place can be determined from the following set of equations:

$$|\beta(x_0)| = 9{\cdot}06\beta_c$$

$$a(x_0) = S(\eta_i)(\eta_i^2 - 1)^{1/2} + \frac{yb \sin 2\theta_B}{2\pi(\chi_h\chi_{\bar{h}})^{1/2}}\left(\frac{1}{x_0^2+y^2} - \frac{1}{x_i^2+y^2}\right) = 0.$$

In this way we may obtain the theoretical dependence of the y of maximal fringe intensity on η_i. Indeed, by solving the above set of equations we readily obtained the same type of dependence of the maximum fringe intensity that can be seen in the simulations. The agreement is, however, only qualitative. This is because the wavefields (I_4 and I_5) arriving at the crystal surface interfere. Certainly, it is also an important factor that the region where the trajectories change dramatically and wavefield creation takes place is quite small ($2|x_0|$ of the order of 20 μm) compared with the wave-front width in the real topographic conditions where a plane wave was employed.

5. Conclusion

In this paper a very important result concerning X-ray propagation has been obtained: geometrical optics results can be used if one takes into account the creation of new wavefields which occurs along every beam trajectory at the point where $a = 0$ with an intensity directly related to the value of the strain gradient at this point. This result has been demonstrated here in the Bragg case but it is general enough to be valid also in the Laue case.

Such an 'extended' geometrical-optics treatment when applied to any practical type of distortion, *e.g.* a dislocation line, should lead to a better understanding of the contrast origin in X-ray topographs.

As pointed out, geometrical optics cannot be applied in the total reflection range of the Bragg case. It seems, however, that there should be a similarity between the mechanism of the new wavefield creation in strongly deformed crystals and the phenomena occurring during total reflection. In this respect, it is obviously worthwhile to study this mechanism in more detail.

The authors wish to thank Dr Bedynska for her consent to reproduce here one of her simulations (Fig. 6). They are also grateful to Dr Y. Epelboin whose representation routine was a helpful tool for the visualization of the phenomenon of new-wavefield creation (Fig. 3). One of us (JG) acknowledges the financial support of CNRS for subsidizing a one-year stay at the Laboratoire de Minéralogie–Cristallographie in Paris.

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Acta Cryst. (1984). **A40**, 522–526

A Matrix Basis for CBED Pattern Analysis

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(*Received* 31 *July* 1983; *accepted* 4 *April* 1984)

Abstract

A simple construction procedure is given for convergent-beam electron diffraction (CBED) pattern matrices as symmetry elements in diffraction space coordinates. These are constructed from a limited set of point-group elements, namely those belonging to the layer groups of Alexander & Hermann [*Z. Kristallogr.* (1929), **70**, 328–345]. As a result a transformation is found between crystal and diffraction space in which the three-dimensional crystal symmetries transform into four-dimensional intensity distributions. Equivalent anti-symmetric matrices which operate on amplitudes rather than intensities are found for non-symmorphic space-group elements.

0108-7673/84/050522-05$01.50

I. Introduction

It is well known that the Seitz matrices (Seitz, 1935) can readily be transformed into reciprocal space, and that the transformed matrices offer a precise description of the kinematic X-ray diffraction pattern symmetries. The 3×3 rotational component, R, of these matrices is even more simply handled, becoming simply its transpose, R^T, in reciprocal space. Furthermore, a matrix-based nomenclature for the space-group symbols has recently been proposed by Hall (1981) with the aim of giving a more explicit notation.

The current problem in electron diffraction, of analyzing zone-axis pattern symmetries, is superficially at least more complicated. At first, it is not immediately obvious that an equally simple transformation between crystal space and pattern space will apply. CBED patterns result from intense N-beam dynamic interaction and are not, as are their counterpart in X-ray diffraction, subject to the restrictions of the Laue group. Their information content is correspondingly high. An analysis by Tanaka, Sekii & Nagasawa (1983), for example, has shown that 202 of the 230 three-dimensional space groups can be identified from characteristics associated with CBED pattern extinctions.

