

- ARNOLD, E., ERICKSON, J. W., FOUT, G. S., FRANKENBERGER, E. A., HECHT, H. J., LUO, M., ROSSMANN, M. G. & RUECKERT, R. R. (1984). *J. Mol. Biol.* **177**, 417-430.
- ARNOLD, E. & ROSSMANN, M. G. (1986). *Proc. Natl Acad. Sci. USA*, **83**, 5489-5493.
- BHAT, T. N. & BLOW, D. M. (1982). *Acta Cryst.* **A38**, 21-29.
- BIESECKER, G., HARRIS, J. I., THIERRY, J. C., WALKER, J. E. & WONACOTT, A. J. (1977). *Nature (London)*, **266**, 328-333.
- BLOOMER, A. C., CHAMPNESS, J. N., BRICOGNE, G., STADEN, R. & KLUG, A. (1978). *Nature (London)*, **276**, 362-368.
- BRICOGNE, G. (1976). *Acta Cryst.* **A32**, 832-847.
- BUEHNER, M., FORD, G. C., MORAS, D., OLSEN, K. W. & ROSSMANN, M. G. (1974). *J. Mol. Biol.* **82**, 563-585.
- CALLAHAN, P. L., MIZUTANI, S. & COLONNO, R. J. (1985). *Proc. Natl Acad. Sci. USA*, **82**, 732-736.
- CROWTHER, R. A. (1969). *Acta Cryst.* **B25**, 2571-2580.
- ERICKSON, J. W., FRANKENBERGER, E. A., ROSSMANN, M. G., FOUT, G. S., MEDAPPA, K. C. & RUECKERT, R. R. (1983). *Proc. Natl Acad. Sci. USA*, **80**, 931-934.
- FINCH, J. T. & KLUG, A. (1959). *Nature (London)*, **183**, 1709-1714.
- FLETTERICK, R. J. & STEITZ, T. A. (1976). *Acta Cryst.* **A32**, 125-132.
- GAYKEMA, W. P. J., HOL, W. G. J., VEREIJKEN, J. M., SOETER, N. M., BAK, H. J. & BEINTEMA, J. J. (1984). *Nature (London)*, **309**, 23-29.
- HARRISON, S. C., OLSON, A. J., SCHUTT, C. E., WINKLER, F. K. & BRICOGNE, G. (1978). *Nature (London)*, **276**, 368-373.
- HENDRICKSON, W. A. (1981). *Refinement of Protein Structures*, edited by P. A. MACHIN, J. W. CAMPBELL & M. ELDER, pp. 1-8. Daresbury: Science and Engineering Research Council.
- HENDRICKSON, W. A. & LATTMAN, E. E. (1970). *Acta Cryst.* **B26**, 136-143.
- HOGLE, J. M., CHOW, M. & FILMAN, D. J. (1985). *Science*, **229**, 1358-1365.
- JOHNSON, J. E. (1978). *Acta Cryst.* **B34**, 576-577.
- JOHNSON, J. E., AKIMOTO, T., SUCK, D., RAYMENT, I. & ROSSMANN, M. G. (1976). *Virology*, **75**, 394-400.
- JONES, T. A. (1978). *J. Appl. Cryst.* **11**, 268-272.
- KAHN, R., FOURME, R., GADFT, A., JANIN, J. & ANDRÉ, D. (1982). *J. Appl. Cryst.* **15**, 330-337.
- KONNERT, J. H. & HENDRICKSON, W. A. (1980). *Acta Cryst.* **A36**, 344-350.
- LILJAS, L., UNGE, T., JONES, T. A., FRIDBERG, K., LÖVGREN, S., SKOGLUND, U. & STRANDBERG, B. (1982). *J. Mol. Biol.* **159**, 93-108.
- LUO, M., VRIEND, G., KAMER, G., MINOR, I., ARNOLD, E., ROSSMANN, M. G., BOEGE, U., SCRABA, D. G., DUKE, G. M. & PALMENBERG, A. C. (1987). *Science*, **235**, 182-191.
- MCARTHUR, G. R. (1983). Tech. Note NCAR-TH/166-IA, May. National Center for Atmospheric Research, Boulder, Colorado.
- MAIN, P. (1967). *Acta Cryst.* **23**, 50-54.
- MAIN, P. & ROSSMANN, M. G. (1966). *Acta Cryst.* **21**, 67-72.
- NORDMAN, C. E. (1980). *Acta Cryst.* **A36**, 747-754.
- RAYMENT, I. (1983). *Acta Cryst.* **A39**, 102-116.
- RAYMENT, I., BAKER, T. S., CASPAR, D. L. D. & MURAKAMI, W. T. (1982). *Nature (London)*, **295**, 110-115.
- ROSSMANN, M. G. (1976). *Acta Cryst.* **A32**, 774-777.
- ROSSMANN, M. G. (1979). *J. Appl. Cryst.* **12**, 225-238.
- ROSSMANN, M. G. (1984). *Biological Systems: Structure and Analysis*, edited by G. P. DIAKUN & C. D. GARNER, pp. 28-40. Daresbury: Science and Engineering Research Council.
- ROSSMANN, M. G., ARNOLD, E., ERICKSON, J. W., FRANKENBERGER, E. A., GRIFFITH, J. P., HECHT, H. J., JOHNSON, J. E., KAMER, G., LUO, M., MOSSER, A. G., RUECKERT, R. R., SHERRY, B. & VRIEND, G. (1985). *Nature (London)*, **317**, 145-153.
- ROSSMANN, M. G., ARNOLD, E. & VRIEND, G. (1986). *Acta Cryst.* **A42**, 325-334.
- ROSSMANN, M. G. & BLOW, D. M. (1962). *Acta Cryst.* **15**, 24-31.
- ROSSMANN, M. G. & BLOW, D. M. (1963). *Acta Cryst.* **16**, 39-45.
- ROSSMANN, M. G. & HENDERSON, R. (1982). *Acta Cryst.* **A38**, 13-20.
- ROSSMANN, M. G., LESLIE, A. G. W., ABDEL-MEGUID, S. S. & TSUKIHARA, T. (1979). *J. Appl. Cryst.* **12**, 570-581.
- SCHEVITZ, R. W., PODJARNY, A. D., ZWICK, M., HUGHES, J. J. & SIGLER, P. B. (1981). *Acta Cryst.* **A37**, 669-677.
- SCHILDKAMP, W. & BILDERBACK, D. (1986). *Nucl. Instrum. Methods*. In the press.
- SIM, G. A. (1959). *Acta Cryst.* **12**, 813-815.
- SIM, G. A. (1960). *Acta Cryst.* **13**, 511-512.
- STANWAY, G., HUGHES, P. J., MOUNTFORD, R. C., MINOR, P. D. & ALMOND, J. W. (1984). *Nucl. Acids Res.* **12**, 7859-7875.
- TEN EYCK, L. F. (1973). *Acta Cryst.* **A29**, 183-191.
- VARGHESE, J. N., LAVER, W. G. & COLMAN, P. M. (1983). *Nature (London)*, **303**, 35-40.
- VRIEND, G., ROSSMANN, M. G., ARNOLD, E., LUO, M., GRIFFITH, J. P. & MOFFAT, K. (1986). *J. Appl. Cryst.* **19**, 134-139.
- WANG, B. C. (1985). *Meth. Enzymol.* **115**, 90-112.

