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References

BEARDEN, J. A. (1967). *Rev. Mod. Phys.* **39**, 78-124.
CROMER, D. T. (1983). *J. Appl. Cryst.* **16**, 437.
CROMER, D. T. & LIBERMAN, D. (1970). *J. Chem. Phys.* **53**, 1891-1898.
DESLATTES, R. D. & DEBEN, H. S. (1959). *Phys. Rev.* **115**, 71-74.
HENDRICKSON, W. A. (1985). *Trans. Am. Crystallogr. Assoc.* **21**, 11-21.
HENDRICKSON, W. A., TROUP, J. M. & ZDANSKY, G. (1986). Private communication.
International Tables for X-ray Crystallography (1974). Vol. IV. Birmingham: Kynoch Press. (Present distributor Kluwer Academic Publishers, Dordrecht.)
JAMES, R. W. (1982). *The Optical Principles of the Diffraction of X-rays*, p. 138. Woodbridge: Ox Bow.
JOHNSON, C. K. (1965). *ORTEP*. Report ORNL-3794. Oak Ridge National Laboratory, Tennessee, USA.
KRAUSE, M. O. & OLIVER, J. H. (1979). *J. Phys. Chem. Ref. Data*, **8**, 329-338.
PÄHLER, A. & HENDRICKSON, W. A. (1987). Private communication.
PHILLIPS, J. C., CERINO, J. A. & HODGSON, K. O. (1979). *J. Appl. Cryst.* **12**, 592-600.
ROGERS, D. (1981). *Acta Cryst.* A**37**, 734-741.
STERN, E. A. (1974). *Phys. Rev. B*, **10**, 3027-3037.
TEMPLETON, D. H. & TEMPLETON, L. K. (1982). *Acta Cryst.* A**38**, 62-67.
TEMPLETON, D. H. & TEMPLETON, L. K. (1985*a*). *Acta Cryst.* A**41**, 133-142.
TEMPLETON, D. H. & TEMPLETON, L. K. (1985*b*). *Acta Cryst.* A**41**, 365-371.
THOMPSON, S. D., CARROLL, D. G., WATSON, F., O'DONNELL, M. & MCGLYNN, S. P. (1966). *J. Chem. Phys.* **45**, 1367-1379.
WAGENFELD, H. (1975). *Anomalous Scattering*, edited by S. RAMASESHAN & S. C. ABRAHAMS, pp. 13-24. Copenhagen: Munksgaard.
ZACHARIASEN, W. H. (1967). *Theory of X-ray Diffraction in Crystals*, p. 114. New York: Dover.
ZDANSKY, G. (1968). *Ark. Kemi*, **29**, 443-448.

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Structure Analysis of Modulated Molecular Crystals. V. Symmetry Restrictions for One-Dimensionally Modulated Crystals

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Abstract

Symmetry restrictions that apply to amplitudes of displacement waves when atoms or rigid molecules are located at special positions in a modulated structure are tabulated for various crystallographic symmetry elements. The tables can be used for both the atomic and molecular models and are valid for harmonics of arbitrary order.

Introduction

The (3 + 1) superspace-group formalism described by de Wolff (1974) and de Wolff, Janssen & Janner (1981) has found widespread application in the description of one-dimensionally modulated crystals. For a given space group the amplitudes of the displacement waves are restricted when an atom or rigid molecule is located at a special position of the basic space group (*e.g.* Yamamoto, 1980, 1983; Kucharczyk, Paciorek & Uszynski, 1986; Gao, Gajhede, Mallinson, Petříček & Coppens, 1988). We present here a simple method for obtaining the restrictions and the derivation of general rules. The results are summarized in Table 1.

In order to illustrate our derivation, we will first summarize a number of relevant expressions given by de Wolff, Janssen & Janner (1981). A four-dimensional symmetry operation $\hat{S}$ is defined by

$$\hat{S}(x_1, x_2, x_3, x_4) = (x_1', x_2', x_3', x_4')$$

where the x_i are the coordinates with respect to the four-dimensional bases. $\hat{S}$ is described by the matrix

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Table 1. *Symmetry restriction of translational and rotational displacements as a function of symmetry operators*

Symbols of symmetry operators are chosen as in *International Tables for Crystallography* (1983), Tables 11.2 and 11.3. A minus sign indicates the component is not restricted. Non-zero restricted values are indicated by the symbols A, B, C and D, which are defined as local values for a given $\bar{\tau}$.