In spite of their inherent value, however, recent debate over interpretation (Ishizuka, 1982; Eades, Shannon & Buxton, 1983) serves to illustrate the point that current methods of analysis, which include scattering diagrams and analytical approximations (Ishizuka, 1983), are insufficiently precise to allow unequivocal interpretations of CBED pattern symmetries. On the other hand, matrices of appropriate order provide the logical means for writing down the symmetry operations within the CBED pattern, and ultimately for forming the appropriate diffraction group.

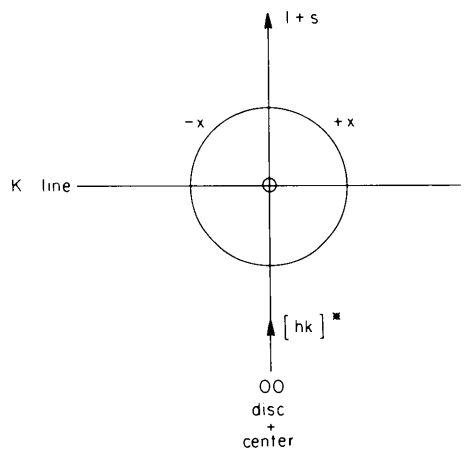

Fig. 1. The system of axes, relative to a specific hk disc, in which an origin is defined by the intersection of the K line or trace of $\zeta_h = 0$ for that reflexion, and the diffracting vector drawn from the pattern center (see text).

A revision of CBED nomenclature can be envisaged in three stages:

(1) provision of a matrix expression for CBED pattern symmetry elements;

(2) derivation of appropriate groups for both pattern space (diffraction groups) and crystal space (space groups);

(3) subsequent provision of an interrelation between the resulting groups and the space groups of three-dimensional crystallography.

If this were achieved there could be available something of the algebraic background currently available to kinematic X-ray diffraction.

In the present paper, which forms the first part of a series, we give a recipe for the construction of 4×4 CBED symmetry elements from a restricted set of point-group operators, namely those belonging to the space groups of two-dimensionally periodic layers (Alexander & Hermann, 1929), known as the 'layer groups'. These matrices refer to coordinates in CBED pattern space.

II. Coordinate system

We first define a system of axes applicable to the pattern, which, incidentally, is closely similar to the system adopted in the original Cowley & Moodie (1957) paper on dynamic diffraction, and proceed to define any *point* in the CBED pattern (a CBED pattern consists of a set of intensity *discs*) by the column vector

$$\begin{pmatrix} h \\ k \\ l+s \\ x \end{pmatrix}.$$

Here, (h, k) describes a specific reflexion disc, or order, and $(l+s, x)$ defines a point within that order with respect to the set of orthogonal axes determined by (i) the K line for that order (defined in the Fig. 1 caption) and (ii) the $[hk]^*$ diffracting vector drawn from the zone-axis center in the pattern. These axes intersect at an origin for that disc, *i.e.* the 'central point' defined earlier (Goodman, 1975). The ordinate s is then a specific excitation error ($s = -\zeta_h$ in the Cowley–Moodie notation), l is taken parallel to s (see Fig. 1), and the abscissa x is a collective N-beam excitation error ζ_N, for ζ_h = constant.†

† Here l has the restrictions or requirements similar to those implicit in the Cowley–Moodie notation (Cowley & Moodie, 1957, p. 615), which uses $U(hk)$ for the zero-layer ($l = 0$) wave amplitude. In point of detail, the Cowley–Moodie nomenclature defines the excitation errors, ζ_h, to represent a z-axis component of kinetic energy, where this axis is taken as coincident with the incident-beam direction, so that $\zeta_0 = 0$, whereas the present definitions would have the l index running parallel to the surface normal of the lamellar crystal. This latter definition has obvious advantages in symmetry analysis.

The formulation here is made convenient for principal zone axes, where the main symmetry occurs, and where there is most need for symmetry-interactive expressions.

The combination of indices as $(l+s)$ is correct for zone axes orthogonal to the principal $hk0$ net. This applies to systems of higher symmetry: hexagonal, tetragonal, cubic and orthorhombic. The combination index takes into account the overlapping of shape transforms which occurs between layers defined by different l values, $l=0, 1, 2, \ldots$. Thus, more than one value of l contributes to a given point on the shape transform since s is a continuous variable. This overlap effect, found within the summation over l of the N-beam scattering equations (Cowley & Moodie, 1957) provides one of the two categories of 'upper-layer interaction' in N-beam diffraction, the other category being that provided by dynamic scattering paths between non-overlapping transforms.