Acta Cryst. (1987). **A43**, 361-369

Three-Beam Diffraction in a Finite Perfect Crystal

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Abstract

By using the Laplace transform on the Takagi-Taupin equations for three coupled waves in a perfect crystal it has been possible to obtain general boundary-value Green functions for the wave fields D_o , D_h and D_g . For a crystal shaped as a parallelepiped the integrated power P_h is calculated in the kinematical limit by

suitable integrations over one divergence angle and over the entrance and exit surfaces. The result, which is expressed as a function of the deviation from the Bragg condition for the third wave, is continuous through the three-beam point, and gives the expected asymmetry associated with the invariant phase of the product of the three structure factors involved. The asymptotic behaviour is the same as that obtained

from pseudo-two-beam formulations based on standard plane-wave theories. In the expression for the integrated power the dimensions of the crystal, scaled to appropriate extinction lengths, occur as parameters. The movement of the reciprocal-lattice point g owing to the rotation of the crystal when P_h is to be measured is taken explicitly into account. When this movement is negligible or small, it is found that the diffracted power in the vicinity of the three-beam point shows oscillations due to a functional dependence corresponding to the Laue interference function. Both *Umweganregung* and *Aufhellung* situations are covered.

Introduction

Many-beam X-ray diffraction, and especially three-beam diffraction, is of importance in structural analysis for two main reasons: (i) the possibility offered for experimental determination of phases of reflections from single crystals; (ii) the need for correcting intensity data from effects which cause deviation from standard two-beam kinematical theory. The last point is of special importance when accurate structure factors are necessary, for instance in charge density studies.

Here we shall focus our attention on the invariant-phase information of structure factors which appears in the primary diffracted intensity near a three-beam point. Early works of Lipscomb (1949), Miyake & Kambe (1954), Kambe (1957) and Hart & Lang (1961) showed that the intensity of diffracted beams (both for X-rays and electrons) was affected by the relative phases of the structure factors involved. Later works concerning X-ray diffraction, especially by Colella (1974), Post (1977, 1979, 1983), Chapman, Yoder & Colella (1981), Chang (1981, 1982*a, b*, 1984, 1986), Chang & Valladares (1985) Juretschke (1982*a, b*, 1984), Høier & Aanestad (1981), Høier & Marthinsen (1983) and Hümmer & Billy (1982, 1986), have explored different topics related to multiple diffraction and the phase problem in detail. These works use the plane-wave dynamical theory of X-ray diffraction [*cf.* Batterman & Cole (1964) or Pinsker (1978)] as a starting point for the analysis, and the many-beam effects are usually associated with perturbations of the ordinary two-beam dispersion-surface branches.

In this work we have chosen Takagi-Taupin wave equations (Takagi 1962, 1969; Taupin 1964) for description of the dynamical scattering processes. By using these equations it is possible in a straightforward way to examine dynamical three-beam diffraction effects in finite crystals.

Here absorption and coupling of different polarization states have been neglected and the calculations have been performed with a crystal shaped as a parallelepiped with edges defined by the three wave vectors involved. These simplifications are mainly for

mathematical convenience as our main purpose has been to show the potential inherent in the Takagi-Taupin formalism.

Boundary-value Green functions for the wave fields

Field equations and boundary conditions

By combining Maxwell's equation for the electrical displacement field

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

with the Fourier expansion of the periodic electrical susceptibility

$$\chi_e(\mathbf{r}) = \sum_q \chi_q \exp(-2\pi i \mathbf{q} \cdot \mathbf{r}) \quad (2)$$

and the plane-wave expansion

$$\mathbf{D}(\mathbf{r}, t) = \sum_p \mathbf{D}_p(\mathbf{r}) \exp[2\pi i(\nu t - \mathbf{k}_p \cdot \mathbf{r})] \quad (3)$$

to first order in χ one obtains the Takagi-Taupin equations (Takagi, 1962, 1969; Taupin 1964; Authier, Malgrange & Tournarie 1968) for a perfect crystal:

$$(i/\pi)(\mathbf{k}_p \cdot \nabla) \mathbf{D}_p = [K^2(1 + \chi_0) - \mathbf{k}_p^2] \mathbf{D}_p + \sum_{q \neq p} \chi_{p-q} [\mathbf{k}_p^2 \mathbf{D}_q - (\mathbf{k}_p \cdot \mathbf{D}_q) \mathbf{k}_p]. \quad (4)$$

Here K is the wave number of the incoming vacuum wave, $K = 1/\lambda$, and \mathbf{k}_p is a crystal wave vector, *i.e.*

$$\mathbf{k}_p = \mathbf{k}_0 + \mathbf{p}, \quad (5)$$

\mathbf{p} being a reciprocal-lattice vector.

At this point the following assumptions are made:

(i) Effects associated with refraction and average absorption are neglected. Thus $\chi_0 \equiv 0$.

(ii) Any interactions between different polarization states (*i.e.* $\hat{\sigma}$ and $\hat{\pi}$) of the vector amplitudes $\{\mathbf{D}_q\}$ are neglected.

(iii) Friedel's law is fulfilled, *i.e.* $\chi_{p-q}^* = \chi_{q-p}$; thus anomalous absorption effects are not included.

These assumptions will greatly simplify the mathematical treatment, and the constraints imposed thereby will be considered in more detail in future work. With respect to (ii) numerical analysis by Aanestad (1979) has shown that when $\hat{\sigma}$ - $\hat{\pi}$ coupling terms are omitted an error of about 5% will arise in the calculated dispersion-surface branches. The same assumption is also made in pseudo-two-beam calculations, *cf.* Høier & Marthinsen (1983). Chang (1986) has shown that anomalous scattering has effects on the determination of invariant phases in near-absorption-edge multiple diffraction, an aspect which as a consequence of (iii) is not covered in the present work.

The Takagi-Taupin equations are then written

$$\begin{aligned} \partial D_p / \partial s_p &= 2\pi i \{ |\mathbf{k}_p| - K \} D_p \\ &\quad - \pi i K \sum_{q \neq p} \chi_{p-q} (\hat{\mathbf{e}}_p \cdot \hat{\mathbf{e}}_q) D_q \end{aligned} \quad (6)$$

where s_p is a coordinate along the base vector $\hat{\mathbf{s}}_p = \mathbf{k}_p / |\mathbf{k}_p|$, and D_p is the component of the wave field along the polarization vector $\hat{\mathbf{e}}_p$ ($\hat{\sigma}$ or $\hat{\pi}$).

By introducing the following quantities

$$\alpha_p = \{ |\mathbf{k}_p| - K \} \quad (7a)$$

$$\kappa_{pq} = -\pi K \chi_{p-q} (\hat{\mathbf{e}}_p \cdot \hat{\mathbf{e}}_q) = |\kappa_{pq}| \exp(i\varphi_{p-q}) \quad (7b)$$

we can write

$$\partial D_p / \partial s_p = 2\pi i \alpha_p D_p + i \sum_{q \neq p} \kappa_{pq} D_q. \quad (8)$$

α_p is equal to the excitation error, the distance from reciprocal point p to the Ewald sphere, and $1/|\kappa_{pq}|$ is equal to the extinction length. Explicitly,

$$|\kappa_{pq}| = (r_e \lambda / V_c) |F_{p-q}(\hat{\mathbf{e}}_p \cdot \hat{\mathbf{e}}_q)|. \quad (9)$$

Here r_e is the classical electron radius, V_c the unit-cell volume and F_q the structure factor of reflection q . When the first term on the right-hand side of (8) is removed by the phase transformation

$$D_p = \bar{D}_p \exp\left(2\pi i \sum_q \alpha_q s_q\right) \quad (10)$$

we obtain the set of equations

$$\partial \bar{D}_p / \partial s_p = i \sum_{q \neq p} \kappa_{pq} \bar{D}_q. \quad (11)$$

In the case of three-beam interaction, *i.e.* $p, q \in (o, h, g)$, the equations become:

$$\partial \bar{D}_o / \partial s_o = i\kappa_{oh} \bar{D}_h + i\kappa_{og} \bar{D}_g \quad (12a)$$

$$\partial \bar{D}_h / \partial s_h = i\kappa_{ho} \bar{D}_o + i\kappa_{hg} \bar{D}_g \quad (12b)$$

$$\partial \bar{D}_g / \partial s_g = i\kappa_{go} \bar{D}_o + i\kappa_{gh} \bar{D}_h. \quad (12c)$$

To obtain a solution to this set of equations, the boundary conditions must be specified. Restricting ourselves to *Laue* diffraction the so-called boundary-value Green functions are obtained by applying the conditions:

$$D_o(0, s_h, s_g) = \delta(s_h) \delta(s_g) \quad (13a)$$

$$D_h(s_o, 0, s_g) = 0 \quad (13b)$$

$$D_g(s_o, s_h, 0) = 0. \quad (13c)$$

The same boundary conditions will also apply to the fields $\{\bar{D}_p\}$.

