Misorientation Contrast of Crystal Subgrain Boundaries in Berg–Barrett X-ray Micrographs

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(Received 14 January 1974; accepted 3 August 1974)

A stereographic projection method of analysis has been developed for analyzing the misorientation contrast of crystal subgrain boundaries which are observed in Berg–Barrett X-ray micrographs. The boundary appearance is described in terms of the geometry of the boundary with respect to the crystal surface, the angular misorientation of the adjacent subgrains, and the X-ray diffraction conditions. The rotation axis for the misorientation of adjacent subgrains, including the sense of the rotation, and the amount of misorientation is determined from the width and relative contrast of the boundary in various images. Experimental results are described for subgrain boundaries in zinc crystals solidified along different crystallographic growth directions. The Burgers vectors of the dislocations composing the subgrain boundary are indirectly determined by this method.

Subgrain boundaries in X-ray diffraction images

Dislocation subgrain boundaries are easily observed in X-ray micrographs such as those which are obtained with the Berg–Barrett technique (Newkirk, 1959; Schultz & Armstrong, 1964; Wu, Arnstein & Armstrong, 1971). Fig. 1(a) and (b) is an example of the subgrain boundary (and dislocation) structure which is revealed in 10T3 and 1013 reflections obtained with Co Ka radiation from matching (0001) cleavage surfaces of a zinc crystal. In each micrograph the boundaries are recognized either as prominent black bands of relatively greater X-ray intensity or as white bands with occasionally resolved black edges. Two different X-ray effects are responsible for the boundaries being observed. In the first instance, the misorientation between adjacent subgrains generally requires that different incident and diffracted beam directions are needed to satisfy the Bragg condition in each subgrain so that the X-ray intensities overlap each other to produce a dark track in the X-ray image or are separated so as to produce a region of zero reflected X-ray intensity. In the second instance, the strain fields of the dislocations within a subgrain boundary affect the integrated intensity of reflected X-rays within a narrow zone along the boundary so that an enhanced X-ray intensity occurs to delineate the boundary position (Newkirk, 1959).

The image contrast associated with dislocations within a subgrain boundary is directly related to the enhanced X-ray intensity which is observed for individual dislocations within the subgrain volumes or within otherwise perfect crystals. This contrast has been described for the Berg–Barrett technique by Newkirk (1959), Chikawa & Newkirk (1967), and Weismann (1968), and for the Lang (1959) X-ray technique most recently by Authier (1970) and Kato & Patel (1973). The dislocation images within an annealed subgrain boundary are predictably very narrow because of the complementary strain fields of dislocations (Lang, 1964; Futagami, 1967). Misorientation contrast is not described nearly so well, particularly not with regard to its being useful for determining the axis of rotation (including its sense) for the misorientation of adjacent subgrains. It is the purpose of this report to describe an analysis for specifying the misorientation contrast of subgrain boundaries; and to demonstrate the utility of the analysis for determining, mainly, the various misorientation axes of subgrain boundaries in zinc crystals solidified along different crystallographic directions.

Misorientation contrast via the stereographic projection

The crystal of Fig. 1 was solidified along [0001]. Furthermore, a number of the boundaries in this figure are observed to run parallel or nearly parallel to (1010), (1120), and other low-index directions. The boundary positions in the crystal cross section change very little along the crystal-growth axis and so they constitute essentially a lineage structure (Chalmers, 1962) of {1120}, {1010} and other low-index boundary surfaces.

The reversed white-to-black contrast of certain boundaries, such as AB, AC, AD and AE, in the matching micrographs of Fig. 1(a) and (b) should seem a natural observation on the basis of the description which has been given for misorientation contrast. The X-ray result follows from the observation that a simple tilt, say, about any [hkl0] will be reversed when viewed from opposite sides of the same (0001) surface. In the general case, however, the appearance of a subgrain boundary in an X-ray image is dependent on the total diffraction geometry, including in addition to the
boundary misorientation the relative position and shape of the X-ray source, the orientation of the diffracting plane, the crystal surface orientation, and the orientation of the boundary itself.

