

## Studies in Domain Crystallography. I. The Composite Reciprocal Lattice of a Multidomain ('Hybrid') Crystal

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(Received 2 August 1961)

The diffraction geometry of a 'hybrid' single crystal composed of a finite number of domain orientations is interpreted in terms of a composite reciprocal lattice formed by the superposition of the reciprocal lattices of the individual domains. A procedure is described whereby the composite reciprocal lattice can be used to index oscillation photographs and the method is illustrated by an example of anomalous domain orientations in the structural transition of potassium cyanide.

### Introduction

When a solid undergoes a thermal transformation that involves a loss of crystallographic symmetry elements, there are two possibilities regarding the mutual orientation of the parent and transformed lattices. Either the new phase appears in a completely random orientation relative to the original phase or else specific orientation relations exist between the two lattices. For the former possibility there is no restriction on the number of allowed domain orientations whereas, in the latter, the number of allowed orientations of the new phase is restricted to the number of independent ways in which the orientation relations can be realized. It follows that a single crystal may transform irreversibly into a randomly oriented powder or reversibly into a fixed number of domain orientations, depending on the thermodynamic parameters and the transformation mechanism. The multidomain crystal has been termed a 'hybrid' single crystal (Ubbelohde & Woodward, 1945*a, b*; Ubbelohde, 1956) because the average properties remain those of a single crystal. Early examples of crystals exhibiting such behaviour were ammonium chloride (Dinichert, 1944), potassium dihydrogen phosphate and arsenate (Ubbelohde & Woodward, 1945*a, b*) and Rochelle salt (Ubbelohde & Woodward, 1947). Many more examples are now known including other ferroelectric crystals (Megaw, 1957) and some alkali metal cyanides (Verweel & Bijvoet, 1940; Bijvoet & Lely, 1940; Cimino, Parry & Ubbelohde, 1959).

When a single crystal undergoes a transformation into its 'hybrid' form, the X-ray diffraction pattern of the crystal undergoes characteristic change corresponding to the transformation of the original reciprocal lattice into a composite lattice formed by the superposition of the reciprocal lattice of the new phase in all allowed orientations. In consequence a single Bragg reflexion of the original phase splits into a group of reflexions whose number and relative disposition is determined by the parameters of the new

lattice, its orientation relative to the original lattice and the diffraction geometry. In principle, granted that there is no limitation on the amount of time available for the investigation, all details of the composite reciprocal lattice can be obtained by the use of existing techniques such as precession or Weissenberg photographs. If, on the other hand, the maximum amount of information is required quickly, then undoubtedly the simple oscillation photograph is to be preferred. The present paper is concerned with a method for the detailed interpretation of such oscillation photographs from 'hybrid' crystals.

### The composite reciprocal lattice

If the volume of the individual domains in a 'hybrid' crystal is sufficiently small, then each allowed domain orientation will be equally probable and the point-group symmetry of the original and composite reciprocal lattices should be identical. For the 'hybrid' crystal, this reciprocal-lattice symmetry, which will be subsequently referred to as the transformation symmetry, comprises:

- (1) The intradomain point-group symmetry, i.e. those symmetry elements of the original point group which are preserved throughout the transformation.
- (2) The interdomain symmetry (not in general a point group) i.e. those symmetry elements of the original point group which are not present in the intradomain symmetry.

Only the interdomain symmetry operations give rise to different domain orientations, the intradomain point group merely reproducing each domain orientation a certain number of times. It should be emphasized that the intradomain symmetry is not necessarily the true point-group symmetry of the reciprocal lattice of a single domain but only those elements that are unchanged in orientation throughout the

transformation. Any other symmetry elements in the new reciprocal lattice will be reproduced by the interdomain symmetry operators, giving rise to additional elements of symmetry that do not conform to a crystallographic point group. Thus the reciprocal lattice of potassium cyanide transforms from cubic,  $m3m$ , to orthorhombic,  $mmm$ , diffraction symmetry although the intradomain symmetry is only  $2/m$  for the normal transformation  $C \rightarrow R$  and  $\bar{1}$  for the abnormal transformation  $C \rightarrow R'$  to be discussed later.