Symbol of symmetry operation	$\bar{\tau}$	Translational (U_a^x, U_b^x, U_c^x) (U_a^y, U_b^y, U_c^y)	Rotational (V_a^x, V_b^x, V_c^x) (V_a^y, V_b^y, V_c^y)
		$\varepsilon = 1$	
2 0, 0, z	0	(0, 0, −) (0, 0, −)	(0, 0, −) (0, 0, −)
	1/2	(−, −, 0) (−, −, 0)	(−, −, 0) (−, −, 0)
m $x, y, 0$	0	(−, −, 0) (−, −, 0)	(0, 0, −) (0, 0, −)
	1/2	(0, 0, −) (0, 0, −)	(−, −, 0) (−, −, 0)
Hexagonal tetragonal m x, x, z	0	(A, A, −) (B, B, −)	(C, $-C$, 0) (D, $-D$, 0)
	1/2	(A, $-A$, 0) (B, $-B$, 0)	(C, C, −) (D, D, −)
Hexagonal tetragonal m $x, -x, z$	0	(A, $-A$, −) (B, $-B$, −)	(C, C, 0) (D, D, 0)
	1/2	(A, A, 0) (B, B, 0)	(C, $-C$, −) (D, $-D$, −)
Hexagonal m $x, 2x, z$	0	(A, $2A$, −) (B, $2B$, −)	(−, 0, 0) (−, 0, 0)
	1/2	(−, 0, 0) (−, 0, 0)	(C, $2C$, −) (D, $2D$, −)
Hexagonal m $2x, x, z$	0	($2A$, A, −) ($2B$, B, −)	(0, −, 0) (0, −, 0)
	1/2	(0, −, 0) (0, −, 0)	($2C$, C, −) ($2D$, D, −)
Hexagonal m $x, 0, z$	0	(−, 0, −) (−, 0, −)	(C, 2C, 0) (D, $2D$, 0)
	1/2	(A, $2A$, 0) (B, $2B$, 0)	(−, 0, −) (−, 0, −)
Hexagonal m 0, y, z	0	(0, −, −) (0, −, −)	($2C$, C, 0) ($2D$, D, 0)
	1/2	($2A$, A, 0) ($2B$, B, 0)	(0, −, −) (0, −, −)
3 0, 0, z	0	(0, 0, −) (0, 0, −)	(0, 0, −) (0, 0, −)
	1/3	($A/2-\sqrt{3}B/2$, A, 0) ($\sqrt{3}A/2+B/2$, A, 0)	($C/2-\sqrt{3}D/2$, C, 0) ($\sqrt{3}C/2+D/2$, C, 0)
	2/3	($A/2+\sqrt{3}B/2$, A, 0) ($-\sqrt{3}A/2+B/2$, A, 0)	($C/2+\sqrt{3}D/2$, C, 0) ($-\sqrt{3}C/2+D/2$, C, 0)

Table 1. (*cont.*)

Symbol of symmetry operation	$\bar{\tau}$	Translational (U_a^x, U_b^x, U_c^x) (U_a^y, U_b^y, U_c^y)	Rotational (V_a^x, V_b^x, V_c^x) (V_a^y, V_b^y, V_c^y)
4 0, 0, z	0	(0, 0, −) (0, 0, −)	(0, 0, −) (0, 0, −)
	1/4	(A, B, 0) (B, $-A$, 0)	(C, D, 0) (D, $-C$, 0)
	1/2	(0, 0, 0) (0, 0, 0)	(0, 0, 0) (0, 0, 0)
	3/4	(A, B, 0) ($-B$, A, 0)	(C, D, 0) ($-D$, C, 0)
6 0, 0, z	0	(0, 0, −) (0, 0, −)	(0, 0, −) (0, 0, −)
	1/6	($A/2-\sqrt{3}B/2$, A, 0) ($\sqrt{3}A/2+B/2$, A, 0)	($C/2-\sqrt{3}D/2$, C, 0) ($\sqrt{3}C/2+D/2$, C, 0)
	1/3	(0, 0, 0) (0, 0, 0)	(0, 0, 0) (0, 0, 0)
	1/2	(0, 0, 0) (0, 0, 0)	(0, 0, 0) (0, 0, 0)
	2/3	(0, 0, 0) (0, 0, 0)	(0, 0, 0) (0, 0, 0)
	5/6	($A/2+\sqrt{3}B/2$, A, 0) ($-\sqrt{3}A/2+B/2$, A, 0)	($C/2+\sqrt{3}D/2$, C, 0) ($-\sqrt{3}C/2+D/2$, C, 0)
		$\varepsilon = -1$	
$\bar{1}$ 0, 0, 0	0	(−, −, −) (0, 0, 0)	(0, 0, 0) (−, −, −)
	1/2	(0, 0, 0) (−, −, −)	(−, −, −) (0, 0, 0)
2 0, 0, z	0	(−, −, 0) (0, 0, −)	(−, −, 0) (0, 0, −)
	1/2	(0, 0, −) (−, −, 0)	(0, 0, −) (−, −, 0)
Hexagonal tetragonal 2 $x, x, 0$	0	(A, $-A$, −) (B, B, 0)	(C, $-C$, −) (D, D, 0)
	1/2	(A, A, 0) (B, $-B$, −)	(C, C, 0) (D, $-D$, −)
Hexagonal tetragonal 2 $x, -x, 0$	0	(A, A, −) (B, $-B$, 0)	(C, C, −) (D, $-D$, 0)
	1/2	(A, $-A$, 0) (B, B, −)	(C, $-C$, 0) (D, D, −)
Hexagonal 2 $x, 0, 0$	0	(A, $2A$, −) (−, 0, 0)	(C, $2C$, −) (−, 0, 0)
	1/2	(−, 0, 0) (A, $2A$, −)	(−, 0, 0) (C, $2C$, −)
Hexagonal 2 0, y, 0	0	($2A$, A, −) (0, −, 0)	($2C$, C, −) (0, −, 0)
	1/2	(0, −, 0) ($2A$, A, −)	(0, −, 0) ($2C$, C, −)

