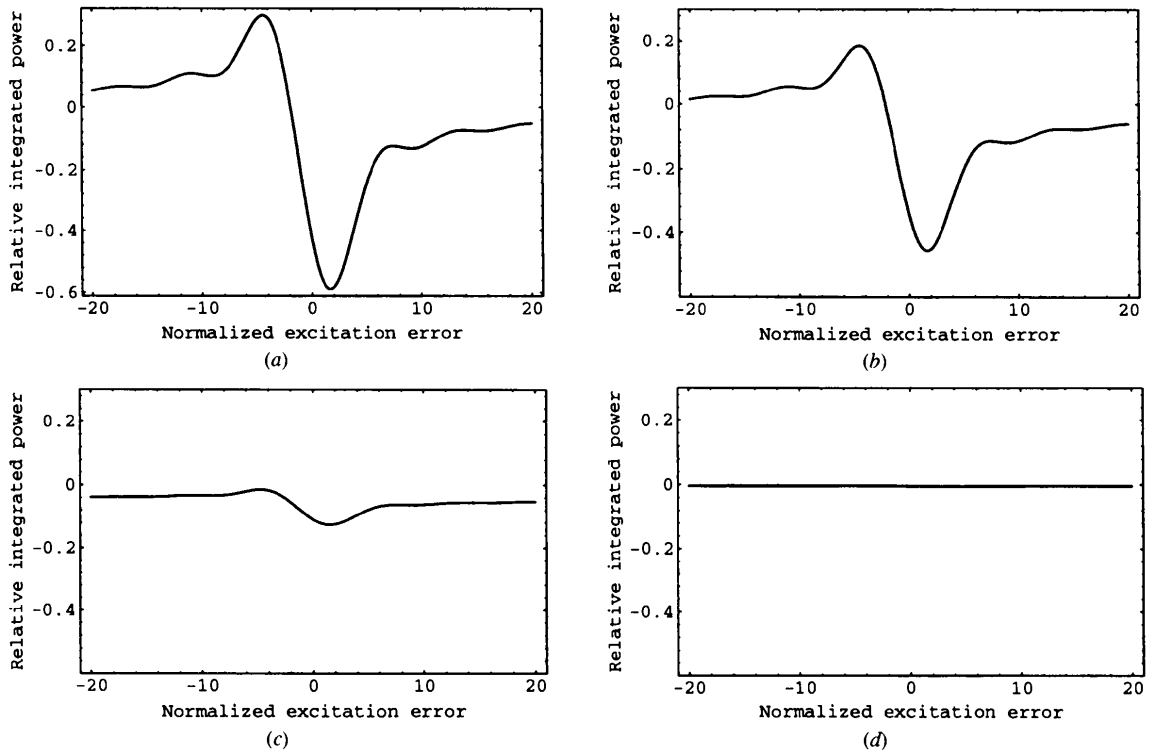

Fig. 3. Relative integrated power as a function of the normalized excitation error, $\xi_g = 2\pi\alpha_g l_g$, for different long-range parameters, E. $|\eta_{oh}| = 0.1$, $|\eta_{go}| = |\eta_{hg}| = 0.28$. The short-range parameter is chosen such that $\tau/\Lambda_{oh} = 0.1$ and the invariant triplet phase sum is $\phi_\Sigma = 45°$. (*a*) $E = 1$, $\mu_o = \mu_h = \mu_g = 0$; (*b*) $E = 0.9$, $\mu_o = \mu_h = 0.19$, $\mu_g = 0.024$; (*c*) $E = 0.5$, $\mu_o = \mu_h = 0.75$, $\mu_g = 0.093$; (*d*) $E = 0.1$, $\mu_o = \mu_h = 0.99$, $\mu_g = 0.12$.

5. Discussion

Using the above results, we may assess the influence of crystal imperfection on the ψ profiles. The plots are given in terms of integrated power relative to the kinematical value. However, since the incoherent parts of the scattering are not taken into consideration in the present treatment, this does not ensure a relative two-beam level at zero – as for the perfect-crystal case.