The only important system for which this combination index is incomplete at a principal setting is the monoclinic system in alternative (non-orthogonal) settings. In this case, since c is not parallel to c^*, the overlapping effect does not occur. Then the only complication is that there are two components of s required in the general diffraction vector, which becomes

$$\begin{pmatrix} h+s_\perp \\ k \\ l+s_\parallel \\ x \end{pmatrix}.$$

This complication is not referred to again as it adds nothing to the analysis, which remains valid, with the above understanding for monoclinic settings. (The triclinic system has not been discussed since it has only one possible non-translational symmetry.)

III. Rotational symmetry elements

Writing r^2 for a 2×2 symmetry element from the groups of plane rotations R^2 (referred to reciprocal-space coordinates) we have the following general form for a CBED pattern matrix:

$$\left(\begin{array}{cc|cc} r^2 & & 0 & 0 \\ & & 0 & 0 \\ \hline 0 & 0 & p & 0 \\ 0 & 0 & 0 & q \end{array}\right),$$

in which p and q can have the values $+1$ or -1. This matrix operates on the above column matrix in h, k, $l+s$, x to give h_1, k_1, $(l+s)_1$, x_1 *etc.*, the intensity- (or amplitude-) equivalent points in the CB pattern.

A useful subdivision into *direct* and *indirect* symmetry elements results in the following.

Direct symmetry operations, in which the $l+s$ index is unchanged, can, by choice, be represented by a 3×3 matrix, by omitting the third row and column above, *viz*

$$\begin{pmatrix} r^2 & 0 \\ 0 & \pm1 \end{pmatrix}, \quad \text{operating on} \begin{pmatrix} h \\ k \\ x \end{pmatrix}.$$

These operations are pure rotations about a vertical axis and the dichromatic index (the q index from above) is given by $\det|r^2|$. This is $+1$ for a proper rotation and -1 for an improper rotation (mirror reflexion).

Indirect symmetry elements (*i.e.* those invoking reciprocity) involve a change in the sign of $l+s$ and so require the 4×4 rotation matrix,

$$\left(\begin{array}{cc|cc} r^2 & & 0 & 0 \\ & & 0 & 0 \\ \hline 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & \pm1 \end{array}\right) = \left(\begin{array}{ccc|c} r^{2,1} & & & 0 \\ & & & 0 \\ & & & 0 \\ \hline 0 & 0 & 0 & \pm1 \end{array}\right) = r^{2,2}$$

operating on the full CB diffraction vector:

$$r^{2,2}\begin{pmatrix} h \\ k \\ l+s \\ x \end{pmatrix}.$$

Here, $r^{2,2}$ is an element of the doubly dichromatic rotation groups $R^{2,2}$, the sign of the dichromatic indices being determined by: p, whether or not reciprocity is involved (direct and indirect categories); and q, the three-dimensional nature of the rotation element (*i.e.* whether proper or improper).

The matrix elements of $r^{2,2}$ can be constructed from the elements $r^{2,1}$, which are the transposed rotation elements from the layer groups.

In this nomenclature the Buxton, Eades, Steeds & Rackham (1976) CBED pattern operator $\underline{R}$ is given by

$$\begin{pmatrix} E & 0 \\ 0 & -E \end{pmatrix} \quad \text{with } E = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}.$$

Symmetries of the central beam of CBED patterns are higher than those of the whole pattern since the two origins of the constituent matrices are coincident. Since both direct and indirect symmetries act independently, if we take one axis, $\mathbf{s}_0$, to be coincident with a principal crystallographic axis $\mathbf{h}$, the symmetry is given by

$$r^2\begin{pmatrix} p & 0 \\ 0 & q \end{pmatrix}\begin{pmatrix} s_0 \\ x_0 \end{pmatrix}$$

with $\mathbf{x}_0$ orthogonal to $\mathbf{s}_0$. It can easily be seen that the rotational group concerned has the character of

the Laue group since for a crystallographic center of symmetry both r^2 and $\begin{pmatrix} p & 0 \\ 0 & q \end{pmatrix}$ are equal to $-E$, so that the property of centrosymmetry cannot be determined.