Method of solution

With the given boundary conditions it is possible to obtain the solutions for the wave fields by applying the Laplace-transform technique. The transformation

of the fields is written

$$\begin{aligned} \tilde{D}_p(t_o, t_h, t_g) &= \int_0^\infty ds_o \int_0^\infty ds_h \int_0^\infty ds_g \bar{D}_p(s_o, s_h, s_g) \\ &\quad \times \exp[-(t_o s_o + t_h s_h + t_g s_g)] \\ &= L_3\{\bar{D}_p(s_o, s_h, s_g)\} \end{aligned} \quad (14)$$

with $p \in (o, h, g)$.

Performing a Laplace transformation of Takagi's equation (12) using boundary conditions (13), we arrive at the following set of algebraic equations:

$$\begin{bmatrix} t_o & -i\kappa_{oh} & -i\kappa_{og} \\ -i\kappa_{ho} & t_h & -i\kappa_{hg} \\ -i\kappa_{go} & -i\kappa_{gh} & t_g \end{bmatrix} \begin{bmatrix} \tilde{D}_o \\ \tilde{D}_h \\ \tilde{D}_g \end{bmatrix} = \begin{bmatrix} 1 \\ 0 \\ 0 \end{bmatrix}. \quad (15)$$

The expressions for the transformed fields are then obtained by Cramer's rule

$$\begin{bmatrix} \tilde{D}_o \\ \tilde{D}_h \\ \tilde{D}_g \end{bmatrix} = \frac{1}{\det \Lambda} \begin{bmatrix} t_h t_g + \kappa_{hg} \kappa_{gh} \\ i\kappa_{ho} t_g - \kappa_{hg} \kappa_{go} \\ i\kappa_{go} t_h - \kappa_{gh} \kappa_{ho} \end{bmatrix}. \quad (16)$$

Explicitly, the determinant of the system matrix Λ becomes

$$\det \Lambda = t_o t_h t_g + \gamma_{hg} t_o + \gamma_{og} t_h + \gamma_{oh} t_g + i\Gamma_{ohg} \quad (17)$$

where

$$\gamma_{oh} = \kappa_{oh} \kappa_{ho} = |\kappa_{oh}|^2 \quad (18a)$$

$$\gamma_{og} = \kappa_{og} \kappa_{go} = |\kappa_{og}|^2 \quad (18b)$$

$$\gamma_{hg} = \kappa_{hg} \kappa_{gh} = |\kappa_{hg}|^2 \quad (18c)$$

and

$$\begin{aligned} \Gamma_{ohg} &= \kappa_{ho} \kappa_{og} \kappa_{gh} + \kappa_{oh} \kappa_{go} \kappa_{hg} \\ &= 2|\kappa_{ho}| |\kappa_{go}| |\kappa_{hg}| \cos(\varphi_h + \varphi_g + \varphi_{g-h}) \\ &= 2|\kappa_{ho}| |\kappa_{go}| |\kappa_{hg}| \cos \varphi_\Sigma \end{aligned} \quad (18d)$$

with φ_Σ as the invariant sum of phases of the three structure factors involved. The fields $\bar{D}_p(s_o, s_h, s_g)$ are now found by the inverse Laplace transformation

$$\begin{aligned} \bar{D}_p(s_o, s_h, s_g) &= [1/(2\pi i)^3] \iiint_{\{L\}} dt_o dt_h dt_g \tilde{D}_p(t_o, t_h, t_g) \\ &\quad \times \exp(t_o s_o + t_h s_h + t_g s_g) \\ &= L_3^{-1}\{\tilde{D}_p(t_o, t_h, t_g)\}, \end{aligned} \quad (19)$$

where the paths of integrations $\{L\}$ are to the right of all poles in \tilde{D}_p . Notice that a similar approach was used by Kato (1980) in his work on the statistical dynamical theory of crystal diffraction.

Results

Without presenting the mathematical details* we give the final results for the wave fields:†

$$\begin{aligned} \bar{D}_o(s_o, s_h, s_g) &= \delta(s_h)\delta(s_g) - [(\gamma_{og}s_o s_g)^{1/2}/s_g] \\ &\times J_1[2(\gamma_{og}s_o s_g)^{1/2}]\delta(s_h) \\ &- [(\gamma_{oh}s_o s_h)^{1/2}/s_h] \\ &\times J_1[2(\gamma_{oh}s_o s_h)^{1/2}]\delta(s_g) \\ &+ \sum_{k=0}^{\infty} \sum_{l=0}^{\infty} (-i)^k (1/k!l!) (1/s_h s_g) \\ &\times \frac{J_{2l+k-1}\{2[(\gamma_{og}s_o + \gamma_{hg}s_h)s_g]^{1/2}\}}{[(\gamma_{og}s_o + \gamma_{hg}s_h)s_g]^{(2l+k-1)/2}} \\ &\times \frac{J_{l+k-1}[2(\gamma_{oh}s_o s_h)^{1/2}]}{(\gamma_{oh}s_o s_h)^{(l+k-1)/2}} \\ &\times (\Gamma_{ohg}s_o s_h s_g)^k (\gamma_{hg}s_h s_g)^l (\gamma_{og}s_o s_g)^l; \end{aligned} \quad (20a)$$

$$\begin{aligned} \bar{D}_h(s_o, s_h, s_g) &= i\kappa_{ho}J_0[2(\gamma_{oh}s_o s_h)^{1/2}]\delta(s_g) \\ &+ \sum_{k=0}^{\infty} \sum_{l=0}^{\infty} (-i)^k (1/k!l!) \\ &\times \left(\frac{i\kappa_{ho}}{s_g} \frac{J_{2l+k-1}\{2[(\gamma_{og}s_o + \gamma_{hg}s_h)s_g]^{1/2}\}}{[(\gamma_{og}s_o + \gamma_{hg}s_h)s_g]^{(2l+k-1)/2}} \right. \\ &\times \frac{J_{l+k}[2(\gamma_{oh}s_o s_h)^{1/2}]}{(\gamma_{oh}s_o s_h)^{(l+k)/2}} \\ &- \kappa_{hg}\kappa_{go} \frac{J_{2l+k}\{2[(\gamma_{og}s_o + \gamma_{hg}s_h)s_g]^{1/2}\}}{[(\gamma_{og}s_o + \gamma_{hg}s_h)s_g]^{(2l+k)/2}} \\ &\times \left. \frac{J_{l+k}[2(\gamma_{oh}s_o s_h)^{1/2}]}{(\gamma_{oh}s_o s_h)^{(l+k)/2}} \right) \\ &\times (\Gamma_{ohg}s_o s_h s_g)^k (\gamma_{hg}s_h s_g)^l (\gamma_{og}s_o s_g)^l; \end{aligned} \quad (20b)$$

$$\begin{aligned} \bar{D}_g(s_o, s_h, s_g) &= i\kappa_{go}J_0[2(\gamma_{og}s_o s_g)^{1/2}]\delta(s_h) \\ &+ \sum_{k=0}^{\infty} \sum_{l=0}^{\infty} (-i)^k (1/k!l!) \\ &\times \left(\frac{i\kappa_{go}}{s_h} \frac{J_{2l+k}\{2[(\gamma_{og}s_o + \gamma_{hg}s_h)s_g]^{1/2}\}}{[(\gamma_{og}s_o + \gamma_{hg}s_h)s_g]^{(2l+k)/2}} \right. \\ &\times \left. \frac{J_{l+k-1}[2(\gamma_{oh}s_o s_h)^{1/2}]}{(\gamma_{oh}s_o s_h)^{(l+k-1)/2}} \right) \end{aligned}$$