In all but the simplest of cases, the disposition of reciprocal-lattice points (r.l.p.) in the composite reciprocal lattice will be too complex to be handled conveniently by conventional graphical methods, and in the method to be described therefore the composite reciprocal lattice is represented analytically. The general reciprocal-lattice vector,  $\mathbf{H}_1$ , of the reference domain is resolved into components parallel to three reference axes. These axes are conveniently chosen to be the principal axes of the original point group, for this simplifies the subsequent application of the interdomain symmetry operators. Thus for a cubic crystal, the reference axes  $x, y, z$  are taken parallel to the crystallographic axes of the unit cell. Any reciprocal-lattice vector in the cubic phase  $\mathbf{H}_0$  can then be written:

$$\mathbf{H}_0 = h\mathbf{a}_0^* + k\mathbf{b}_0^* + l\mathbf{c}_0^* .$$

After transformation, this same vector in the reference domain changes to

$$\mathbf{H}_1 = h\mathbf{a}_1^* + k\mathbf{b}_1^* + l\mathbf{c}_1^* .$$

The new reciprocal-lattice vectors  $\mathbf{a}_1^*, \mathbf{b}_1^*, \mathbf{c}_1^*$ , are not necessarily the simplest vectors of the new reciprocal lattice but merely represent the change in orientation of  $\mathbf{a}_0^*, \mathbf{b}_0^*$  and  $\mathbf{c}_0^*$ . The vector  $\mathbf{H}_1$  can be projected on to the axes  $x, y, z$  to give the coordinates  $X, Y, Z$  of the corresponding r.l.p. Thus

$$\begin{aligned} X &= (1/a_0^*) \mathbf{H}_1 \cdot \mathbf{a}_0^* , \\ Y &= (1/b_0^*) \mathbf{H}_1 \cdot \mathbf{b}_0^* , \\ Z &= (1/c_0^*) \mathbf{H}_1 \cdot \mathbf{c}_0^* . \end{aligned}$$

The coordinates of the *same* r.l.p.,  $hkl$ , in an alternative domain orientation, can be derived by operating upon the coordinates for the reference domain with the symmetry operator  $\mathbf{S}$  that relates the two domain orientations, so that

$$X'Y'Z' = \mathbf{S} \cdot (XYZ) .$$

By referring each allowed domain orientation in turn to the reference domain, this procedure will generate the coordinates of all identical r.l.p.,  $hkl$ , in the composite reciprocal lattice.

### Practical procedure

However, although this sequence of operations is useful for describing the way in which the composite

reciprocal lattice can be generated, it is not the most convenient way of obtaining the results in practise. Then the problem is that a particular reflexion,  $hkl$ , of the high-symmetry form transforms into a 'split' reflexion, the components of which must be identified by their crystallographic indices and also, possibly, by the domain orientation from which they originate. What is required, therefore, is not the coordinates of the same r.l.p. in different regions of the composite reciprocal lattice, but the coordinates of the different r.l.p. that form the components of a 'split' group in the same region of reciprocal space. To find these coordinates requires a simple modification of the procedure described above.

Before transformation, any two r.l.p. common to the form  $\{hkl\}$  are related by certain elements of point-group symmetry. Thus if  $hkl$  is taken as the reference plane, then any other plane in the form can be represented by  $\mathbf{S} \cdot (hkl)$  where  $\mathbf{S}$  is some symmetry operator (or set of symmetry operators). For example if  $\mathbf{S} = 4^1$ , then  $\mathbf{S} \cdot (hkl) = (khl)$ . After transformation the r.l.p.,  $hkl$ , will have coordinates  $X_I Y_I Z_I$  relative to the reference axes, while  $\mathbf{S} \cdot (hkl)$  will have coordinates  $X_S Y_S Z_S$  when referred to the same axes, i.e. within the same domain. If now the reference axial system is operated upon by  $\mathbf{S}^{-1}$ , then  $\mathbf{S}^{-1} \cdot (X_S Y_S Z_S)$  will represent the coordinates of the r.l.p.  $\mathbf{S} \cdot (hkl)$  in a domain related by symmetry  $\mathbf{S}^{-1}$  to the original domain. In the high symmetry crystal structure, the coordinates  $X_I Y_I Z_I$  and  $X'_S Y'_S Z'_S = \mathbf{S}^{-1} \cdot (X_S Y_S Z_S)$  are identical, but if  $\mathbf{S}$  is an element of *interdomain* symmetry after the crystal has transformed, then this will no longer be the case and the original r.l.p. will be 'split'.

