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WITH PARTICULAR REFERENCE TO A RECENT PLANE MATCHING APPROACH*

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ON THE STRUCTURE OF HIGH ANGLE GRAIN BOUNDARIES
WITH PARTICULAR REFERENCE TO A RECENT PLANE MATCHING APPROACH

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Introduction

Recently, Pumphrey(1) has attempted to explain the periodic line structures in grain boundaries, which have been observed by transmission electron microscopy(2-10), on the basis of a plane matching (PM) approach which involves the mismatch between certain atomic planes across the boundary. In this approach attention is focused on families of atomic planes which impinge upon the boundary from the adjoining grains in such a manner that their traces in the boundary are only slightly mismatched, i.e., the spacings and orientations of the sets of parallel traces in the boundary are only slightly different. The situation is as shown in Fig. 1 where the traces of one family of planes entering the bound-

FIG. 1

Slightly mismatched traces in the boundary of planes impinging from adjoining Crystals 1 and 2.

ary from Crystal 1 are present at a spacing s_1 and are slightly rotated by the angle θ with respect to a corresponding set of traces due to planes in Crystal 2. This overall array of traces produces a Moiré pattern which possesses parallel bands of high opacity (relatively bad atomic fit) lying along the dashed

lines. Between these bands are corresponding bands of high transparency (relatively good atomic fit). The spacing, d , between bands (Fig. 1) is given by

$$d = \frac{s_1 s_2}{[s_1^2 + s_2^2 - 2s_1 s_2 \cos \theta]^{\frac{1}{2}}}, \quad (1)$$

and the angle, ρ , between the bands and the traces due to Crystal 1 is given by

$$\rho = \sin^{-1} \left(\frac{d \sin \theta}{s_2} \right). \quad (2)$$

Pumphrey suggests that atomic relaxation occurs in the lattice in the vicinity of these Moiré bands, and that strain fields are then produced which are imaged in the microscope as a single set of parallel lines which has the same geometry as the bands. He then claims that this approach provides a better explanation of the observed boundary lines than the grain boundary dislocation (GBD) model which has been employed previously for this purpose by the writers(5-7,10). This latter model describes the boundary as a fit-misfit structure which is composed of a network of GBD's embedded in a relatively low energy interface which is generally periodic in the boundary plane.

The main purposes of the present note are:

- (1) to show explicitly the relationship between the PM approach and the GBD model.
- (2) to point out various limitations of the PM approach.
- (3) to show that the PM description is actually not an independent "model" but is merely another way of representing the line structures of certain boundaries which are already well accounted for by the more general GBD model.

Low Angle Boundaries

For simplicity we start with a consideration of low angle boundaries. An example consisting of a [001] twist boundary in a fcc structure is shown in Fig. 2. The unrelaxed atomic misfit across the boundary is shown in Fig. 2a where the filled circles represent the atomic positions in the [002] plane in Crystal 1 just above the boundary, and the open circles represent the $(\frac{1}{2}, 0, 0)$ and $(0, \frac{1}{2}, 0)$ interstices in the [002] plane in Crystal 2 just below the boundary. In this case it is well known(5,11) that relaxation in the boundary region occurs which produces large square patches of perfect fit, and the misfit is forced into a square network of lines which is easily shown to be a network of screw GBD's. The boundary is well described, therefore, by the GBD model consisting of a network of GBD's embedded in the otherwise perfect lattice. Now, if we focus attention on the slightly mismatched traces of the (111) planes which run approximately vertically in Fig. 2c, we see that they form Moiré bands running horizontally at an angle

$$\rho \approx \pi/2 \quad (3)$$

FIG. 2

Low angle $[001]$ twist boundary in fcc structure. (a) Unrelaxed misfit across boundary. (b) Relaxed fit-misfit structure. (c) Moiré bands formed by traces of (111) planes in unrelaxed structure. (d) Traces of (111) planes in relaxed structure. (e) Moiré bands formed by traces of (020) planes in unrelaxed structure. (f) Traces of (020) planes in relaxed structure.