* Details of the intermediate mathematical calculations have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 43522 (8 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

† Since the expressions for the wave fields correspond to a source given by a Dirac function, a more appropriate name would be wave-field densities.

$$\begin{aligned} &- \frac{i\kappa_{go}}{s_h} (\gamma_{hg}s_h s_g) \\ &\times \frac{J_{2l+k+1}\{2[(\gamma_{og}s_o + \gamma_{hg}s_h)s_g]^{1/2}\}}{[(\gamma_{og}s_o + \gamma_{hg}s_h)s_g]^{(2l+k+1)/2}} \\ &\times \frac{J_{l+k}[2(\gamma_{oh}s_o s_h)^{1/2}]}{(\gamma_{oh}s_o s_h)^{(l+k)/2}} \\ &- \kappa_{gh}\kappa_{ho} \frac{J_{2l+k}\{2[(\gamma_{og}s_o + \gamma_{hg}s_h)s_g]^{1/2}\}}{[(\gamma_{og}s_o + \gamma_{hg}s_h)s_g]^{(2l+k)/2}} \\ &\times \frac{J_{l+k}[2(\gamma_{oh}s_o s_h)^{1/2}]}{(\gamma_{oh}s_o s_h)^{(l+k)/2}} \\ &\times (\Gamma_{ohg}s_o s_h s_g)^k (\gamma_{hg}s_h s_g)^l (\gamma_{og}s_o s_g)^l. \end{aligned} \quad (20c)$$

These expressions should then be multiplied by the unit step functions $\theta(s_o)\theta(s_h)\theta(s_g)$ as the wave fields are zero outside the pyramid defined by the unit vectors \hat{s}_o , \hat{s}_h and \hat{s}_g . In the three-beam case the diffraction equations (12) and the boundary conditions (13) are symmetric with respect to the indices h and g . Thus an alternative expression for \bar{D}_g can be obtained from (20b) through permutation of h and g . The equivalence of the expression for \bar{D}_g with the one given in (20c) can be proved by series expansion.

It is readily shown that the solutions for the wave fields satisfy the partial differential equations (12). Furthermore we obtain the limits

$$\lim_{u \rightarrow 0} \bar{D}_o(u, s_h, s_g) = \delta(s_h)\delta(s_g) \quad (21a)$$

while

$$\lim_{u \rightarrow 0} \bar{D}_h(s_o, u, s_g) = i\kappa_{ho}\bar{D}_o^{(2)}(s_o, s_g) + i\kappa_{hg}\bar{D}_g^{(2)}(s_o, s_g) \quad (21b)$$

and

$$\lim_{u \rightarrow 0} \bar{D}_g(s_o, s_h, u) = i\kappa_{go}\bar{D}_o^{(2)}(s_o, s_h) + i\kappa_{gh}\bar{D}_h^{(2)}(s_o, s_h) \quad (21c)$$

where the fields $\bar{D}_o^{(2)}(s_o, s_p)$ and $\bar{D}_p^{(2)}(s_o, s_p)$, $p \in \{h, g\}$, are the standard expressions for the two-beam wave fields (Kato, 1974; Bremer & Thorkildsen, 1986). The limiting expression for \bar{D}_h (\bar{D}_g) gives the field values after a unique diffraction into the \hat{s}_h (\hat{s}_g) direction. In an analysis of two-beam diffraction a corresponding result was found by Becker (1977).

To compare with results previously obtained, the general solutions should be evaluated in the limits (i) $\gamma_{og} \rightarrow 0$ and (ii) $\gamma_{hg} \rightarrow 0$. In both cases the only term in the double sum which survives is the one with $k = l = 0$. The expressions for the wave field \bar{D}_h in the two cases become:

$$(i) \quad \bar{D}_h(s_o, s_h, s_g) = i\kappa_{ho} \{ J_0 [2(\gamma_{oh}s_o s_h)^{1/2}] \delta(s_g) - [(\gamma_{hg}s_h s_g)^{1/2} / s_g] J_0 [2(\gamma_{oh}s_o s_h)^{1/2}] \times J_1 [2(\gamma_{hg}s_h s_g)^{1/2}] \}; \quad (22a)$$

$$(ii) \quad \bar{D}_h(s_o, s_h, s_g) = i\kappa_{ho} \{ J_0 [2(\gamma_{oh}s_o s_h)^{1/2}] \delta(s_g) - [(\gamma_{og}s_o s_g)^{1/2} / s_g] J_0 [2(\gamma_{oh}s_o s_h)^{1/2}] \times J_1 [2(\gamma_{og}s_o s_g)^{1/2}] \}. \quad (22b)$$

These results are in perfect agreement with those obtained using integral equations (Bremer & Thorkildsen, 1986).

Integrated power for a crystal with the shape of a parallelepiped

Power as a function of excitation errors

The crystal considered in this work is cut in the shape of a parallelepiped confined by the unit vectors \hat{s}_o , \hat{s}_h and \hat{s}_g . The actual dimensions are l_o , l_h and l_g along the corresponding directions (Fig. 1). The field amplitude D_h at a point P , ($s_o(P)$, l_h , $s_g(P)$), on the exit surface due to a point source at S , (0 , $s_h(S)$, $s_g(S)$), on the entrance surface is given by

$$D_h(P \leftarrow S) = D_o^{(e)} i\kappa_{ho} \left[J_0 [2(\gamma_{oh}\Delta_o \Delta_h)^{1/2}] \delta(\Delta_g) + \sum_{k=0}^{\infty} \sum_{l=0}^{\infty} (-i)^k (1/k!l!) \times \left(\frac{1}{\Delta_g} \frac{J_{2l+k-1} \{ 2[(\gamma_{og}\Delta_o + \gamma_{hg}\Delta_h)\Delta_g]^{1/2} \}}{[(\gamma_{og}\Delta_o + \gamma_{hg}\Delta_h)\Delta_g]^{(2l+k-1)/2}} \times \frac{J_{l+k} [2(\gamma_{oh}\Delta_o \Delta_h)^{1/2}]}{(\gamma_{oh}\Delta_o \Delta_h)^{(l+k)/2}} \right) \right]$$

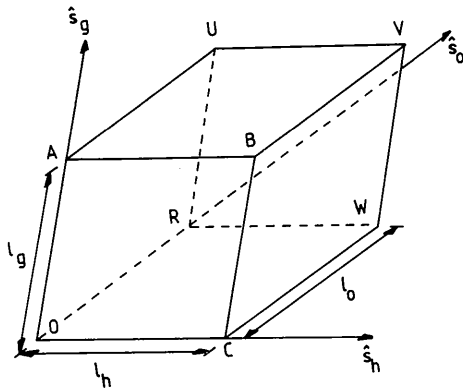


Fig. 1. Actual crystal cut in the shape of a parallelepiped. The unit vectors \hat{s}_o , \hat{s}_h and \hat{s}_g are parallel to \mathbf{OR} , \mathbf{OC} and \mathbf{OA} . The crystal dimensions are $|\mathbf{OR}| = l_o$, $|\mathbf{OC}| = l_h$ and $|\mathbf{OA}| = l_g$. Entrance surface is $OABC$, exit surface is $CBVW$.

