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Conditions for equality of intensities of electron beams diffracted by twinned crystals.

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A diffracted beam **h** will maintain a constant intensity on passing between the two parts of a twinned crystal if there exists at least one point operator R in the set of equivalent operators describing the boundary such that: $R\mathbf{h} = \mathbf{h}$ (1) and $R\mathbf{k}_0 = \mathbf{k}_0$ (2), where $\mathbf{k}_0$ is the wave vector of the incident beam. For the zero beam, relation (2) becomes: $R\mathbf{k}_0 = \pm\mathbf{k}_0$ (2′).

Conventional electron micrographs of twins generally exhibit a change in background intensity at the interface in both bright- and dark-field images. However, for particular directions of the incident beam, some diffracted beams may show no intensity change between adjacent twinned domains (Portier, Gratias & Fayard, 1977). A well-known example is given by bright-field images which never show an intensity change between inverse crystals. A theoretical explanation has been given by (Serneels, Snykers, Delavignette, Gevers & Amelinckx, 1973) using the column approximation and two-dimensional dynamical diffraction.

The role of upper-layer interactions in electron diffraction symmetries has been studied in detail for the convergent-beam method (Goodman, 1974; Steeds, Tatlock & Hampson, 1973) which shows the relation between the crystal symmetry and the symmetries of the beam patterns.

The present work is an attempt to give the geometrical conditions under which an electron beam diffracted by a crystal in a given direction maintains constant intensity when diffracted by an adjacent twin crystal. When this condition is fulfilled a conventional electron micrograph, exposed with this diffracted beam, reveals the same illuminating background intensity for the two adjacent twin crystals, except at the overlap region where interface fringes are generally found. We shall disregard here these fringes and use the three-dimensional dynamical diffraction formulation (Cowley & Moodie, 1957) within the column approximation.

We designate by $(R|\tau)$ one of the geometrical space operations – in Seitz (1936) notation – which relate the homologous points $\mathbf{r}^{\mathrm{I}}$ and $\mathbf{r}^{\mathrm{II}}$ of the crystal twins I and II:

$$(R|\tau)\,\mathbf{r}^{\mathrm{I}} = \mathbf{r}^{\mathrm{II}} = R\mathbf{r}^{\mathrm{I}} + \tau. \tag{1}$$

The crystal potentials are related by:

$$V^{\mathrm{I}}[(R|\tau)\,\mathbf{r}] = V^{\mathrm{II}}(\mathbf{r})$$

and their Fourier components by:

$$F^{\mathrm{II}}(R^{-1}\mathbf{h}) = F^{\mathrm{I}}(\mathbf{h})\exp(2i\pi\mathbf{h}.\tau). \tag{2}$$

Using the same method as Gjønnes & Moodie (1965) for the extinction conditions in electron diffraction, we consider any given multiscattering path in crystal I:

$$\mathbf{h}_1, \mathbf{h}_2, \ldots, \mathbf{h} - \sum_{i=1}^{n-1} \mathbf{h}_i$$

corresponding to the general term of the series involved in dynamical scattering (Cowley & Moodie, 1962):

$$F^{\mathrm{I}}(\mathbf{h}_1)F^{\mathrm{I}}(\mathbf{h}_2)\ldots F^{\mathrm{I}}\left(\mathbf{h} - \sum_{i=1}^{n-1}\mathbf{h}_i\right).Z(\zeta_1, \zeta_2, \ldots, \zeta)$$

for the expression of the scattered wave in the $\mathbf{h}$ direction. ζ_i are the excitation errors of the $\sum_{j=1}^{l}\mathbf{h}_j$ spots and Z is a totally symmetric function of ζ_i.

According to relation (2) an equivalent term is given in crystal II by the multiscattering path:

$$R^{-1}\mathbf{h}_1, R^{-1}\mathbf{h}_2, \ldots R^{-1}\mathbf{h} - \sum_{i=1}^{n-1} R^{-1}\mathbf{h}_i$$

for the $R^{-1}\mathbf{h}$ direction, with equal modulus and a constant $[\exp(2i\pi\mathbf{h}.\tau)]$ phase change, only if the Z functions are equal, *i.e.* if

$$R\mathbf{k}_0 = \mathbf{k}_0. \tag{3}$$

Relation (3) leads to the equality of intensity between the spots $\mathbf{h}$ and $R^{-1}\mathbf{h}$. There will then be no change in background intensity if $\mathbf{h}$ and $R^{-1}\mathbf{h}$ are superimposed:

$$R\mathbf{h} = \mathbf{h}. \tag{4}$$

These two conditions (3) and (4) give the geometrical rule for an $\mathbf{h}$ spot to exhibit no intensity change on the two sides of a twin boundary.

The central spot needs particular attention: every multiscattering path being closed, it can always be equivalently described in two ways:

$$\mathbf{h}_1, \mathbf{h}_2, \ldots, \mathbf{h}_{n-1}, 0 \quad \text{and} \quad -\mathbf{h}_{n-1}, -\mathbf{h}_{n-2}, \ldots, -\mathbf{h}_1, 0.$$

The second path is equivalent to the first path but with $-\mathbf{k}_0$ wave vector for the incident beam, so that relation (3) now has to include the minus sign:

$$R\mathbf{k}_0 = \pm\mathbf{k}_0. \tag{5}$$

The minus sign is obviously superfluous for centrosymmetric structures but has to be considered in the non-centrosymmetric case. In particular relation (5) is always satisfied for an inversion boundary ($R = \bar{1}$) and for any orientation of the incident beam: the Friedel law is always satisfied for bright-field images in three-dimensional diffraction.