Table 1. (*cont.*)

Symbol of symmetry operation	$\bar{\tau}$	Translational (U_a^x, U_b^x, U_c^x) (U_a^y, U_b^y, U_c^y)	Rotational (V_a^x, V_b^x, V_c^x) (V_a^y, V_b^y, V_c^y)
Hexagonal 2 x, 2x, 0	0	(−, 0, −) (A, 2A, 0)	(−, 0, −) (C, 2C, 0)
	1/2	(A, 2A, 0) (−, 0, −)	(C, 2C, 0) (−, 0, −)
Hexagonal 2 2x, x, 0	0	(0, −, −) (2A, A, 0)	(0, −, −) (2C, C, 0)
	1/2	(2A, A, 0) (0, −, −)	(2C, C, 0) (0, −, −)
m x, y, 0	0	(0, 0, −) (−, −, 0)	(−, −, 0) (0, 0, −)
	1/2	(−, −, 0) (0, 0, −)	(0, 0, −) (−, −, 0)
$\bar{4}$ 0, 0, z	0	(0, 0, −) (0, 0, 0)	(0, 0, 0) (0, 0, −)
	1/4	(0, 0, A) (0, 0, −A)	(0, 0, C) (0, 0, −C)
	1/2	(0, 0, 0) (0, 0, −)	(0, 0, −) (0, 0, 0)
	3/4	(0, 0, A) (0, 0, A)	(0, 0, C) (0, 0, C)

equation

$$\left(\begin{array}{c|c} \mathbf{R} & \mathbf{0} \\ \hline -\mathbf{m}^* & \varepsilon \end{array}\right)\begin{pmatrix} x_1 \\ x_2 \\ x_3 \\ x_4 \end{pmatrix}+\begin{pmatrix} s_1 \\ s_2 \\ s_3 \\ \delta \end{pmatrix}=\begin{pmatrix} x_1' \\ x_2' \\ x_3' \\ x_4' \end{pmatrix} \tag{1}$$

where **R** and $\mathbf{m}^*$ are 3×3 and 1×3 matrices respectively and $\varepsilon=\pm1$. R, m^* and ε describe the rotational part of $\hat{S}$, while the translational part is represented by $\mathbf{s}=(s_1, s_2, s_3)$ and δ.