$$+ \frac{i\kappa_{hg}\kappa_{go}}{\kappa_{ho}} \frac{J_{2l+k} \{ 2[(\gamma_{og}\Delta_o + \gamma_{hg}\Delta_h)\Delta_g]^{1/2} \}}{[(\gamma_{og}\Delta_o + \gamma_{hg}\Delta_h)\Delta_g]^{(2l+k)/2}} \times \frac{J_{l+k} [2(\gamma_{oh}\Delta_o \Delta_h)^{1/2}]}{(\gamma_{oh}\Delta_o \Delta_h)^{(l+k)/2}} \times (\Gamma_{ohg}\Delta_o \Delta_h \Delta_g)^k (\gamma_{hg}\Delta_h \Delta_g)^l (\gamma_{og}\Delta_o \Delta_g)^l \times \exp(2\pi i \alpha_h \Delta_h) \exp(2\pi i \alpha_g \Delta_g) \times \theta(\Delta_o) \theta(\Delta_h) \theta(\Delta_g). \quad (23)$$

Here $D_o^{(e)}$ is the amplitude of the incoming vacuum wave and

$$\Delta_o = s_o(P) - s_o(S) = s_o(P) \quad (24a)$$

$$\Delta_h = s_h(P) - s_h(S) = l_h - s_h(S) \quad (24b)$$

$$\Delta_g = s_g(P) - s_g(S), \quad (24c)$$

where the last equality applies to the actual crystal geometry.

The lowest-order expansion of (23) becomes

$$D_h(P \leftarrow S) = D_o^{(e)} i\kappa_{ho} [(1 - \gamma_{oh}\Delta_o \Delta_h) \delta(\Delta_g) - \gamma_{og}\Delta_o - \gamma_{hg}\Delta_h + i(|\kappa_{hg}\kappa_{go}|/|\kappa_{ho}|) \times \exp(-i\varphi_g)] \exp(2\pi i \alpha_h \Delta_h) \times \exp(2\pi i \alpha_g \Delta_g). \quad (25)$$

Now the field $D_h(P)$ due to an incoming plane wave is obtained by integrating (25) over the entrance [cf. Bremer & Thorkildsen (1986)]:

$$D_h(P) = J \int dS \cdot \hat{s}_o D_h(P \leftarrow S). \quad (26)$$

The Jacobian J is introduced to transform the source Dirac field density function from a representation in coordinates (t_1, t_2) normal to the incident direction to the coordinates s_h and s_g ; explicitly,

$$J = |\partial(s_o, s_h, s_g) / \partial(s_o, t_1, t_2)|. \quad (27)$$

However, this factor is cancelled when the surface integration is performed using a coordinate system spanned by \hat{s}_o , \hat{s}_h and \hat{s}_g . Expressed in the new coordinates Δ_h and Δ_g the field $D_h(P)$ becomes

$$D_h(P) = \int_0^{l_h} d\Delta_h \int_0^{s_g(P)} d\Delta_g D_h(P \leftarrow S). \quad (28)$$

Since we have the amplitude of the diffracted wave at P , we can calculate the beam intensity

$$I_h(P) = D_h(P) D_h^*(P). \quad (29)$$

The power P_h is then obtained by integrating I_h over the exit surface:

$$P_h = \int dP \cdot \hat{s}_h I_h(P) = J^{-1} \int_0^{l_o} d\Delta_o \int_0^{l_g} ds_g(P) I_h(P). \quad (30)$$

With the following set of dimensionless quantities:

$$|\eta_{oh}|^2 = \gamma_{oh} l_o l_h \quad (31a)$$

$$|\eta_{og}|^2 = \gamma_{og} l_o l_g \quad (31b)$$

$$|\eta_{hg}|^2 = \gamma_{hg} l_h l_g \quad (31c)$$

$$\xi_h = 2\pi\alpha_h l_h \quad (31d)$$

$$\xi_g = 2\pi\alpha_g l_g, \quad (31e)$$

the power is expressed as

$$\begin{aligned} P_h &= P_h(\xi_h, \xi_g) \\ &= I_o v |\kappa_{ho}|^2 l_h \{2f_1(\xi_h) - |\eta_{oh}|^2 f_1(\xi_h) \\ &\quad - 4(|\eta_{og}| |\eta_{hg}| / |\eta_{oh}|) [\cos \varphi_{\Sigma} f_1(\xi_h) f_2(\xi_g) \\ &\quad - \sin \varphi_{\Sigma} f_1(\xi_h) f_1(\xi_g)] - 2|\eta_{og}|^2 f_1(\xi_h) f_1(\xi_g) \\ &\quad - 2|\eta_{hg}|^2 [f_1(\xi_h) f_1(\xi_g) + f_4(\xi_h) f_2(\xi_g)] \\ &\quad + 4(|\eta_{og}|^2 |\eta_{hg}|^2 / |\eta_{oh}|^2) f_1(\xi_h) f_3(\xi_g)\}. \end{aligned} \quad (32)$$

Here $I_o = |D_o^{(e)}|^2$ is the intensity of the incident beam, and v is the volume of the crystal ($v = J^{-1} l_o l_h l_g$). The functions f_i are given by

$$f_1(u) = (1/u^2)(1 - \cos u) = \frac{1}{2} \sin^2(u/2)/(u/2)^2 \quad (33a)$$

$$f_2(u) = (1/u)[1 - (1/u) \sin u] \quad (33b)$$

$$f_3(u) = (1/u^2)[1 - (1/u) \sin u] \quad (33c)$$

$$f_4(u) = (1/u^2)[\sin u - (2/u)(1 - \cos u)]. \quad (33d)$$

In fact $f_1(\xi_h)$ and $f_1(\xi_g)$ are the Laue interference functions evaluated in the vicinity of the reciprocal-lattice points h and g respectively. One of the interesting aspects of (32) is that the power depends on the invariant phase sum θ_{Σ} . Notice especially the combination of $\cos \theta_{\Sigma}$ and the odd function $f_2(\xi_g)$ which varies with ξ_g^{-1} for large values of ξ_g , thus having long-range effect. This is the basis for using three-beam diffraction to obtain phase information and will be discussed in more detail later on. Another point that should not be overlooked is that (32) represents only a second-order approximation, being a series expansion up to the order of $|\eta|^2$. This limits its usefulness to cases where $|\eta| < 1$, or in other words to cases where the crystal dimensions do not exceed the extinction lengths involved. Thus (32) should be viewed as the kinematical limit of the diffracted power.

Integrated power

In what follows we shall assume the diffraction conditions usually employed with a four-circle diffractometer with \hat{s}_o and \hat{s}_h defining the diffraction plane. As pointed out by Høier & Marthinsen (1983), the excitation errors α_h and α_g will depend on two external divergence angles which give the position of the incident wave vector relative to the Laue point.