and a spacing

$$d \approx \frac{s}{\theta} . \quad (4)$$

These results are predicted by Eqs. (1) and (2) when $s_1 = s_2 = s$ and $\theta \ll 1$. The configuration of these traces in the relaxed boundary configuration is shown schematically in Fig. 2d where it is seen that they have relaxed into a pattern of distortion which reproduces the linear features of the Moiré pattern in Fig. 2c. It is readily seen that the similar relaxation of the slightly mismatched traces of the $(\bar{1}11)$ planes which run approximately horizontally would produce a similar set of lines of distortion which would run vertically and complete a square network. In this case the line structures according to the PM approach and the GBD model are identical and, of course, agree with the linear pattern of the fit-misfit structure (Fig. 2b).

Examination of Fig. 2a shows that there are many other families of boundary traces which are also slightly mismatched. Consider, for example, the traces of the unrelaxed (020) planes in Fig. 2e which produce Moiré bands which run along [010]. These Moiré bands do not reproduce the line structure of the boundary. (The relaxation of these traces produces the square network seen in Fig. 2f which, of course, corresponds to the fit-misfit structure and the GBD network.) The use of these planes in the PM description, therefore, does not reproduce the boundary line structure, and it is obvious that in order to reproduce the correct result, the mismatched traces must be specially selected so that the corresponding Moiré bands run parallel to the misfit lines (GBD's) which the boundary relaxation produces.

There are many boundaries where the PM approach, at least in the simple form proposed by Pumphrey, does not lead to the correct result regardless of which set of mismatched traces are chosen. For example, in the case of a $[111]$ twist boundary it is well known that the fit-misfit structure consists of a hexagonal network of GBD's (11,12). In this boundary the line structure does not coincide with the Moiré bands produced by any sets of mismatched planar traces. In general, the PM description cannot be used in any case where the line structure does not consist of families of straight parallel lines, i.e., Moiré bands. There also does not seem to be any simple way in which the PM approach can be used to describe the complex line structures which develop when foreign (extrinsic) GBD's join the boundary and interact with periodic networks of GBD's (10,13).

We also mention a difficulty which arises in the application of the PM approach to certain tilt boundaries. Consider the classic example of the asymmetric low angle tilt boundary in Fig. 3. In this case two slightly tilted families of planes, each with spacing t , enter the boundary from Crystals 1 and 2 to produce families of traces of spacing s_1 and s_2 in the boundary which run

FIG. 3
Asymmetric low angle tilt
boundary. Tilt angle = α .

parallel to the tilt axis. When $\phi < (90 - \alpha/2)$, these traces produce a parallel Moiré pattern consisting of bands running parallel to the tilt axis. In this case $\theta = 0$, and Eq. (1) yields an expression for the spacing which we write in the form

$$\frac{1}{d} = \left| \frac{1}{s_1} - \frac{1}{s_2} \right| \quad (5)$$

to emphasize the fact that the Moiré bands are due to the difference between $1/s_1$, the density of traces due to planes entering the boundary from Crystal 1 and $1/s_2$, the corresponding trace density associated with Crystal 2. Since $s_1 = t/\cos(\phi - \alpha/2)$ and $s_2 = t/\cos(\phi + \alpha/2)$, we have

$$d = \frac{t}{\alpha \sin \phi} \quad (6)$$

which is just the expression for the spacing of the dislocations with Burgers vector t in such a boundary (14). The PM description and GBD model are therefore in agreement in this case. However, when $\phi > (90 - \alpha/2)$, the flux of planes from Crystal 2 changes sign. The line structure (GBD) density in the boundary is then determined by the total density of traces in the boundary, since every nearly vertical plane which impinges on the boundary produces a line (GBD). In this case no Moiré pattern is produced as a result of the "beating" of the two families of parallel traces against each other. Such a situation is clearly demonstrated in the case of the symmetrical tilt boundary illustrated in Fig. 4a,b. Here all planes from Crystals 1 and 2 intersect the boundary symmetrically (Fig. 4a), and there is no Moiré type mismatch. However, atomic relaxation produces a fit-misfit boundary structure consisting of a stack of edge dislocations, i.e., observable lines running parallel to the tilt axis at a spacing given by $d = t/\alpha$ (15) as illustrated in Fig. 4b. We note that Eqs. (5) and (6) still hold in this case, and the PM approach may still be applied formally by taking the Moiré mismatch as the sum of all the traces in the boundary. However, in our opinion this application of the PM approach is rather artificial.