It is clear that the dynamical perturbation due to three-beam interaction will decrease as the crystal imperfection increases. This is illustrated in Fig. 3 for a crystal having different values of the long-range parameter E. The calculations are carried out for an anisotropic sample in which $l_o = l_h = \frac{1}{8} l_g$ but where the intrinsic strengths of the reflections (*i.e.* the κ_{pq} parameters) are the same. The profile in Fig. 3(*a*) represents the perfect-crystal case in which the scattering is purely coherent. Reducing the value of E, we simultaneously increase the values of the 'effective absorption' parameters μ_o, μ_g, μ_h. For the case of $E = 0.9$ (Fig. 3*b*), representing a slightly distorted crystal, we notice the resulting shift towards decreasing values of relative integrated power. This is due to the

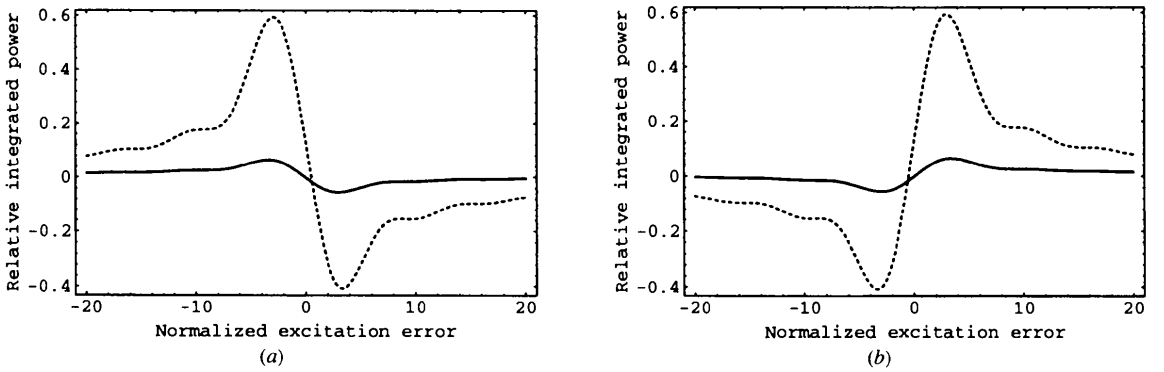

Fig. 4. ψ profiles for a centrosymmetric case. $|\eta_{oh}| = 0.1$, $|\eta_{go}| = |\eta_{hg}| = 0.28$. Dashed line: perfect-crystal case ($E = 1$); solid line: distorted-crystal case ($E = 0.5$, $\tau/\Lambda_{oh} = 0.1$). (*a*) $\phi_\Sigma = 0°$, (*b*) $\phi_\Sigma = 180°$.