At the diffractometer three-beam diffraction is studied using the so-called ψ -scan technique (Busing & Levy, 1967), *i.e.* by rotating the crystal around the reciprocal-lattice vector \mathbf{h} . Thus it is convenient to use the angle ψ together with the angles ε_1 and ε_2 , which give the horizontal and vertical divergence of the incoming beam, as independent variables in the expressions for the excitation errors. From the definition (7a) it follows that

$$\alpha_p = \hat{s}_p \cdot \Delta \mathbf{p}, \quad p \in (h, g), \quad (34)$$

where $\Delta \mathbf{p}$ denotes the deviation from the exact Bragg position for the reciprocal-lattice point p . Using the angle definitions given in Fig. 2 [*cf.* Zachariasen (1945)], we find for infinitesimal rotations

$$\alpha_h = (\sin 2\theta_{oh}/\lambda) \varepsilon_1 \quad (35a)$$

$$\begin{aligned} \alpha_g &= \alpha_g(\psi) + (\cos \chi_g \sin \varphi_{og}/\lambda) \varepsilon_1 - (\sin \chi_g/\lambda) \varepsilon_2 \\ &\rightarrow \alpha_g(\psi) + (K_{og} \sin 2\theta_{og}/\lambda) \varepsilon_1. \end{aligned} \quad (35b)$$

The geometrical factor, K_{pq} in general, has been defined by Moon & Shull (1964):

$$K_{pq} = \cos \chi_p \cos \chi_q \sin \varphi_{pq} / \sin 2\theta_{pq}. \quad (35c)$$

In what follows we shall not explicitly take the vertical divergence into consideration in our calculations; ε_2 will be set to zero. Equations (35a) and (35b) show that normally both α_h and α_g will depend on the horizontal divergence. The integrated power is now obtained by integrating (32) with respect to ε_1 .

$$\begin{aligned} P_h(\xi_g) &= \int_{-\infty}^{\infty} d\varepsilon_1 P_h[\xi_h(\varepsilon_1), \xi_g(\varepsilon_1)] \\ &= I_o v |\kappa_{ho}|^2 (\lambda / \sin 2\theta_{oh}) \\ &\quad \times \{1 - \frac{1}{2} |\eta_{oh}|^2 - 2(|\eta_{og}| |\eta_{hg}| / |\eta_{oh}|) \\ &\quad \times [\cos \varphi_{\Sigma} f_2(\xi_g) - \sin \varphi_{\Sigma} f_1(\xi_g)] \\ &\quad - |\eta_{og}|^2 f_1(\xi_g) - |\eta_{hg}|^2 [f_1(\xi_g) + f_4(\xi_g)] \\ &\quad + 2(|\eta_{og}|^2 |\eta_{hg}|^2 / |\eta_{oh}|^2) f_3(\xi_g)\}, \end{aligned} \quad (36)$$

with

$$\xi_g = \xi_g(\psi) = 2\pi\alpha_g(\psi) l_g. \quad (37a)$$

Furthermore, with the quantity X as parameter,

$$X = |K_{og} \sin 2\theta_{og} / \sin 2\theta_{oh}| (l_g / l_h), \quad (37b)$$

the functions f_i become

$$\begin{aligned} f_1(u) &= (1/u^2)(1 - \cos u) \\ &\quad + (X/u^2)[1 + \cos u - (2/u) \sin u] \end{aligned} \quad (38a)$$

$$\begin{aligned} f_2(u) &= (1/u)[1 - (1/u) \sin u] \\ &\quad + (X/u^2)[\sin u - (2/u)(1 - \cos u)] \end{aligned} \quad (38b)$$

$$\begin{aligned} f_3(u) &= (1/u^2)[1 - (1/u) \sin u] \\ &\quad + (X/u^2)[\frac{1}{2} + (\sin u)/u - (3/u^2)(1 - \cos u)] \end{aligned} \quad (38c)$$

$$f_4(u) = (X/u^2)\{1 + \cos u - (2/u) \sin u - X[\cos u - 4(\sin u)/u + (6/u^2)(1 - \cos u)]\}. \quad (38d)$$

These expressions are valid for $X < 1$. The relevant expressions for $X > 1$ are given in the Appendix. Notice that for $X = 0$ the functions f_i are identical to the functions f_i , $i = 1, 2, 3$, which is obvious as $X = 0$ corresponds to the situation where the excitation error for the secondary beam (\mathbf{k}_g) does not depend on ε_1 .

The term in (36) proportional to $|\eta_{oh}|^2$ is the first-order correction of the integrated power due to primary extinction. This term will be neglected from now on. Introducing P_h^o for the kinematical two-beam integrated power

$$P_h^o = I_o v |\kappa_{ho}|^2 \lambda / \sin 2\theta_{oh}, \quad (39)$$

we have for the relative change in the integrated power due to three-beam interaction:

$$\begin{aligned} \Delta P_h(\xi_g) / P_h^o = & -2(|\eta_{og}||\eta_{hg}|/|\eta_{oh}|) \\ & \times [\cos \varphi_\Sigma f_2(\xi_g) - \sin \varphi_\Sigma f_1(\xi_g)] \\ & - |\eta_{og}|^2 f_1(\xi_g) - |\eta_{hg}|^2 [f_1(\xi_g) + f_4(\xi_g)] \\ & + 2(|\eta_{og}|^2 |\eta_{hg}|^2 / |\eta_{oh}|^2) f_3(\xi_g). \end{aligned} \quad (40)$$

This expression is the key result and shows how phase information can be obtained from measurements of integrated power for various values of ξ_g , i.e. by changing the excitation error for beam g .

Discussion

In Figs. 3-5 we have shown the results for the relative change in the integrated power in three different cases:

- (i) $|\eta_{oh}| = 0.05, |\eta_{og}| = 0.4, |\eta_{hg}| = 0.4$ (Fig. 3)
- (ii) $|\eta_{oh}| = 0.4, |\eta_{og}| = 0.05, |\eta_{hg}| = 0.4$ (Fig. 4)
- (iii) $|\eta_{oh}| = 0.05, |\eta_{og}| = 0.05, |\eta_{hg}| = 0.4$ (Fig. 5).

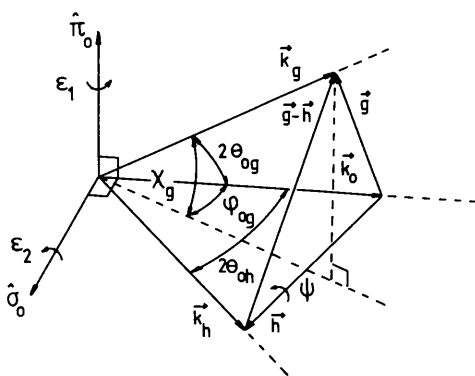


Fig. 2. Definitions of the angles involved when calculating how the excitation errors depend on the beam divergence and crystal rotation.

In all three cases $X = 0$, i.e. $K_{og} = 0$; thus the position of the reciprocal-lattice point g relative to the Ewald sphere is not affected by rotation of the crystal. The invariant phase sum φ_Σ has been given the values 0 (a), 180 (b), 90 (c) and 270° (d). Case (i) corresponds to what is usually called *Umweganregung*, case (ii) to *Aufhellung*, while case (iii) is an intermediate state. Notice the different scales along the ordinate axis in the three figures.

It is clear that these profiles carry phase information. For a centrosymmetric crystal we find that when the reciprocal-lattice point g is in a position inside the Ewald sphere corresponding to a negative value of ξ_g , the integrated power is enhanced when $\varphi_\Sigma = 0$ and reduced with respect to the two-beam result when $\varphi_\Sigma = 180^\circ$, and *vice versa* for a positive value of ξ_g . This result corresponds to the sign rule proposed by Chang (1982a).

The factor which determines the asymptotic behaviour of the diffraction profiles for a centrosymmetric crystal is the combination of $\cos \varphi_\Sigma$ and the odd function f_2 which varies asymptotically as ξ_g^{-1} . This is in accordance with results obtained from dynamical plane-wave theory (Juretschke 1982a, b; Højer & Marthinsen 1983).