Consider the combined X-ray and crystal geometries which are shown on the stereographic projection (Newkirk, 1959; Armstrong & Wu, 1973) of Fig. 2. In this case a section of an [0001] standard projection is shown for a cleaved zinc crystal containing two subgrains, designated 1 and 2. The subgrains are misoriented by 10° counterclockwise rotation about their common [0001] as shown by the [10T0]1, and [10T0]2 positions at the eastern region of the equator. The trace of the subgrain boundary, containing the [0001] growth direction, is taken to lie along an average [01T0] between the two subgrains. Also shown near the center of the projection are the various poles, labeled {10T3}N, for which favorable Berg–Barrett micrographs may be taken with Co Kα radiation. To obtain the 10T3 Berg–Barrett reflection one initially orients the crystal so that the [0001] is perpendicular to the incident X-ray beam and the [1210] is vertical. The crystal is then rotated through an angle, α, around the average [1210] to satisfy the Bragg condition for 10T3. In Fig. 2, however, the position of the X-ray beam source is shown to be rotated through the angle α(=6.1°) for convenience of analysis. Following Newkirk (1959), the 10T3 reflecting circle is drawn on the projection at an angle of [(π/2)−θB] from the X-ray source, where θB, the Bragg angle, is 41.6° for Co Kα radiation. The reflecting circle is shown to have finite angular dimensions which for illustration are somewhat larger, say, 2° than would normally be employed in an actual diffraction experiment. For this case, both the (10T3)N1 and (10T3)N2 fall easily within the reflecting circle, thus allowing subgrains 1 and 2 to give simultaneous 10T3 reflections.

The diffracted beams from the two subgrains are found from tracing along great circles containing the X-ray source and the respective poles, at angles of [(π/2)−θB] from each of them, as marked for the positions 1' and 2' in Fig. 2. The appearance of the subgrain boundary is found in the X-ray image from considering that the boundary in the crystal surface is to be viewed from the two diffracted beam directions and, therefore, the angular rotation which is required to place the [0001] surface normal at the diffracted beam positions, 1' and 2', determines the angular displacement in the stereographic projection of all points on the boundary for each subgrain. In this way, the points R and S on the subgrain boundary are shifted to their respective pairs of image positions, R1, R2 and S1, S2, as shown in the figure. Because the image position of subgrain 2' is displaced to the left relative to 1', as compared with the actual subgrain positions, it must occur that a black band of overlapping X-ray intensities from the two subgrain volumes is recorded at the boundary position in the X-ray image. Of course, adjacent points of the subgrain volumes at the boundary are actually shifted parallel to the boundary as well as perpendicular to it. This boundary appearance is due solely to the misorientation of the subgrain
volumes and, hence, is termed misorientation contrast. The combined operations which are involved in determining the boundary appearance in the X-ray image demonstrate the dependence of misorientation contrast on the total diffraction geometry. The misorientation contrast, thus described, is clearly independent of the contrast produced by dislocation strain fields. Though Fig. 2 is determined for a symmetric misorientation of the subgrains about [0001] and a symmetric position of the subgrain boundary between them, no significant differences seem to result from considering asymmetric situations.

An analysis similar to that which has been described for Fig. 2 has been applied to various boundary orientations having a fixed misorientation of 10° rotation of subgrain 2 with respect to subgrain 1 about [0001], as shown in Table 1. The boundary orientation of Fig. 2 is shown as the 10T3 reflection in the second row, second column, of Table 1. For this particular case, alternating black and white boundary images are indicated for pairs of lattice reflections which are rotated 180° from each other; however, the black versus white sequence of images which occur for consecutive pairs of images rotated 60° about [0001] is not nearly so obvious. The relative shift in the X-ray images of adjacent points along the boundary direction in (0001) is indicated in each image by the generally oblique cut-off edge determining the length of the various image segments.

The importance to image contrast of the misorientation axis for the subgrain rotations is demonstrated by comparing the foregoing results with those which are obtained for a subgrain boundary whose rotation axis is contained within (0001), say [10T0], as shown for subgrains 1 and 2 in Fig. 3. In this case, the subgrain misorientation is taken as 2°, which is directly comparable to the divergence of the incident X-ray beam. The procedure for determining a boundary image is indicated in Fig. 3 for the 1013 and 01T3 reflections.

The boundary is observed in each instance to appear white because of the separation of the reflections from the adjacent subgrain volumes. These results are shown in Table 2 together with those results which were obtained for various boundary orientations, all having this same misorientation across them. The unchanging boundary appearances in Table 2 as compared to those in Table 1 show immediately that a hexad rotation axis for the misorientation may be easily identified.