As an example of this procedure, suppose that for the reference domain

$$\begin{aligned} X_I &= ha_{11} + ka_{12} + la_{13} \\ Y_I &= ha_{21} + ka_{22} + la_{23} \\ Z_I &= ha_{31} + ka_{32} + la_{33} \end{aligned}$$

then when  $\mathbf{S} = 4^1$ ,  $h \rightarrow k$ ,  $k \rightarrow \bar{h}$ , and

$$\begin{aligned} X_S &= ka_{11} - ha_{12} + la_{13} \\ Y_S &= ka_{21} - ha_{22} + la_{23} \\ Z_S &= ka_{31} - ha_{32} + la_{33} . \end{aligned}$$

Now  $\mathbf{S}^{-1} = 4^3$  so that  $X'_S = -Y_S$ ,  $Y'_S = X_S$ , and

$$\begin{aligned} X'_S &= ha_{22} - (ka_{21} + la_{23}) \\ Y'_S &= ka_{11} - (la_{13} - ha_{12}) \\ Z'_S &= la_{33} - (ha_{32} - ka_{31}) . \end{aligned}$$

### Indexing of 'split' reflexions

The method described above provides one set of coordinates  $X'_S Y'_S Z'_S$  for each domain orientation in the hybrid crystal and for each set of indices,  $hkl$ . In simple cases where the rotation axis of the crystal is parallel to one of the reference axes,  $x, y$  or  $z$ , then one coordinate, say  $Z'_S$ , can be immediately inter-

preted as  $\zeta$  while  $\xi$  can be expressed as  $[(X'_S)^2 + (Y')^2]^{\frac{1}{2}}$  and this procedure is appropriate in the examples that follow.

Although there is no reason why the subsequent indexing should not follow a completely analytical procedure, it has been found more convenient to use the normal graphical method in practice. In the analytical method, the azimuthal angle  $\varphi$  between the X-ray beam and one of the reference axes, ( $y$ ), defines the limits of oscillation of the crystal,  $\varphi_1$  and  $\varphi_2$ . For a reflexion to occur within this oscillation range,  $X'_S$  and  $Y'_S$  must satisfy the following relation

$$\cos \varphi_1 \mp [1 - \zeta^2 - (X'_S - \sin \varphi_1)^2]^{\frac{1}{2}} \\ \mp Y'_S \mp \cos \varphi_2 \mp [1 - \zeta^2 - (X'_S - \sin \varphi_2)^2]^{\frac{1}{2}} \quad (\varphi_1 < \varphi_2)$$

where  $X'_S$  has positive or negative values according to the sign of  $h$ ,  $k$  and  $l$ . Hence knowing  $X'_S$  and  $\zeta (=Z'_S)$ , it is possible to decide whether a particular r.l.p. could have reflected within the oscillation range.

In practice, allowance has to be made for the fact that  $\varphi_1$  and  $\varphi_2$  are known only to certain limits of accuracy and for the possible spread of the r.l.p. in reciprocal space due to domain size effects. Although it is possible to allow for both these effects to some extent, it was found simpler in the case of potassium cyanide to use the normal graphical method, drawing the two extreme positions of the reflecting circle in the correct orientation relative to a square grid which represented the  $x$  and  $y$  axes and on which any particular r.l.p. could be located.

#### Application of the method to diffraction photographs of KCN

In order to illustrate the way in which the method can be used in practice, one crystal of potassium cyanide, which during a recent investigation (Cimino, Parry & Ubbelohde, 1959) was found to give an abnormal diffraction pattern after transformation, will be considered in detail. Only a few diffraction photographs were recorded before this crystal was accidentally destroyed, but by interpreting these in detail it was possible to confirm that this crystal had undergone the normal structural transition, the abnormal feature being the crystallographic relations controlling the orientation of the domains relative to the original cubic lattice.