Rotation elements of $R^{2,2}$ given above have a 'semi-arithmetic' definition, *i.e.* they include the elements m, m', 2, 2′ (Hall, 1981, nomenclature), which are specifically aligned to the axial system. The relevance of this definition with regard to CBED classification is discussed in another publication (Goodman, 1984*b*).

CBED matrices provide a basis for further group treatment and the transformation $R^{2,1} \rightleftharpoons R^{2,2}$ permits a simple derivation of diffraction groups for symmorphic space groups.

IV. Extinctive space-group elements

Compound symmetry elements which arise in non-symmorphic space groups can be defined, in a manner parallel to that demonstrated by Hall (1981) for X-ray kinematic diffraction, as anti-symmetric versions of the rotation matrices, with the matrices operating on diffraction amplitudes rather than intensities.* This permits a compact statement of the Gjønnes & Moodie (1965) rules for dynamic extinction. The anti-symmetries generated are shown schematically in Fig. 2.

Hence, the vertical glide plane, conventionally symbolized by a, is represented by

$$(-1)^h \left(\begin{array}{cc:cc} 1 & 0 & 0 & \\ 0 & -1 & & \\ \hdashline & & 1 & 0 \\ 0 & & 0 & -1 \end{array}\right), \quad \text{operating on} \quad \begin{pmatrix} h \\ k \\ l+s \\ x+\delta \end{pmatrix},$$

which is anti-symmetric in x for h odd (Fig. 2*b*). Summation over + and − terms results in the generation of Gjønnes–Moodie (G–M) '*A*' bands in the $k = 0$ reflexions, and for all values of $l+s$, when $x \simeq 0$, the width of the bands being a function of an arbitrarily small finite value chosen for δ. δ can be regarded as a half-width for dynamic interference.

Similarly, the horizontal twofold screw axis, 2_1, is represented by

$$(-1)^h \left(\begin{array}{cc:cc} 1 & 0 & & \\ 0 & -1 & 0 & \\ \hdashline & & -1 & 0 \\ 0 & & 0 & 1 \end{array}\right), \quad \text{operating on} \quad \begin{pmatrix} h \\ k \\ l+s+\delta \\ x \end{pmatrix},$$

which is anti-symmetric in $(l+s)$ for h odd (Fig. 2*c*). The horizontal glide plane, a, is similarly

$$(-1)^h \left(\begin{array}{cc:cc} 1 & 0 & & \\ 0 & 1 & & \\ \hdashline & & -1 & 0 \\ & & 0 & -1 \end{array}\right) \begin{pmatrix} h \\ k \\ (l+s)+\delta' \\ x+\delta'' \end{pmatrix},$$

where there is a need to distinguish between extinction widths δ' and δ''. This leads to a *diagonal* anti-symmetry (Fig. 2*d*), and generates the Gjønnes & Moodie condition ($A \cap B$). There is no specific requirement here for orthogonal axes (illustrated in Fig. 2*d*), and this extinction will occur in a two-dimensional set of $hk0$ reflexions, in rows with h odd. (Those with h even will have a centrosymmetric distribution.)

This gives the three basic types of compound symmetry elements, drawn from the same restricted set of groups, $R^{2,1}$. Allowing for permutations of the translation vector and rotation axes amongst the crystallographically allowed directions space groups can be formed in these coordinates. General three-