By the same method of analysis, Table 3 has been produced to show the misorientation contrast associ-
ated with a simple twist boundary, i.e. one for which the rotation axis for the subgrain misorientation is orthogonal to the subgrain boundary surface. A misorientation of 2° rotation about the \((10\overline{1}0)\) for an \(\{10\overline{1}0\}\) boundary surface, or 2° rotation about \((11\overline{2}0)\) for an \(\{11\overline{2}0\}\) boundary surface, has been employed. In this case, the shape and size of the X-ray source and, consequently, the divergence of the X-ray beam is especially important in determining the nature of the misorientation contrast. For Table 3 a square X-ray source whose angular width is 2° has been utilized so as to allow both subgrain volumes to diffract for all reflections. Interestingly enough, several of the boundary images which are shown in Table 3 are also strikingly different from those presented in the previous tables.

**Experimental observations**

The preceding stereographic-projection analysis of misorientation contrast gives a description of the boundary appearance in terms of its angular width whereas any subgrain boundary which is observed in an X-ray micrograph has an actual width that is proportional both to this angular displacement and to the distance between the crystal surface and the X-ray film. The dependence of the measured boundary width on the boundary has been described for a simple tilt boundary (Wu, Arnstein & Armstrong, 1971; Kranjc, 1968).

Within a micrograph, it should be expected that the boundary image would be different according to the changing boundary location. This effect is, however, a generally negligible consideration with respect to detecting any alteration in the appearance of a particular boundary because the maximum change in boundary width per unit boundary length is given by the product \((\Delta \theta) (\sin \alpha)\) which, for \(\Delta \theta \approx 2°\) and \(\alpha = 6°\), has a value of \(\sim 4 \times 10^{-3}\). This explains why no change in boundary appearance is observed along the length of a boundary even at the relatively low magnification of the boundary images shown in Fig. 1(a) and (b).

An example of the major effect of the boundary orientation on misorientation contrast is shown for the single offset boundary which is revealed in the X-ray micrograph of Fig. 4. This micrograph shows at a larger magnification an adjacent area of the film plate from which Fig. 1(a) was produced. The diagonal boundary in Fig. 4 [\(\angle \text{AE in Fig. 1(a)}\]] is parallel to \([01\overline{1}0]\) while the offset segment is very nearly parallel to \([10\overline{1}0]\). The abrupt change in boundary direction through an angle of \(30°\) for the offset segment has produced a contrast reversal. The method by which this may be understood is directly modeled in Fig. 5. In this figure, it is shown that the same misorientation between subgrains which produces a reasonably wide angular separation of the subgrain images for one boundary orientation also produces a relatively narrow boundary of overlapping X-ray intensities for another boundary orientation. On the basis of this analysis, the rotation axis for the misorientation of the two subgrains in Fig. 4 is determined to be \([00\overline{1}0]\), that is, of opposite sense to that described for Fig. 2 and Table 1. A neighboring segmented subgrain boundary within this same \([00\overline{1}0]\) axis crystal is shown in Fig. 6. The major portion of this boundary is also parallel to \([01\overline{1}0]\) while the black offset segment is nearly parallel to \([10\overline{1}0]\) and the white reset segment with black edges is parallel to \([\overline{1}00]\). An analysis of the misorientation contrast for this boundary gives a rotation axis, \([00\overline{1}0]\), opposite to that established for the neighboring boundary in Fig. 4.

At one position along the \([\overline{1}010]\) boundary, \(\angle \text{AC}\), shown in Fig. 1(a), a deformation twin with associated accommodation kink boundaries (Armstrong, 1964) was produced by impacting the specimen surface with a

![Fig. 3. Misorientation contrast of subgrain boundary with rotation axis [10\overline{1}0] parallel to the trace of the subgrain boundary in the (0001) surface.](image)