The structural change which occurs at about  $-106^\circ\text{C}$ . has already been described (Bijvoet & Lely, 1940; *Structure Reports*, 1955). Briefly, the cyanide groups assume parallel orientation causing the face-centred cubic lattice of the high temperature form to shear through an angle of about  $11^\circ$  giving an  $F$ -centred monoclinic cell with  $a=c$ . The smallest crystallographic unit cell is therefore orthorhombic: one axis is parallel to the  $b$  axis of the monoclinic cell

while the other two axes are the semidiagonals of  $(101)_m$ .†

The normal transformation  $C \rightarrow R$  takes place in such a way that, of the three faces of the new unit cell in its monoclinic representation, two remain parallel to faces of the original cubic unit cell. Thus although  $(100)_m$  and  $(001)_m$  are structurally equivalent, only one remains parallel to its orientation in the cubic crystal. In order that this face should be distinguishable, it was consistently taken to be  $(001)_m$ . The orientation relations for the reference domain are therefore:

$$(001)_c \parallel (001)_m \\ [100]_c \parallel [100]_m$$

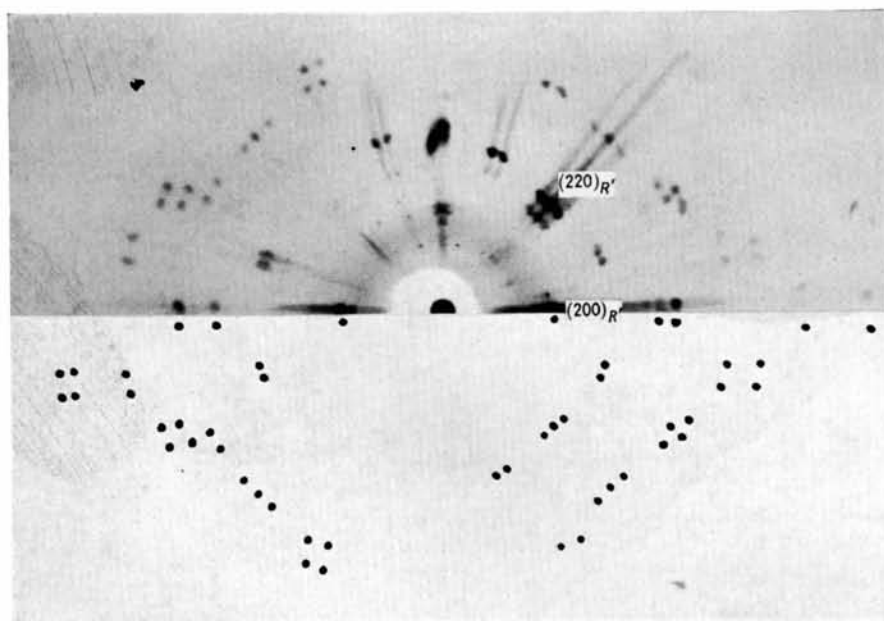
If the reference axes  $x, y, z$ , are taken parallel to  $a_m^*$ ,  $b_m^*$  and  $c_m^*$  respectively, the composite reciprocal-lattice coordinates of a r.l.p.,  $hkl$ , originating from the reference domain, are

$$X = ha_m^* \sin \beta^* \\ Y = kb_m^* \\ Z = lc_m^* + ha_m^* \cos \beta^*$$

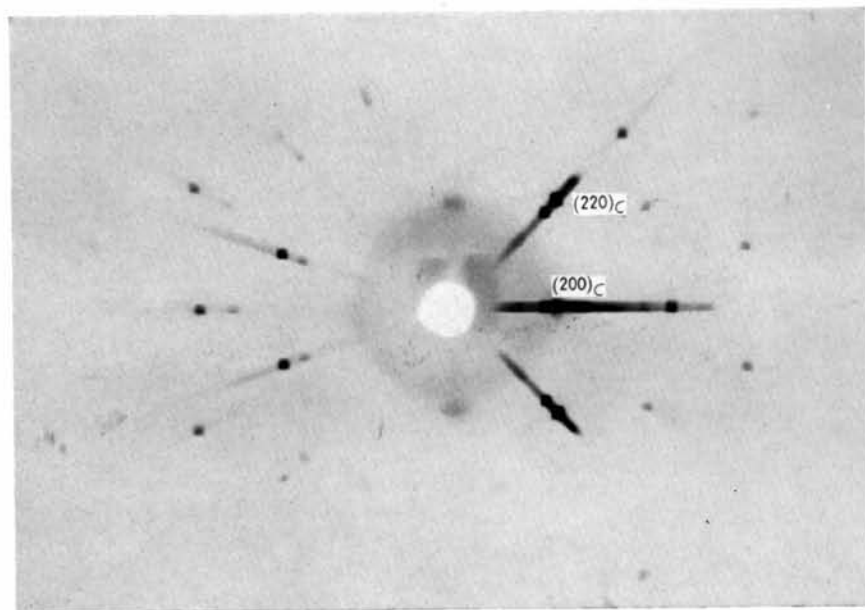
The intradomain symmetry follows from the orientation relations and is  $2/m$  (Fig. 2). Thus the 48 general r.l.p.,  $hkl$ , in the cubic reciprocal lattice are related in groups of four by the intradomain symmetry giving rise to  $48/4=12$  independent domain orientations. Hence to define the composite reciprocal lattice completely, it is necessary to select twelve operations of interdomain symmetry,  $S^{-1}$ . One possible selection is shown in Table 1 together with the coordinates  $X'_S Y'_S Z'_S$  that result. If the oscillation axis is taken as  $z$ , it can be seen that for  $(200)_R$ , where  $k$  and  $l$  are both zero, most components have  $\zeta (=Z'_S)=0$ , i.e. they lie on the zero layer line, in general agreement with experiment.