Table 1 illustrates relations (3) and (5) for the cases where R is a crystallographic operation (merohedral twins or twins generated by a length-preserving phase transition). As an example of the use of the table let us consider the inversion twins obtained by ordering of $LiFe_5O_8$ (Portier, Gratias & Fayard, 1977). The point group of the ordered structure being {432}, an inversion boundary is characterized by the multiplication of the inversion $\bar{1}$ by the elements of {432}:

$$\{R\} = \bar{1} \times \{432\}.$$

We consider the [111] zone axis diffraction (Fig. 1). There is no centre of symmetry in the two-dimensional projection of the crystal and inverse domains generally show a difference

of background intensity. However, it may be possible to obtain equal intensity for dark-field micrographs performed with **h** diffraction spots satisfying relation (4) for at least one point operator of $\{R\}$. Such is the case for the $\langle 1\bar{1}0\rangle$ and $\langle 11\bar{2}\rangle$-type reflections because $R_1 = \bar{1} \times 2_{[001]} = m_{[001]}$ and $R_2 = \bar{1} \times 2_{[01\bar{1}]} = m_{[01\bar{1}]}$ leave respectively $\bar{1}10$ and $\bar{2}11$ invariant. From Table 1 it can be shown that $R = m_{[001]}$ may not satisfy relation (3) for the [111] zone axis because this mirror is neither parallel nor perpendicular to the zone axis ($\mathbf{k}_0$ is not invariant by this mirror). The $\langle 110\rangle$ spots exhibit a difference in background intensity between inverse domains. On the contrary, the $m_{[01\bar{1}]}$ mirror being parallel to the zone axis whatever the orientation of $\mathbf{k}_0$ such that $\mathbf{k}_0$ is contained in m, the $\mathbf{h} = \bar{2}11$ spot shows no background intensity change. In this case the projection of the Ewald sphere centre is located on the A geometrical locus of the table, which is the intersection of the R mirror with the diffracting zone. This result holds for zero- and upper-layer interactions. Moreover, the $\langle 110\rangle$-type reflections of the pattern deduced from each other by the R-mirror have exchanged intensities between the inverse domains, because relation (3) is satisfied. If we try to obtain equal intensities in inverse domains for all the $\langle 112\rangle$-type spots in the pattern, the $\mathbf{k}_0$ vector has to be simultaneously invariant with respect to the different R mirrors involved ($m_{[1\bar{1}0]}, m_{[10\bar{1}]}, m_{[0\bar{1}1]}$): the projection of the Ewald sphere must exactly correspond to the central spot O (intersection of $\mathbf{k}_0$ with the diffracting zone); in this case the actual trigonal symmetry of the pattern clearly appears because the $\bar{1}10$, $10\bar{1}$ and $0\bar{1}1$ spots exhibit the same intensity for a given crystal (and are exchanged with the $1\bar{1}0$, $\bar{1}01$, $01\bar{1}$ spots for the inverse crystal). The experimental observations of these symmetry properties have been shown previously (Portier *et al.*, 1977).

Table 1. *Conditions under which a diffracting spot* $\mathbf{h} = R\mathbf{h}$ *exhibits no intensity change between crystal twins*

O intersection of $\mathbf{k}_0$ with the diffracting plane.
A intersection of the *R* operation with the diffracting plane.

	Location of the projection of the centre of the Ewald sphere on the diffracting plane	
	Upper-layer interactions	Zero-layer interactions
R		
Even axis perpendicular to $\mathbf{k}_0$	Only central spot *O*	*A*
Axis parallel to $\mathbf{k}_0$	Only central spot *O*	
Mirror perpendicular to $\mathbf{k}_0$	Only central spot *O*	Always
Mirror parallel to $\mathbf{k}_0$	*A*	
Inversion centre	Only central spot Always	

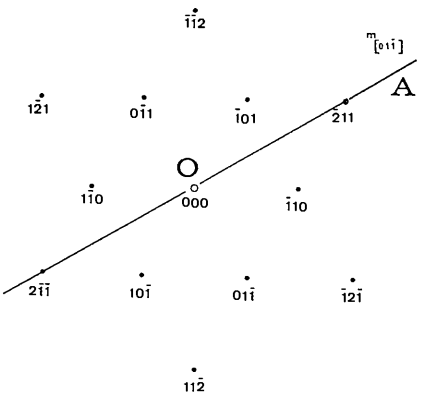

Fig. 1. Example of use of the table: geometrical locus *A* of the projection of the Ewald sphere for the $\bar{2}11$ reflection to show no backgound intensity change between two inverse domains of point group 432. If $\mathbf{k}_0$ is exactly projected on *O* all the 112-type reflections of the pattern show no background intensity change.

References

COWLEY, J. M. & MOODIE, A. F. (1957). *Acta Cryst.* **10**, 609–619.

COWLEY, J. M. & MOODIE, A. F. (1962). *J. Phys. Soc. Jpn*, **17**, B-II, 86.

GJØNNES, J. & MOODIE, A. F. (1965). *Acta Cryst.* **19**, 65–67.

GOODMAN, P. (1974). *Nature (London)*, **251**, 698–701.

PORTIER, R., GRATIAS, D. & FAYARD, M. (1977). *Philos. Mag.* **36**, 421–436.

SEITZ, F. (1936). *Ann. Math.* **37**, 17.

SERNEELS, R., SNYKERS, M., DELAVIGNETTE, P., GEVERS, R. & AMELINCKX, S. (1973). *Phys. Status Solidi B*, **58**, 277–292.

STEEDS, J. W., TATLOCK, G. J. & HAMPSON, J. (1973). *Nature (London)*, **241**, 435.