In the case of a non-centrosymmetric crystal Juretschke (1982b), using a pseudo-two-beam treatment, has discussed some asymptotic aspects of the diffraction profile in the vicinity of the three-beam

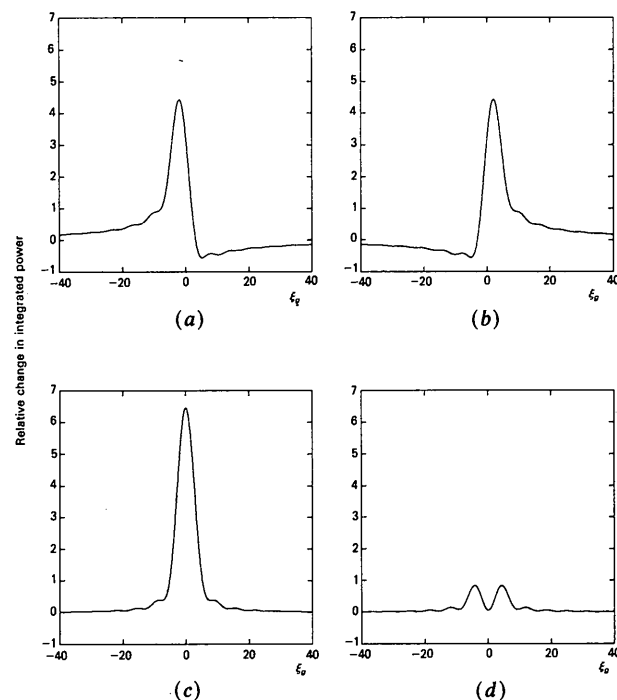


Fig. 3. Relative change in integrated power as function of ξ_g . $|\eta_{oh}| = 0.05, |\eta_{og}| = 0.4, |\eta_{hg}| = 0.4, X = 0$. (a) $\varphi_\Sigma = 0$; (b) $\varphi_\Sigma = 180^\circ$; (c) $\varphi_\Sigma = 90^\circ$; (d) $\varphi_\Sigma = 270^\circ$.

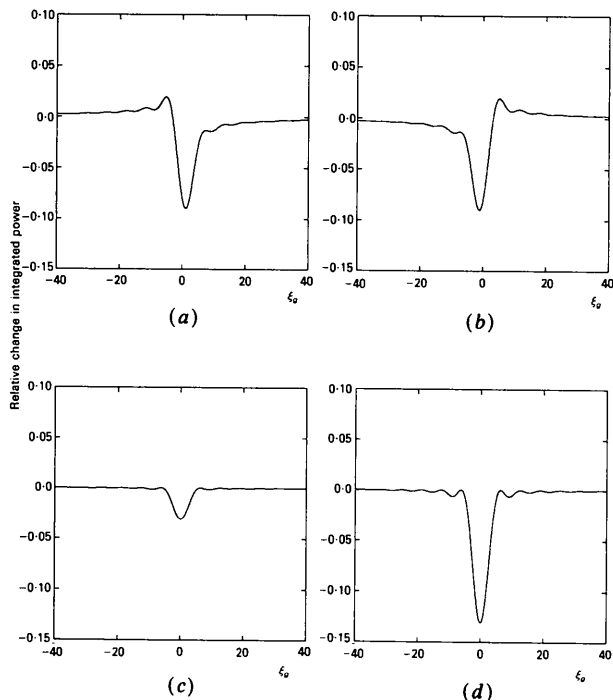


Fig. 4. Relative change in integrated power as functions of ξ_g . $|\eta_{oh}| = 0.4$, $|\eta_{og}| = 0.05$, $|\eta_{hg}| = 0.4$; $X = 0$. (a) $\varphi_\Sigma = 0^\circ$; (b) $\varphi_\Sigma = 180^\circ$; (c) $\varphi_\Sigma = 90^\circ$; (d) $\varphi_\Sigma = 270^\circ$.

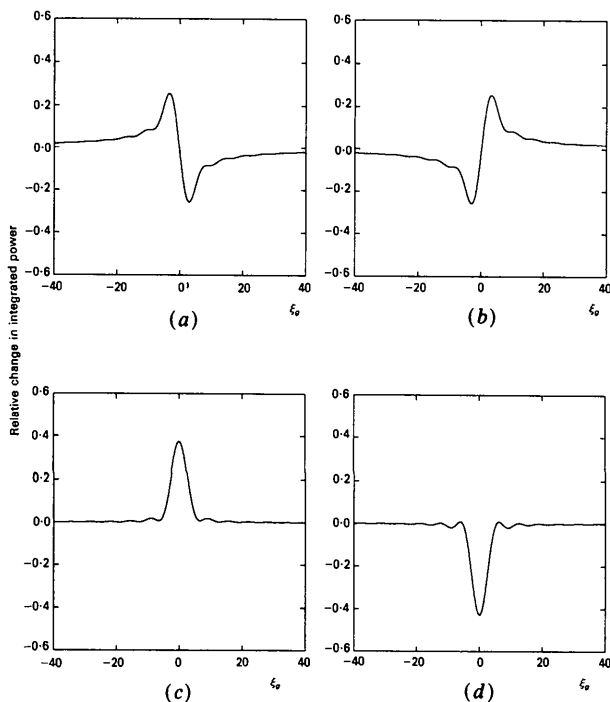


Fig. 5. Relative change in integrated power as function of ξ_g . $|\eta_{oh}| = 0.05$, $|\eta_{og}| = 0.05$, $|\eta_{hg}| = 0.4$; $X = 0$. (a) $\varphi_\Sigma = 0^\circ$; (b) $\varphi_\Sigma = 180^\circ$; (c) $\varphi_\Sigma = 90^\circ$; (d) $\varphi_\Sigma = 270^\circ$.

point. However, as is seen from Figs. 3–5 for $\varphi_\Sigma = 90^\circ$ and 270° , it is of importance to have information of the diffracted power at the exact three-beam point, where the pseudo-two-beam formulation diverges. Notice that phase information is also carried by the first-order term proportional to $(\sin \varphi_\Sigma) f_1$. Since f_1 varies with ξ_g^{-2} this term becomes important close to the three-beam point. When $\xi_g \rightarrow 0$ both first- and second-order contributions become important, their relative weight being determined by the $|\eta|$'s. The fact that the diffraction power is symmetric in ξ_g when $\varphi_\Sigma = 90^\circ$ and 270° is known from plane-wave dynamical theory, e.g. Ewald & Héno (1968).

For a finite crystal we have shown that the natural parameters in three-beam diffraction are ratios of crystal dimensions to extinction lengths, the $|\eta|$ parameters, and the natural variables are products of crystal dimensions and excitation errors, the ξ variables, all being dimensionless. Thus in addition to structure factors and crystal orientation, crystal dimension shows up as a very important quantity when the perturbation of the two-beam power is to be calculated. This aspect is not covered by standard dynamical plane-wave theory, and shows the usefulness of Takagi's (1962, 1969) formulation.