**Table 3. Summary of stereographic projection analysis of the misorientation contrast of a twist boundary**

<table>
<thead>
<tr>
<th>Subgrain Boundary Orientation</th>
<th>Rotation Axis</th>
<th>Misorientation Contrast</th>
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<tbody>
<tr>
<td></td>
<td>[10\overline{1}0]</td>
<td></td>
</tr>
<tr>
<td></td>
<td>[\overline{2}10]</td>
<td></td>
</tr>
<tr>
<td></td>
<td>[1\overline{1}0]</td>
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<td></td>
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<td>[\overline{2}10]</td>
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focused laser beam (Wu, Armstrong & Lee, 1972). The deformation twin crossed the subgrain boundary and thereby served as a special marker for measuring the displacement of subgrain images both parallel to and perpendicular to the direction of the boundary. The results for observation of this twin-boundary intersection in six \{10\text{13}\} reflections are shown in Fig. 7. An optical micrograph of the deformed region is shown in the central square of the figure surrounded by the appropriately oriented \{10\text{13}\} reflections. The vertical twin lamella runs to the center of the optical micrograph from the top of the figure. An accommodation kink band on the left side of the twin is visible as a relatively wide, dark band. These two features are somewhat obscured in the optical micrograph by the extensive surface roughness produced by microkinking deformation and, also, by oxidation of the specimen surface which occurred as a result of the laser damaging process. The \{01\text{0}0\} boundary is not recognizable in this unetched optical micrograph but the boundary runs horizontally from left to right through the middle of the figure. The 10\text{13} reflection, corresponding to that area in Fig. 1 which was obtained prior to deformation of the specimen, is shown at the far left of Fig. 7. It is observed from all of the X-ray micrographs that the subgrain boundary appears as a black band in the 01\text{13}, 10\text{13} and \text{110}3 reflections and as a white band in the other reflections. These results were compared with those contained in Table 1. The black or white boundary images appear to be reversed from those shown in the fourth row of the table. The maximum boundary image width occurs for the 10\text{13} and 10\text{13} reflections, as predicted. These facts, then, suggest that the rotation axis for the misorientation of the subgrains is \text{[0001]}\text{.} The various features mentioned for Fig. 7 are shown in Fig. 8, which has been produced from the stereographic projection analysis so as to include also the predicted image shifts which occurred parallel to the subgrain boundary. These predicted shifts match nicely with their counterparts in Fig. 7, thus confirming an \text{[0001]} rotation axis for this subgrain boundary misorientation.
Discussion

Determination of the rotation axis for the misorientation across individual subgrain boundaries provides specific information on the geometrical relationship of the subgrains which can be utilized to determine possible dislocation arrangements within the subgrain boundary even when the dislocations themselves are not directly observed. Furthermore, at lines or curves of intersection of annealed subgrain boundaries, an additional condition must be met in that the total misorientation of the subgrains must be conserved so as to prevent the existence of any cumulative long-range stress field due to the dislocations within the terminating subgrain boundaries. The intersection of the four subgrain boundaries $AE$, $AD$, $AC$ and $AB$ at $A$ in Fig. 1 provides an example in which these considerations may be applied. The boundary $AE$ has been identified as having an [0001] rotation axis. The simplest and lowest-energy dislocation array which may account for this rotation would be one having edge dislocations with line vectors along [0001] and Burgers vectors $(a/3)[2110]$ arranged in a tilt boundary. The misorientation contrast of boundaries $AB$ and $AC$ produces the same rotation axis as for $AE$ and therefore, dislocations of Burgers vector $(a/3)[2110]$ and $(a/3)[1210]$ respectively, can be used to account properly for their misorientations. On the basis of its misorientation contrast, boundary $AD$ was found to have a [10$ar{1}0$] rotation axis for which dislocations with line vectors along this direction and Burgers vectors of type $(a/3)[1210]$ will account for this particular rotation. From this information, a three-dimensional picture of the crystal volume in the region of point $A$ has been constructed as shown in Fig. 9. A cleavage step was observed at point $A$ and this surface feature is also shown in Fig. 9.