The elements of the rotational part of the operator are restricted by the equation

$$\mathbf{m}^*=\varepsilon\mathbf{q}-\mathbf{qR} \tag{2}$$

where **q** is the modulation vector, which can be written as the sum of its irrational and rational components, $\mathbf{q}=\mathbf{q}_i+\mathbf{q}_r$. Substitution in (2) gives

$$\mathbf{m}^*=\varepsilon\mathbf{q}_r-\mathbf{q}_r\mathbf{R}$$

and (3)

$$\mathbf{0}=\varepsilon\mathbf{q}_i-\mathbf{q}_i\mathbf{R}.$$

The intrinsic rotational increment of the coordinate x_4, τ, is defined as

$$\tau=\delta-\mathbf{q}_r\mathbf{s}. \tag{4}$$

The displacement of the symmetry-related atom is related to that of the original atom through the equation

$$\mathbf{u}'(x_4')=\mathbf{Ru}[\varepsilon(x_4'-\delta+\mathbf{m}^*\mathbf{r})]. \tag{5}$$

The displacement function **u** is a periodic function, which in its general form is written as a sum of sine and cosine components (Petříček, Coppens & Becker, 1985):

$$\mathbf{u}(x_4)=\sum_{n=0}^{\infty}[\mathbf{U}^x(n)\sin2\pi nx_4+\mathbf{U}^y(n)\cos2\pi nx_4], \tag{6}$$

where n is the order of the harmonic.

General rules for restrictions

A special position of a space group is not changed by at least one of the symmetry operators (excluding the identity operation), or

$$\mathbf{r}=\mathbf{Rr}+\mathbf{s}. \tag{7}$$

A set of such operators forms a point group. From (7) it follows that an intrinsic part of **s**, which represents the projection of **s** into an invariant subspace of the matrix **R**, is equal to zero. Equation (5) can be rewritten for this special position by first using (3), which gives

$$\mathbf{u}'(x_4')=\mathbf{Ru}[\varepsilon(x_4'-\delta+\varepsilon\mathbf{q}_r\mathbf{r}-\mathbf{q}_r\mathbf{Rr})].$$

Substitution of (7) leads to

$$\mathbf{u}'(x_4')=\mathbf{Ru}[\varepsilon(x_4'-\tau+(\varepsilon-1)\mathbf{q}_r\mathbf{r})].$$

Since the displacement of the atom must be unique $\mathbf{u}'(x_4')=\mathbf{u}(x_4')$, or

$$\mathbf{u}(x_4')=\mathbf{Ru}\{\varepsilon[x_4'-\tau+(\varepsilon-1)\mathbf{q}_r\mathbf{r}]\}. \tag{8}$$

For a molecular displacement model the atomic displacements may be described in terms of translations and rotations of the rigid molecule. In the rectilinear approximation one obtains

$$\mathbf{u}(x_4)=\mathbf{U}(x_4)+\mathbf{V}(x_4)\times(\mathbf{r}-\boldsymbol{\rho}), \tag{9}$$

where $\boldsymbol{\rho}$ is the center of rotational displacement of the molecule, and **U** and **V** are vectors describing the translational and rotational molecular displacements, respectively.

For a rigid molecule occupying a special position there are two alternative ways of describing the displacement of a symmetry-related atom within the rigid molecule. One may directly apply the transformation law (8), or use the rigid-body condition (9) with the transformed atomic position. Thus, the following two equations must be satisfied simultaneously:

$$\begin{aligned}\mathbf{u}'(x_4')=&\mathbf{RU}\{\varepsilon[x_4'-\tau+(\varepsilon-1)\mathbf{q}_r\boldsymbol{\rho}]\}\\&+\mathbf{R}(\mathbf{V}\{\varepsilon[x_4'-\tau+(\varepsilon-1)\mathbf{q}_r\boldsymbol{\rho}]\}\times(\mathbf{r}-\boldsymbol{\rho}))\end{aligned} \tag{10a}$$

and

$$\mathbf{u}'(\mathbf{x}_4') = \mathbf{U}(x_4') + \mathbf{V}(x_4') \times (\mathbf{R}\mathbf{r} + \mathbf{s} - \boldsymbol{\rho}). \quad (10b)$$

It follows that

$$\mathbf{U}(x_4') = \mathbf{R}\mathbf{U}\{\varepsilon[x_4' - \tau + (\varepsilon - 1)\mathbf{q}_r\boldsymbol{\rho}]\} \quad (11a)$$

$$\mathbf{V}(x_4') = (\det \mathbf{R})\mathbf{R}\mathbf{V}\{\varepsilon[x_4' - \tau + (\varepsilon - 1)\mathbf{q}_r\boldsymbol{\rho}]\}. \quad (11b)$$

The condition (11a) for the translational displacement of the rigid molecule is the same as that for the individual atomic displacements of an atom at a special position, so that it can be used in both cases.