However, in the abnormal transformation,  $C \rightarrow R'$ , quite a different diffraction pattern was obtained, in which the components of  $(200)_{R'}$  lay on two layer lines that were symmetrically disposed relative to the equator (Fig. 1). The possibility that this crystal had transformed to a new crystal structure was briefly considered but was rejected as highly improbable; the transformation temperature was, within the limits of error ( $\pm 0.3^\circ\text{C}$ .), identical with that of crystals that behaved normally. The more likely explanation was considered to be that the crystal structure was that of the normal low-temperature form but that the orientation of domains relative to the original cubic lattice was different. Assuming this to be so, it was realized that the transformation symmetry could not

† In what follows, the suffix  $c$  will refer to the cubic single crystal, the suffix  $m$  to the transformed unit cell in the monoclinic representation and the suffix  $R$  or  $R'$  to the complete assembly of domains into which the single crystal has transformed.



(a)



(b)

Fig. 1. (a) Observed and calculated diffraction patterns of the one crystal of potassium cyanide which showed the transformation  $C \rightarrow R'$ . The calculated diffraction pattern is based on the domain orientation relations discussed in the text. (b) Corresponding diffraction pattern of another crystal taken after it had reverted to its cubic form at room temperature.

Table 1

Domain number	Interdomain symmetry $S^{-1}$	S. ( $hkl$ )	$S^{-1} \cdot (X_S Y_S Z_S)$ $= X_{S'} Y_{S'} Z_{S'}$	$X_{S'}$	$Y_{S'}$	$Z_{S'}$
1	$I$	$hkl$	$X_S Y_S Z_S$	$hA_1$	$kB$	$lC + hA_2$
2	$4^3_z$	$k\bar{h}l$	$\bar{Y}_S X_S Z_S$	$hB$	$kA_1$	$lC + kA_2$
3	$4^2_z$	$\bar{h}kl$	$\bar{X}_S Y_S Z_S$	$hA_1$	$kB$	$lC - hA_2$
4	$4^1_z$	$\bar{k}hl$	$Y_S \bar{X}_S Z_S$	$hB$	$kA_1$	$lC - kA_2$
5	$3^2_{xyz}$	$lhk$	$Y_S Z_S \bar{X}_S$	$hB$	$kC + lA_2$	$lA_1$
6	$4^1_x$	$\bar{h}lk$	$X_S Z_S \bar{Y}_S$	$hA_1$	$kC + hA_2$	$lB$
7	$3^2_{x\bar{y}z}$	$\bar{l}hk$	$\bar{Y}_S Z_S \bar{X}_S$	$hB$	$kC - lA_2$	$lA_1$
8	$4^3_x$	$h\bar{l}\bar{k}$	$X_S \bar{Z}_S Y_S$	$hA_1$	$kC - hA_2$	$lB$
9	$3^1_{xyz}$	$klh$	$Z_S \bar{X}_S Y_S$	$hC + kA_2$	$kA_1$	$lB$
10	$2_{xz}$	$\bar{l}\bar{k}h$	$Z_S \bar{Y}_S \bar{X}_S$	$hC + lA_2$	$kB$	$lA_1$
11	$3^1_{x\bar{y}z}$	$\bar{k}lh$	$Z_S \bar{X}_S Y_S$	$hC - kA_2$	$kA_1$	$lB$
12	$4^3_y$	$lkh$	$Z_S Y_S \bar{X}_S$	$hC - lA_2$	$kB$	$lA_1$

$$A_1 = a^*_m \sin \beta^*$$

$$A_2 = a^*_m \cos \beta^*$$

$$B = b^*_m$$

$$C = c^*_m$$

be cubic for  $(220)_c$ † transformed into three component reflexions, each with the same  $\theta$  value, and it was verified that the missing reflexions were  $(202)_m$  and  $(20\bar{2})_m$ . Hence it was first necessary to decide what the transformation symmetry might be.