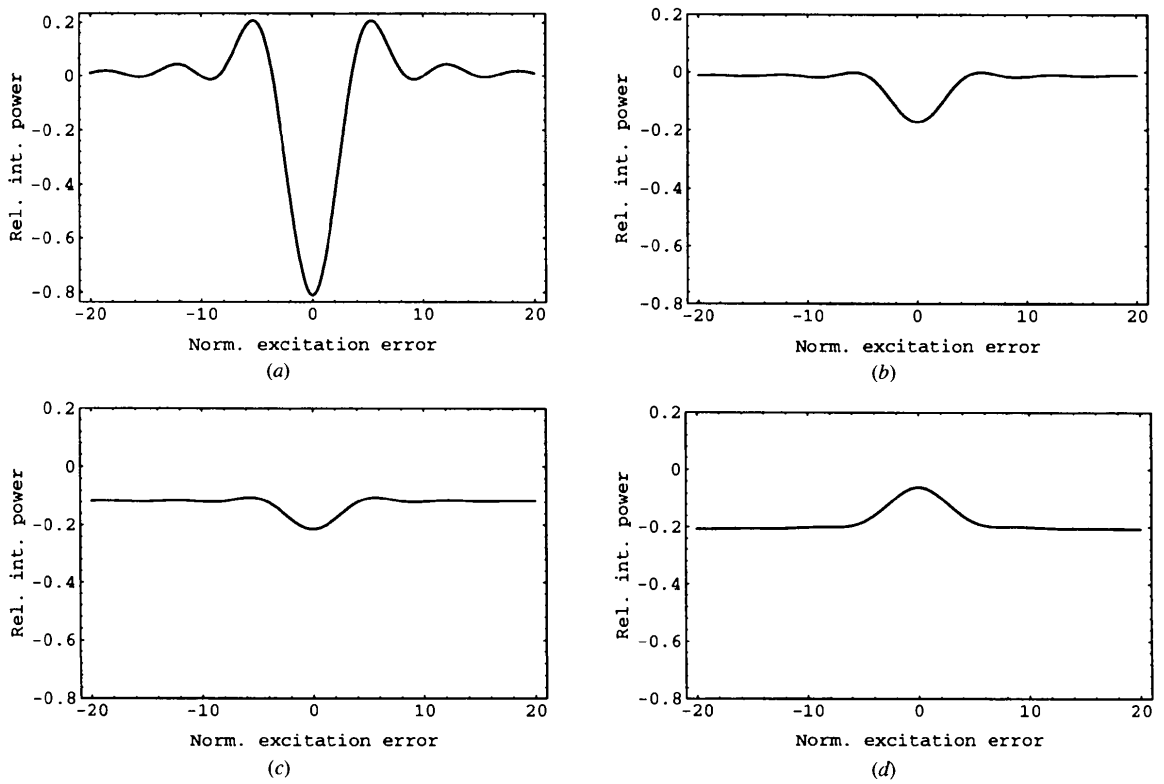

Fig. 5. Effect of increasing the short-range parameter. Isotropic crystal shape ($l_o = l_h = l_g = l$). $|\eta_{oh}| = 0.05$, $|\eta_{go}| = |\eta_{hg}| = 0.3$. Triplet phase sum $\phi_\Sigma = 90°$. (*a*) Perfect-crystal reference case. (*b*) $E = 0.5$, $\tau/l = 0.01$; (*c*) $E = 0.5$, $\tau/l = 0.1$; (*d*) $E = 0.5$, $\tau/l = 0.5$.

missing incoherent contributions. The profile asymmetry is fading but it should in principle be possible to extract reliable phase information for this case. When further reducing the value of E to 0.5 (Fig. 3*c*), the dynamical perturbation due to three-beam interaction diminishes as the coherent contribution to the total scattered power is reduced. In this case, the invariant phase information may be difficult to extract. However, one could still, in favourable cases, be able to separate between $\phi_\Sigma = 0°$ and $\phi_\Sigma = 180°$ in a centrosymmetric case. This is illustrated in Fig. 4. Finally, for heavily distorted crystals – like case (*d*) in Fig. 3 – no dynamical effects are visible at all. Similar trends are found for other crystal geometries and reflection strengths.

An interesting feature to be noted is the dependence of $\sin\phi_\Sigma$ in equation (46), which occurs to first order. For the perfect-crystal case, such dependence is not evident until the second order in the series expansion. For cases in which the invariant triplet phase sum is close to 90°, this term becomes increasingly important and may result in a profile reversal. This is seen in Fig. 5 for $\tau/l = 0.5$, case (*d*), where the dynamical perturbation is clearly visible. The same situation is shown in Fig. 6 – but now with $\phi_\Sigma = -90°$. For such a case, no reversal is observed. The dynamical perturbation in Fig. 6(*d*) is however somewhat stronger than the corresponding case in Fig. 5 and it should hence be possible to assign the correct triplet phase to the profiles. This shows again the importance of always measuring the Friedel-related triplets in experiments (Weckert & Hümmer, 1990).

6. Conclusions

We have shown that the concepts of the statistical dynamical theory can be implemented also for the three-beam case. The calculations confirm that dynamical perturbation effects are observable in 'mosaic' crystals. In addition, we have found that ψ curves involving a phase sum near 90° may experience a profile reversal depending on the degree of crystal imperfection.

The work extends the application of the statistical dynamical theory. Questions related to actual physical models, *cf.* Kato (1976*b*, 1982), for the defect structure of the crystal, leading to estimates for the statistical quantities E and τ, have not been addressed. Because of the lack of variety of such models, it is difficult to verify the consequences of the various approximations used in the theory. The assumptions $\tau \ll \Lambda/E$ and isotropy of E seem to be the most crucial ones when it comes to the question of obtaining reliable phase information from ψ profiles in non-perfect crystals. It is clear that a deeper understanding of the role played by the statistical parameters is necessary for further development of the theory. Some other points should be mentioned:

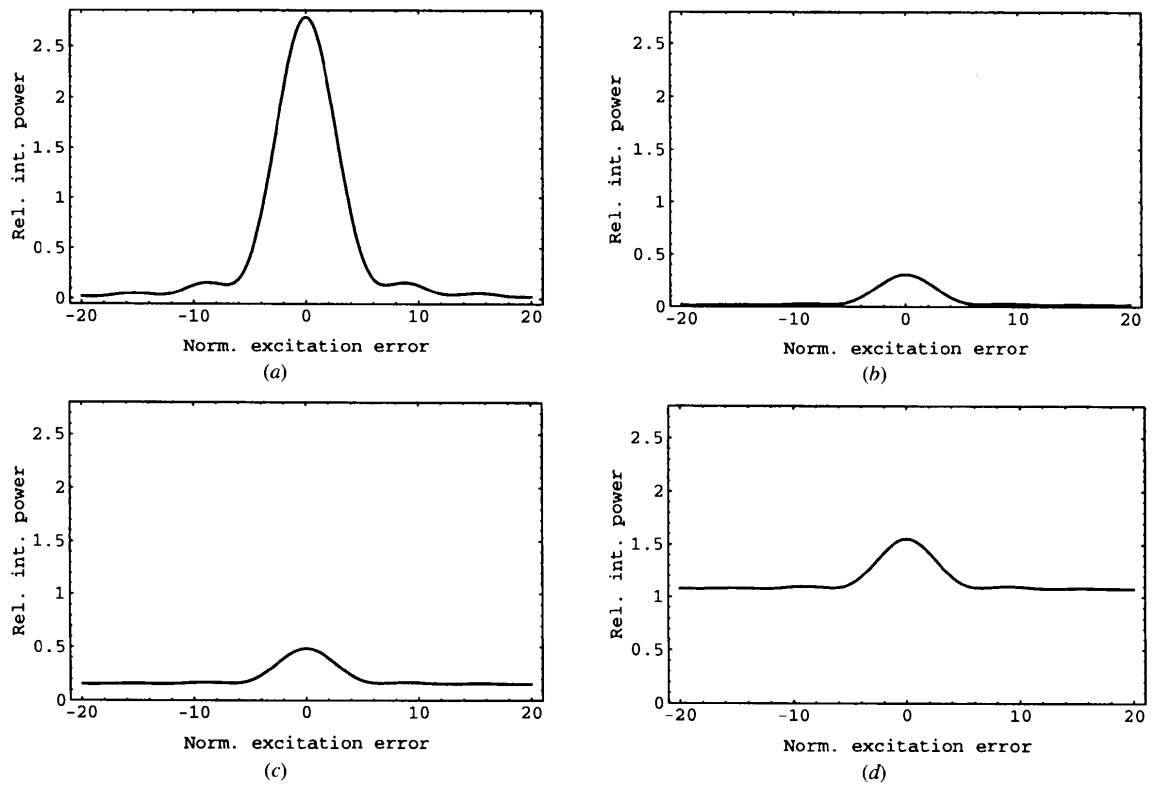

Fig. 6. Same situations as in Fig. 5, but now $\phi_\Sigma = -90°$.

(i) One has to extend the calculations to include the contributions from incoherent scattering events, thereby assessing the total wave fields generated inside the crystal. It is of importance to clarify whether the mixed coherent and incoherent contributions carry any phase information.

(ii) We have focused our attention on the integrated power. The shape of the ψ profiles is then closely linked to the functions $g_1^c(\xi_g)$ and $g_2^c(\xi_g)$, *cf.* equations (48) and (49). A damping and broadening of the profiles will occur with increasing value of the parameter m_g. However, the approximations used impose the restriction $m_g \ll 1$, and g_1^c and g_2^c will change little from the perfect-crystal case. However, a proper discussion related to the actual shape of the profiles might explicitly involve a model for the correlation function and not only its statistical behaviour.

(iii) For a finite crystal, its geometry will also influence the results. It is not clear at the moment how to generalize the calculations to a crystal of a more arbitrary shape or the influence of finiteness on the parameters of the statistical dynamical theory. Becker & Al Haddad (1990) claim that for the statistical assumptions to be valid $l \gg \Lambda$, where l is an average dimension of the crystal. On the other hand, our series-expansion technique is limited to cases where $l < \Lambda$. This is another question to be addressed in forthcoming works.

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