Equations (11) can be rewritten for individual harmonics by using the Fourier summation (6) and the substitutions

$$x = nx_4' \quad \text{and} \quad \bar{\tau} = n\tau - (\varepsilon - 1)\mathbf{q}_r\boldsymbol{\rho} \quad (12)$$

$$\begin{aligned}\mathbf{U}^x(n)\sin 2\pi x + \mathbf{U}^y(n)\cos 2\pi x \\ = \mathbf{R}\mathbf{U}^x(n)\sin[2\pi\varepsilon(x-\bar{\tau})] \\ + \mathbf{R}\mathbf{U}^y(n)\cos[2\pi\varepsilon(x-\bar{\tau})]\end{aligned} \quad (13a)$$

$$\begin{aligned}\mathbf{V}^x(n)\sin 2\pi x + \mathbf{V}^y(n)\cos 2\pi x \\ = (\det \mathbf{R})\mathbf{R}\mathbf{V}^x(n)\sin[2\pi\varepsilon(x-\bar{\tau})] \\ + (\det \mathbf{R})\mathbf{R}\mathbf{V}^y(n)\cos[2\pi\varepsilon(x-\bar{\tau})]\end{aligned} \quad (13b)$$

(note that det **R** is positive for a proper, and negative for an improper axis). Equations (13) make the discussion independent of the order of the harmonics. As (13a) and (13b) are similar, we will discuss mainly the former expression.

It is useful to refer the vectors in (13a) to the orthonormal coordinate system with the z axis along the rotational axis of (det **R**)**R**. The following conclusions can be drawn.

(1) The solution of the homogeneous linear system of equations (13a) for $\varepsilon = -1$ is independent of $\bar{\tau}$. The solution for $\bar{\tau} \neq 0$ can be derived from the solution for $\bar{\tau} = 0$ by using the transformation (as described in the Appendix*)

$$\begin{aligned}\mathbf{U}^x(n) &= \mathbf{U}_0^x(n)\cos\pi\bar{\tau} + \mathbf{U}_0^y(n)\sin\pi\bar{\tau} \\ \mathbf{U}^y(n) &= -\mathbf{U}_0^x(n)\sin\pi\bar{\tau} + \mathbf{U}_0^y(n)\cos\pi\bar{\tau}.\end{aligned} \quad (14)$$

This is in accordance with the fact that for supersymmetry operators with $\varepsilon = -1$, τ can be arbitrarily changed by selecting a different origin in four-dimensional space (see de Wolff, Janssen & Janner, 1981; Petříček, Coppens & Becker, 1985).

(2) Since $z' = (\det \mathbf{R})z$, the z components of $\mathbf{U}^x(n)$ and $\mathbf{U}^y(n)$ have to be zero for supersymmetry operators with $\varepsilon = 1$, unless $\bar{\tau} = 0$ and $\bar{\tau} = 1/2$ for $\det \mathbf{R} = 1$ and $\det \mathbf{R} = -1$, respectively.

(3) The x and y components of $\mathbf{U}^x(n)$ and $\mathbf{U}^y(n)$ have to be zero for supersymmetry operators with $\varepsilon = 1$ unless $\bar{\tau} = \pm\varphi/(2\pi)$ (φ being a rotational angle of **R**) or $\bar{\tau} = \pm\varphi/(2\pi) + \frac{1}{2}$ for $\det \mathbf{R} = 1$ and -1 respectively. The non-zero solutions have the form

$$\begin{aligned}U_y^x(n) &= \pm U_x^y(n) \\ U_x^x(n) &= \mp U_y^y(n)\end{aligned} \quad (15)$$

where the upper sign is for

$$\tau = \varphi/2\pi \qquad (\det \mathbf{R} = 1)$$

or

$$\tau = \varphi/2\pi + \tfrac{1}{2} \qquad (\det \mathbf{R} = -1)$$

and the lower sign is for

$$\tau = -\varphi/2\pi \qquad (\det \mathbf{R} = 1)$$

or

$$\tau = -\varphi/2\pi + \tfrac{1}{2} \qquad (\det \mathbf{R} = -1).$$

Note that (15) implies that the vectors $[U_x^x(n), U_y^x(n)]$ and $[U_x^y(n), U_y^y(n)]$ have the same length and are perpendicular.