FIG. 4

Symmetric tilt boundaries in fcc structure. (a) Unrelaxed low angle boundary. (b) Same as (a) except relaxed. (c) Unrelaxed high angle boundary.

High Angle Boundaries

All of the points regarding boundary line structure which were made above for low angle boundaries also hold for high angle grain boundaries.

Consider, for example, the case of a high angle $[001]$ twist boundary in the fcc structure possessing a twist angle of $\theta = 33.5^\circ$. The unrelaxed structure is shown in Fig. 5a. A schematic drawing of the corresponding relaxed structure is shown in Fig. 5b where it is seen that a fit-misfit structure has formed which in this case consists of square patches of relatively good fit which are separated by a square network of linear regions of poor fit. The areas of relatively good fit consist of an interface which is periodic in two dimensions with a relatively small repeat distance given by $a\sqrt{10}/2^*$ where a is the lattice parameter. It has been shown elsewhere(5) that the network of poor fit actually is an orthogonal network of screw dislocations with Burgers vectors of the type $a/10\langle 310 \rangle$. In this particular case $\langle 310 \rangle$ directions in either crystal are nearly parallel, and planar traces in the boundary parallel to these directions form the Moiré pattern shown in Fig. 5c. The geometry of these traces in the relaxed configuration is shown in Fig. 5d, and we see that the PM approach and the GBD model again lead to the same result when the proper traces (i.e., the $\langle 310 \rangle$ traces) are selected. However, in this boundary $\langle 210 \rangle$ traces are also

* The exact atomic configuration in the interface in these regions of good fit is presently unknown. However, the actual configuration is undoubtedly a relaxed form of the structure which we have drawn in Fig. 5b. This structure contains a high density of coincidence sites (one out of every five atoms is in coincidence across the boundary) and corresponds to the unrelaxed interface which is achieved when the rotation around $[001]$ is exactly 36.9° (5). The basic periodicity of this interface is unaffected by any subsequent atomic relaxation, and we have therefore drawn the unrelaxed configuration for purposes of illustration.

FIG. 5

High angle [001] twist boundary in fcc structure. Twist angle = 33.5° . (a) Unrelaxed misfit across boundary. (b) Relaxed fit-misfit structure. (c) Moiré bands formed by traces parallel to $\langle 310 \rangle$ directions in unrelaxed structure. (d) Traces along $\langle 310 \rangle$ directions in relaxed structure. (e) Moiré bands formed by traces parallel to $\langle 210 \rangle$ directions in unrelaxed structure. (f) Traces along $\langle 210 \rangle$ directions in relaxed structure.

nearly parallel, and, as shown in Figs. 5e,f, the use of these traces does not produce a Moiré pattern which reproduces the correct grain boundary line structure.

Numerous observations have been made of line structures in high angle boundaries which do not consist of families of straight parallel lines which reproduce the geometry of Moiré bands (10,13,16-18). Here again, the results are readily interpreted in terms of GBD's, and the PM approach cannot be applied.

An example of a high angle tilt boundary in which the PM approach (as interpreted rather artificially above for the case of the low angle tilt boundary) may be applied is the symmetric tilt boundary shown in Fig. 4c. In this case the observable line structure consists of a family of edge dislocations with Burgers vectors of the type $a/10\langle 310 \rangle$ lying parallel to the tilt axis (7), and the boundary regions between the dislocations consist of areas of relatively good fit which are periodic in two dimensions and which are again derived from a high coincidence site density situation (7). Here, again there is no Moiré type mismatch across the boundary, and yet the correct result may be obtained with the PM approach if the "mismatch" is formally taken as the sum of the fluxes of nearly vertical $\{310\}$ type planes entering the boundary.