The discussion so far is based on the assumption that $X = 0$. Fig. 6 shows one example of how the

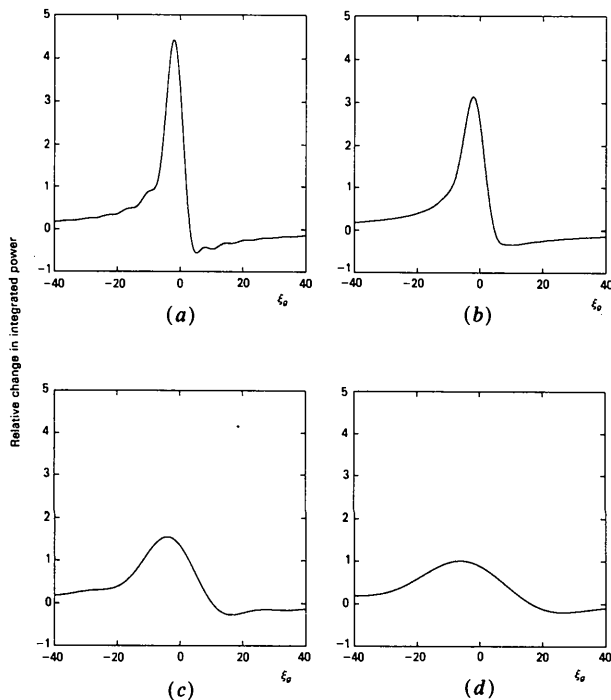


Fig. 6. Relative change in integrated power as function of ξ_g . Effect due to nonzero value of X , i.e. movement of reciprocal-lattice point g relative to the Ewald sphere when the crystal is rotated to measure integrated power. $|\eta_{oh}| = 0.05$, $|\eta_{og}| = 0.4$, $|\eta_{hg}| = 0.4$, $\varphi_\Sigma = 0^\circ$. (a) $X = 0$; (b) $X = 1$; (c) $X = 3$; (d) $X = 5$.

three-beam profiles are influenced by the movement of the reciprocal-lattice point g . The finer details of the diffraction profiles are now smeared out, and the three-beam effect is reduced. Thus in cases where the crystal dimensions are very anisotropic, which implies large values of X , it can be difficult to extract phase information from the profile measurements.

Concluding remarks

The work presented here shows the usefulness of the Takagi-Taupin formalism in treating three-beam diffraction in a perfect finite crystal. With the limitations imposed on the system the wave field inside the crystal is obtained by using the Laplace-transform technique. The integrated power is then calculated through a series expansion combined with suitable integrations. Thus the procedure followed corresponds to the one generally used in work on extinction, cf. Kato (1976) and Becker (1977). The calculations, which are straightforward, although rather lengthy, give analytical results which are not obtained with standard plane-wave theory.

The extension of the formalism to mosaic crystals will be discussed in a forthcoming paper (Thorkildsen & Mo, 1987). Furthermore, within the present formalism, one should think of extensions to cases where absorption effects become important.

The author is indebted to Professor R. Høier for hours of stimulating discussion on the dynamical theory of X-ray diffraction.

APPENDIX

Expressions for the functions $f_i(u)$ when $X > 1$

$$f_1(u) = (1/u^2)[1 + \cos(u/x)] \\ + (X/u^2)[1 - \cos(u/X)] \\ - (2/u) \sin(u/X) \quad (A1)$$

$$f_2(u) = (1/u)[1 + (1/u) \sin(u/X)] + (X/u^2) \\ \times \{-\sin(u/X) - (2/u)[1 - \cos(u/X)]\} \quad (A2)$$

$$f_3(u) = (1/u^2)[1 + \cos(u/X) - (1/2X) \cos(u/X)] \\ + (2/u) \sin(u/X) \\ + (X/u^2)\{ \frac{1}{2}[1 - \cos(u/X)] \\ - (2/u) \sin(u/X) \\ - (3/u^2)[1 - \cos(u/X)] \} \quad (A3)$$

$$f_4(u) = (X/u^2)(1 + \cos(u/X) - (1/X) \cos(u/X)) \\ + (4/u) \sin(u/X) \\ + X\{-(2/u) \sin(u/X) - (6/u^2) \\ \times [1 - \cos(u/X)]\}. \quad (A4)$$

All integrations leading to the expressions for f_i have been performed using a complex formulation with application of the residue theorem.

References

- AANESTAD, A. (1979). PhD thesis. Univ. of Trondheim-NTH, Trondheim.
- AUTHIER, A., MALGRANGE, C. & TOURNARIE, M. (1968). *Acta Cryst.* **A24**, 126–136.
- BATTERMAN, B. W. & COLE, H. (1964). *Rev. Mod. Phys.* **36**, 681–717.
- BECKER, P. (1977). *Acta Cryst.* **A33**, 667–671.
- BREMER, J. & THORKILDSEN, G. (1986). *Acta Cryst.* **A42**, 191–197.
- BUSING, W. R. & LEVY, H. A. (1967). *Acta Cryst.* **22**, 457–464.
- CHANG, S.-L. (1981). *Appl. Phys.* **A26**, 221–226.
- CHANG, S.-L. (1982a). *Phys. Rev. Lett.* **48**, 163–166.
- CHANG, S.-L. (1982b). *Acta Cryst.* **A38**, 516–521.
- CHANG, S.-L. (1984). *Multiple Diffraction of X-rays in Crystals*. Berlin: Springer.
- CHANG, S.-L. (1986). *Phys. Rev. B*, **33**, 5848–5850.
- CHANG, S.-L. & VALLADARES, J. A. P. (1985). *Appl. Phys.* **A37**, 57–64.
- CHAPMAN, L. D., YODER, D. R. & COLELLA, R. (1981). *Phys. Rev. Lett.* **46**, 1578–1581.
- COLELLA, R. (1974). *Acta Cryst.* **A30**, 413–423.
- EWALD, P. P. & HÉNO, Y. (1968). *Acta Cryst.* **A24**, 5–15.
- HART, M. & LANG, A. R. (1961). *Phys. Rev. Lett.* **7**, 120–121.
- HØIER, R. & AANESTAD, A. (1981). *Acta Cryst.* **A37**, 787–794.
- HØIER, R. & MARTHINSEN, K. (1983). *Acta Cryst.* **A39**, 854–860.
- HÜMMER, K. & BILLY, H. W. (1982). *Acta Cryst.* **A38**, 841–848.
- HÜMMER, K. & BILLY, H. W. (1986). *Acta Cryst.* **A42**, 127–133.
- JURETSCHKE, H. J. (1982a). *Phys. Rev. Lett.* **48**, 1487–1489.
- JURETSCHKE, H. J. (1982b). *Phys. Lett.* **92A**, 183–185.
- JURETSCHKE, H. J. (1984). *Acta Cryst.* **A40**, 379–389.
- KAMBE, K. (1957). *J. Phys. Soc. Jpn*, **12**, 13–31.
- KATO, N. (1974). In *X-ray Diffraction*, edited by L. V. AZAROFF, pp. 176–438. New York: McGraw-Hill.
- KATO, N. (1976). *Acta Cryst.* **A32**, 453–457.
- KATO, N. (1980). *Acta Cryst.* **A36**, 770–778.
- LIPSCOMB, W. N. (1949). *Acta Cryst.* **2**, 193–194.
- MIYAKE, S. & KAMBE, K. (1954). *Acta Cryst.* **7**, 218–220.
- MOON, R. M. & SHULL, C. G. (1964). *Acta Cryst.* **17**, 805–812.
- PINSKER, Z. G. (1978). *Dynamical Scattering of X-rays in Crystals*. Berlin: Springer.
- POST, B. (1977). *Phys. Rev. Lett.* **39**, 760–763.
- POST, B. (1979). *Acta Cryst.* **A35**, 17–21.
- POST, B. (1983). *Acta Cryst.* **A39**, 711–718.
- TAKAGI, S. (1962). *Acta Cryst.* **15**, 1311–1313.
- TAKAGI, S. (1969). *J. Phys. Soc. Jpn*, **26**, 1239–1253.
- TAUPIN, D. (1964). *Bull. Soc. Fr. Minéral. Cristallogr.* **87**, 469–511.
- THORKILDSEN, G. & MO, F. (1987). In preparation.
- ZACHARIASEN, W. H. (1945). *Theory of X-ray Diffraction in Crystals*. New York: John Wiley.