Tables of restrictions

Equation (13) can be used to derive the restrictions for translational and rotational amplitudes. They are listed in Table 1 for both $\varepsilon = 1$ and $\varepsilon = -1$. In Table 1, $\bar{\tau}$ is as defined by (12). Its value modulo 1 is to be used to obtain the appropriate restrictions. For a supersymmetry operator with $\varepsilon = 1$, (12) becomes $\bar{\tau} = n\tau$. In this case all possible τ values can be found from the equation

$$m\tau + m(\mathbf{q}_r \cdot \mathbf{s}_0) = l \quad (l \text{ being integer})$$

(see Petříček, 1986), where m is the order of the rotation, and $\mathbf{s}_0$ is an intrinsic part of **s**. But this $\mathbf{s}_0$ is equal zero [see (7)], and thus all possible values of τ are $0, 1/m, 2/m, \ldots, (m-1)/m$. As τ can be arbitrarily changed by a shift of origin for supersymmetry operators with $\varepsilon = -1$, only commonly used values of $\bar{\tau}$ have been selected. Equation (14) can be used to calculate restrictions for other choices of $\bar{\tau}$.

Only the most important orientations of operators and modulation vectors are tabulated. Others can be derived by proper permutation.

* The Appendix has been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP51099 (3pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

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References

GAO, Y., GAJHEDE, M., MALLINSON, P., PETŘÍČEK, V. & COPPENS, P. (1988). *Phys. Rev. B*, **37**, 1825-1831.

International Tables for Crystallography. (1983). Vol. A. Dordrecht: Reidel. (Present distributor Kluwer Academic Publishers, Dordrecht.)

KUCHARCZYK, D., PACIOREK, W. A. & USZYNSKI, I. (1986). Abstr. Proc. Int. Conf. on Polytypes and Modulated Structures, Wrocław, Poland, p. 79.

PETŘÍČEK, V. (1986). Abstr. Proc. Int. Conf. on Polytypes and Modulated Structures, Wrocław, Poland, pp. 46-47.

PETŘÍČEK, V., COPPENS, P. & BECKER, P. J. (1985). *Acta Cryst.* A**41**, 478-483.

WOLFF, P. M. DE (1974). *Acta Cryst.* A**30**, 777-785.

WOLFF, P. M. DE, JANSSEN, T. & JANNER, A. (1981). *Acta Cryst.* A**37**, 625-636.

YAMAMOTO, A. (1980). *Phys. Rev. B*, **22**, 373-379.

YAMAMOTO, A. (1983). *Phys. Rev. B*, **27**, 7823-7825.

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Plane-Wave Theory of Three-Crystal Laue Interferometer

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Dedicated to Professor U. Bonse on the occasion of his 60th birthday

Abstract

The plane-wave theory of a three-crystal Laue interferometer is presented in terms of the amplitudes diffracted by a single slab in the Laue case, using Zachariasen's formalism. Successive applications of the single-slab expressions for the amplitudes lead to the final intensities of the interfering beams present on the back side of the third crystal slab. Numerical examples for X-ray and neutron diffraction show clearly the different contrast relationships in the two cases.

1. Introduction

The theory of a three-crystal symmetric Laue interferometer for spherical waves, in the case of zero absorption (neutrons or X-rays), has been developed by Bauspiess, Bonse & Graeff (1976). Such a theory contains the plane-wave situation as a particular case. An equivalent treatment, with full consideration of absorption, has also been presented by Petrascheck (1979).

Other treatments have appeared in the literature for the plane-wave situation (Bonse & Hart, 1965; Bonse & te Kaat, 1971) and zero or small absorption, for applications to neutron diffraction (Rauch & Suda, 1974; Staudenmann, Werner, Colella & Overhauser, 1980).

The spherical-wave treatment enables one to evaluate the intensity distribution as a function of position on the back of the third crystal slab of the interferometer. While such information is valuable in evaluating the overall performance of an interferometer in terms of the residual strain resulting from crystal defects and fastening techniques, quite often in designing an interferometer the attention is concentrated on the overall counting rate, namely on the integrated intensity emerging from the third slab, for which a plane-wave theory is adequate. Parseval's theorem, in fact, assures us that the integrated intensities are the same whether a plane- or a spherical-wave treatment is employed (Kato, 1968).

In this paper we present a very simple and straightforward derivation for the integrated intensities of some of the beams present behind the third slab of an interferometer made up with three slabs set for Laue diffraction (asymmetric), with full consideration of absorption, so that the formalism can be used in the X-ray case when absorption is important.

We will treat the case of a single slab first, and derive expressions that will then be combined in such a way as to obtain in a straightforward fashion the amplitudes of the waves multiply diffracted by a stack of crystal slabs, as a function of a global phase shift β, introduced along one of the interfering beams. The only critical assumption here is that all the various slabs are exactly coherent in space, which is true when monolithic interferometers are used.

Use will be made throughout this paper of the dynamical theory formalism developed by

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