The crystal was initially mounted on the goniometer with one cubic axis parallel to the oscillation axis and one (100) face perpendicular to the X-ray beam in the  $0^\circ$  position. From this position three oscillation photographs were recorded, covering the range of azimuthal angle from  $0^\circ$  to  $45^\circ$ . Laue photographs of the crystal taken in the  $0^\circ$  position showed  $4mm$  diffraction symmetry after transformation. Apart from the cubic point group  $m\bar{3}m$ , the only other centrosymmetric point group with  $4mm$  as a sub-group is the tetragonal group  $4/mmm$ . Assuming this to be the transformation symmetry, the unique axis of the group must be parallel to the X-ray beam when the crystal is in the  $0^\circ$  position.

The existence of such a unique direction was taken to mean that only one of the reciprocal-lattice vectors of the transformed cell lay on or near this direction. The possibility that this vector was  $b^*_m$  was eliminated by the observation that  $(202)_m$  and  $(20\bar{2})_m$  were missing from  $(220)_{R'}$ , while  $a^*_m$  was ruled out because the divergence of  $a^*_m$  from  $a^*_c$  would be expected to lead to an additional splitting of  $(h0l)_{R'}$  in the  $\zeta$  direction, which was not observed. Hence it was tentatively concluded that  $c^*_m$  was the unique axis and that as in the  $C \rightarrow R$  transformation,  $c^*_m$  remained parallel to  $c^*_c$ . Support for this conclusion came from the observation that a new reflexion, whose  $\xi$  value corresponded either to  $(200)_m$  or  $(002)_m$ , appeared on the

opposite side of the main beam to  $(020)_m$  during an oscillation through the range  $0-15^\circ$ . This is understandable if the reflexion is indexed as  $(\bar{2}00)_m$  because the angle between the reciprocal lattice lines  $(\bar{h}00)_m$  and  $(\bar{h}00)_c$  is about  $11^\circ$  and it was verified on a reciprocal lattice that this orientational change would suffice to allow  $(\bar{2}00)_m$  to reflect during this oscillation range.

One orientation relation was therefore assumed to be  $(001)_c \parallel (001)_m$  while the second relation was suggested by the fact that  $(200)_{R'}$  comprised three components. This is possible only if  $[110]_m$  or  $[\bar{1}\bar{1}0]_m$  remains parallel to  $[110]_c$ . In the former case there are two orientations of  $(220)_m$  (as  $a^*_m \neq b^*_m$ ) whereas in the latter there is only one possible orientation as  $(220)_m$  lies in this zone. The same argument should apply to all reflexions with  $h=k$  and it was verified by inspection that  $(222)_{R'}$  was in fact split into two groups of three reflexions, the angular separation of the triplets being similar to that of  $(220)_{R'}$ .

Hence the preliminary examination of the photographs suggested that in this transformation  $C \rightarrow R'$ , one particular face, (001), and one of the two face diagonals of (001) remain parallel to their original

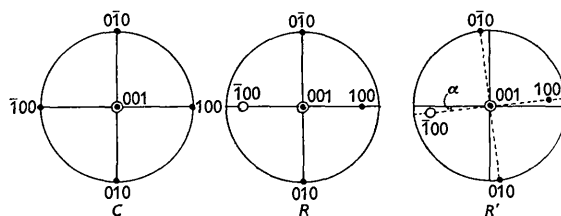


Fig. 2. Stereographic projections of domain orientations in the transformation schemes of potassium cyanide. (a) Original cubic crystal. (b) One of the twelve orientations in the transformation  $C \rightarrow R$ . (c) One of the eight orientations in the transformation  $C \rightarrow R'$ .

† For the indices to conform with the convention previously adopted it is necessary to take the oscillation axis for this crystal to be  $y$  and not  $z$  as before.