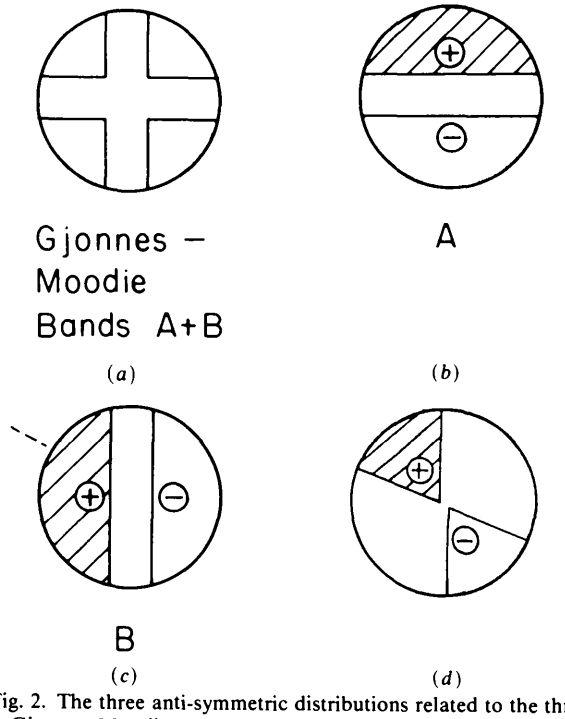

Fig. 2. The three anti-symmetric distributions related to the three Gjønnes–Moodie conditions *A*, *B* and $A \cap B$ (see text). The orientation of the discs is the same as that in Fig. 1.

* The anti-symmetric matrices of this section have a restricted application in being applied to amplitudes rather than intensities. Reciprocity (*e.g.* Moodie, 1972), involving an inversion of the coordinate $(l+s)$, implies only equivalent intensities, as generally applied. Conditions for exact symmetry or anti-symmetry of amplitudes and phases occur only for (*a*) direct symmetries for which $(l+s)$ is unchanged or (*b*) for indirect symmetries when $(l+s)$ is close to (strictly equal to) zero. These are the two conditions *A* and *B* of Gjønnes & Moodie (1965), more recently investigated quantum-mechanically by Portier & Gratias (1981).

The restricted application referred to is therefore relevant, since it refers to just those regions of pattern used in identifying non-symmorphic group symmetry elements.

dimensional extinction conditions outside the layer-group restrictions have not been included at this stage although extensive results along these lines have been produced by Tanaka, Sekii & Nagaswa (1983). Instead, we note that these extended conditions can be found from three-dimensional lattice groups formed from the same restricted set of point groups (Goodman, 1984*b*).

V. Preliminary requirements for application

The central plane approximation

Abstractly the transformation from $R^{2,2}$ to $R^{2,1}$, to the crystal-space coordinates of the layer groups, applies. We now examine the main assumption needed in order to apply this to real crystals. In the central plane approximation (CPA) it is assumed that all horizontal symmetry elements of the space group lie on the central horizontal plane of the crystal. For single symmetry elements this is an approximation which becomes exact periodically with crystal thickness, with the periodicity of the c spacing, and must always be a close approximation for crystals having many repeat distances in the z direction. This approximation is implied in all applications of space-group-determined matrices to pattern intensities. It has also been assumed in other group treatments of CBED symmetries (Tanaka, Sekii & Nagasaw, 1983; Buxton, Eades, Steeds & Rackham, 1976). Its validity was first tested specifically during the study of β-GaS belonging to the space group $P6_3/mmc$, in which horizontal diads occur at intervals of 30° around the [001] axis, separated vertically by $c/4$. In this case all horizontal symmetries were found to be active, as if they belonged to the central plane (Goodman & Whitfield, 1980).

Diffraction symmetries at a zone are higher than would otherwise be expected, owing to the increased possibilities for symmetry interaction under CPA conditions. As a result single symmetry elements can only be examined in isolation at settings sufficiently far from a zone, as illustrated in the above study in the tests for a center of symmetry.

Symmetry-group treatment overcomes the problem of such detailed analysis. Identification of a few pattern characteristics at chosen orientations, particularly if they include dynamic extinctions, has been shown to lead to unequivocal identification of space group (Tanaka, Sekii & Nagasawa, 1983; Goodman, 1984*a*).

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Acta Cryst. (1984). **A40**, 526–531

Triplet Phase Invariants from an Exact Algebraic Analysis of Anomalous Dispersion

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(*Received* 11 *January* 1984; *accepted* 11 *April* 1984)

Abstract

In a previous investigation, a system of exact algebraic equations was derived for any number and type of anomalous scatterers. Solution of the equations provides information concerning intensities of scattering and certain phase differences. In this paper, it is shown that when appropriate combinations of the phase differences and their values are made, the result is the evaluation of the differences of pairs of triplet phase invariants, one associated with the macromolecular structure and the second associated with the structure of the anomalous scatterers. It is usually easy to satisfy the condition that the values of triplet phase invariants associated with the structures of the anomalous scatterers be close to zero.

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