Until the present time, the stereographic projection analysis has been applied to boundaries having the several low-index directions which have been described. It should be interesting to investigate other types of boundaries, however, even though subgrain misorientations with $\langle0001\rangle$ rotation axes have been observed in the present investigation only for crystals solidified along this same growth direction. Rotation axes of this type do not appear have been previously recorded for the misorientation of subgrains in zinc single crystals of any kind. The $\langle0001\rangle$ oriented crystals could themselves be produced only by melting relatively pure zinc material on to an $\langle0001\rangle$ oriented seed crystal and then resolidifying the metal in a controlled manner to produce cylindrical or square-cross-section crystal rods (Gilman & DeCarlo, 1955). Unseeded single crystals generally exhibit rod axes which are directions contained either within or near to being within $\{0001\}$ (Slifkin, 1951; Damiano & Tint, 1961). Crystals solidified by seeding along $\langle10\bar{1}0\rangle$ and $\langle1\bar{1}02\rangle$ growth directions only exhibit subgrains misoriented about rotation axes within $\{0001\}$, as expected. The observed $\langle0001\rangle$ rotation axes for subgrains in crystals of this same orientation may be taken to indicate that during the solidification process dislocations with line vectors contained within $\{0001\}$ climbed into an $\langle0001\rangle$ line orientation and, because of their mutual interaction, formed subgrain boundaries of this particular misorientation. Dislocation image forces should have
operated to produce this result. The crystal seeding process was accomplished very slowly, say, over a period of one hour and the crystal solidification rate was slow, say, 1 cm h⁻¹ so that the crystal growth conditions seem to have been favorable for the process of dislocation climb to occur. An (0001) orientation of dislocation lines for crystals grown along this same direction would provide a continuing sink for solute to be incorporated into the solidifying crystal (Daimano & Tint, 1961; Hulme, 1954), thereby, lessening to a certain extent the need for nucleation of additional individual dislocations within the material.

The angular magnitudes of the subgrain misorientations are not, in general, directly measurable from the separation distance or from the region of overlapping of the subgrain images in an X-ray micrograph because of the dependence of the subgrain boundary width on the X-ray and crystal geometry. This statement applies even for the method of estimating the misorientation of subgrains by measuring the boundary width at two distances from the crystal surface. This point is further demonstrated in Fig. 10 where the apparent angular separation of the diffracted X-ray beams has been computed with the stereographic projection method for a 5° rotation about an [0001] axis of a tilt boundary whose surface orientation is successively taken to be at four different angular positions with respect to the plane of incidence. The angular distance between the two diffracting plane normals, Δθ', is given by the actual misorientation, Δθ = 5°, multiplied by the sine of χ, the angle between [0001] and the diffracting plane normals, i.e. Δθ' = Δθ sin χ = 5° sin 35° = 3°. This angular distance, when referred to the two great circles through the diffracting plane normals (poles) and the X-ray source gives the maximum apparent angular width between the subgrains, Δθm, in terms of Δθ' and θy as Δθm = 2Δθ' sin θy ≈ 4°. The apparent angular width, Δθm, is determined at different angles, Ω, between the projected incident X-ray beam direction and the subgrain boundary direction in (0001) by Δθm multiplied by cos Ω. The orientation dependence in Fig. 10 is also seen to be consistent with the white versus black boundary widths shown in Fig. 4 and with their explanations which are given in Fig. 5. The divergence of the X-ray beam contributes to either reducing the measured boundary width or increasing it, depending on whether the boundary appears white or black respectively.

Some final discussion should be given on the effect of the divergence of the X-ray beam on the resolution of boundaries. The beam divergence affects in a major way the observation of dislocations. For zinc the effect of the beam divergence is such that essentially zero resolution is obtained with 0.5° divergence at a specimen-film distance of 4 mm. For misorientation contrast, the effect of the beam divergence on resolution of the subgrain boundary is rather insensitive to the specimen-film distance so that only a slight blurring of the subgrain boundary image is observed even at film distances significantly larger than 4 mm. The beam divergence must be sufficiently large to allow for the individual subgrains to diffract X-rays but, beyond this, the divergence of the X-ray beam should be either sufficiently small that the subgrains diffract X-rays from nearly individual points on the X-ray source or, separately, the arcs or diffracted intensity from the X-ray source should be oriented so as to make as small an angle as possible within {0001} with the trace of the subgrain boundary. This latter condition may be seen in Tables 1 through 3 to correspond to a decreasing effect of beam divergence on boundary width as the short crossing segments are rotated towards being parallel to the length of the boundaries.
This research has been supported at the University of Maryland by the Engineering Materials Program of the United States National Science Foundation, Grant GH-38751. The X-ray experiments were performed within the X-ray Central Facility, Center of Materials Research, University of Maryland.

References