Table 2

Domain number	Interdomain symmetry $S^{-1}$	$S, (hkl)$	$S^{-1}, (X_S Y_S Z_S)$ $= X_S' Y_S' Z_S'$	$X_S'$	$Y_S'$	$Z_S'$
1	$I$	$hkl$	$XYZ$	$hA_1 + kB_2$	$kB_1 - hA_2$	$lC + hA_3$
2	$4^3$	$\bar{k}hl$	$Y\bar{X}Z$	$hB_1 + kA_2$	$kA_1 - hB_2$	$lC + kA_3$
3	$4^2$	$\bar{h}kl$	$\bar{X}YZ$	$hA_1 + kB_2$	$kB_1 - hA_2$	$lC - hA_3$
4	$4^1$	$k\bar{h}l$	$\bar{Y}\bar{X}Z$	$hB_1 + kA_2$	$kA_1 - hB_2$	$lC - kA_3$
5	$m_x$	$\bar{h}kl$	$\bar{X}YZ$	$hA_1 - kB_2$	$kB_1 + hA_2$	$lC + hA_3$
6	$m_{xy}$	$khl$	$YXZ$	$hB_1 - kA_2$	$kA_1 + hB_2$	$lC + kA_3$
7	$m_y$	$\bar{h}kl$	$X\bar{Y}Z$	$hA_1 - kB_2$	$kB_1 + hA_2$	$lC - hA_3$
8	$m_{\bar{xy}}$	$k\bar{h}l$	$\bar{Y}\bar{X}Z$	$hB_1 - kA_2$	$kA_1 + hB_2$	$lC - kA_3$

$$A_1 = a_m^* \sin \beta^* \cos \alpha \quad B_1 = b_m^* \cos \alpha$$

$$A_2 = a_m^* \sin \beta^* \sin \alpha \quad B_2 = b_m^* \sin \alpha$$

$$A_3 = a_m^* \cos \beta^* \quad C = c_m^*$$

$$\alpha = \tan^{-1}(a/b) - \pi/4$$

orientation during the transformation. This information is sufficient to enable the composite reciprocal lattice to be defined and these conclusions to be confirmed in detail.

Stereographic projections of one domain orientation in  $R$  and  $R'$  are compared in Fig. 2. The reference axes,  $x, y, z$  were chosen as before and the coordinates of  $(hkl)_m$  for the reference domain in  $R'$  can then be shown to be

$$X = ha_m^* \sin \beta^* \cos \alpha + kb_m^* \sin \alpha$$

$$Y = -ha_m^* \sin \beta^* \sin \alpha + kb_m^* \cos \alpha$$

$$Z = lc_m^* + ha_m^* \cos \beta^*$$

where

$$\alpha = \tan^{-1}(a/b) - \pi/4.$$

The intradomain symmetry of this transition is lowered to  $\bar{1}$  so that the 16 general positions in the point group  $4/mmm$  are associated in pairs giving rise to 8 domain orientations. Elements of interdomain symmetry,  $S^{-1}$ , that conveniently specify these domain orientations are given in Table 2, together with the reciprocal-lattice coordinates that result. Taking  $Y_S'$  as  $\zeta$  and  $[(X_S')^2 + (Z_S')^2]^{\frac{1}{2}}$  as  $\xi$  it was confirmed that this composite reciprocal lattice accounted in detail for all reflexions observed on the photographs when the dimensions of the reciprocal cell for the normal low-temperature form were used. Thus the identity of the unit cell of each domain with that in the normal transition  $C \rightarrow R$  was confirmed and the orientation relations specified earlier were justified. As the  $\zeta$  and  $\xi$  coordinates of the 'split' reflexions on the diffraction pattern were difficult to measure with sufficient accuracy, considerable emphasis was given, when

indexing, to the correct geometrical appearance of a group of split reflexions and to the way in which this varied with  $h, k$  and  $l$ . Evidence of the correctness of this interpretation is given therefore not in tabulated form, but as a comparison of an observed photograph with that predicted on the basis of the composite reciprocal lattice that has been deduced (Fig. 1). The measure of agreement is such as to leave no doubt that the composite reciprocal lattice for this transformation has been correctly defined.

The physical significance of these new orientation relations has been discussed elsewhere, (Cimino & Parry, 1961).

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