Discussion of Selected Experimental Observations

Pumphrey claims that the PM approach leads to an explanation of a number of high angle boundary line structures observed by Lévy and Goux (8), Lévy (9) and Schober and Balluffi (7) which cannot be explained by the GBD model. As might be expected from the foregoing, we believe that all of these structures may be satisfactorily explained on the basis of the GBD model. We also believe that there has been some confusion regarding the nature of this line structure, and we therefore consider these observations briefly.

Lévy and Goux (8) and Lévy (9) observed sets of lines which ran parallel to the $[001]$ tilt axis in nearly symmetrical high angle tilt boundaries in aluminum. These lines were shown to be caused by a small twist component around the grain boundary normal which was present in addition to the predominant tilt component around the tilt axis in the boundary plane. The tilt angles, e.g., 37° , 45° , 50° and 53° in ref. (17), were within a few degrees of relatively high coincidence site density misorientations, and according to the GBD model for such boundaries (17), a hexagonal GBD array should take up the twist deviation for such an interface when the tilt angle corresponds to the critical angle associated with a high coincidence site density misorientation. In a case where the tilt angle deviates from such an orientation, an additional tilt subboundary composed of edge GBD's must be superimposed. The writers believe that in the

work of Lévy and Goux(8,9) neither the tilt subboundaries nor the above hexagonal twist subboundaries (with one notable exception as listed below) were actually imaged in the electron microscope. In particular, Lévy and Goux(8,9) imaged only those elements of the hexagonal GBD networks which ran approximately parallel to [001] and which tended to appear as straight lines under the contrast conditions used. Careful inspection of the micrographs presented by Lévy(9) reveals that on Fig. 2 (bottom left and right pictures) arrays of defects were actually present which were approximately perpendicular to the [001] tilt axis. On the bottom left hand side of Fig. 2 of (9) a hexagonal array of GBD's, i.e. a twist subboundary, is clearly imaged in our opinion. We wish to emphasize that this hexagonal structure is in agreement with the GBD model proposed in (17) as far as orientation and spacing is concerned, and that it cannot be explained by the PM approach(1). In other work, the writers investigated the same type of [001] tilt boundaries in gold(7). In the course of that study numerous twist boundaries of hexagonal geometry were observed in boundaries in the vicinity of various high coincidence site density orientations. Their orientation and spacing again were found to be explicable in terms of the GBD model (Schober and Balluffi, unpublished work), and again the observed structures are at variance with the PM approach. Further aspects of this topic are discussed in (17).

Concluding Remarks

We conclude in general that the boundary line structure is fundamentally determined by the manner in which the three-dimensional grain boundary slab region relaxes to form a fit-misfit structure of minimum energy. The resulting fit-misfit structure may always be accounted for by the GBD model. The structure may be described using the PM approach only in certain cases where sets of mismatched planar traces can be found so that the corresponding Moiré bands run parallel to the dislocation lines which the boundary relaxation produces. We have seen that the PM description is actually not an independent "model" but is merely another way of representing the line structures of certain boundaries which are already well accounted for by the more general GBD model.

Finally, we note the current proliferation of models for high angle grain boundaries which is appearing in the literature. Other models include the "secondary" (or "hierarchy of dislocations") GBD model(19-21), the ledge model (21,22) and the structural unit model(22). It is readily shown that all of these models are essentially equivalent and merely emphasize different aspects of the boundary structure. In our work we have preferred to employ the GBD model, since the topological properties of dislocations and displacement fields around them are well known. This has been useful in problems associated with

the analysis of complicated GBD configurations, the estimate of the boundary energy as a function of crystal misorientation, and the interpretation of electron diffraction contrast from the line structure (i.e., the GBD's).

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