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ATOMIC LEVEL OBSERVATION OF GRAIN BOUNDARIES IN METALS

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Abstract

High resolution microscopy has given an explicit, atomistic insight into the nature of the core structure of grain boundaries in metals. Specific issues which have been addressed and to some extent resolved are the occurrence of multiple structures, the transition from low angle to high angle character, the structural unit concept, the relation between the primary and secondary dislocations in a boundary, the spatial extent of the boundary core and the width of the cores of grain boundary dislocations. Much but not all that has been observed by high resolution microscopy was predicted by atomistic computer modelling.

Introduction

The development and direct validation of a general theory for the atomic structure of grain boundaries has proved to be an elusive goal. Nevertheless there is overwhelming evidence from property measurements that grain boundary structure varies systematically with the crystallographic parameters which define a grain boundary [1]. There is also indirect evidence from the relative rotation of unconstrained crystals that the structure of arbitrary grain boundaries is in some, as yet unspecified sense, related to that of the simple periodic grain boundaries which are accessible to atomistic modelling and high resolution imaging techniques [2, 3]. The purpose of this paper is to assess what has been established by high resolution experimental investigations of grain boundary structure in metals and to indicate some avenues for future work.

The crystallography of interfaces has developed along a path quite distinct from that followed by the classical crystallography of single crystals. In the latter case the nature of the perfect structure was well established long before the existence and importance of defects was suspected. However for interfaces the converse was true. In fact it was the observation of interfacial defects, especially dislocations in grain boundaries, which first served to suggest that grain boundaries, in particular high angle boundaries, did indeed possess some kind of presumably ordered, perturbation resistant structure. A similar sense is emerging in the context of interphase interfaces. There is for example a clear similarity between the dislocation structures of low angle grain boundaries and semicoherent interphase interfaces [4]. There are also examples of coincidence site crystallography in interphase
boundaries. The inverse Nishiyama-Wassermann orientation can be thought of as a two
dimensional $\Sigma=3$ coincidence system [5]

Systematic atomistic modelling by molecular statics and molecular dynamics using
computer algorithms to generate relaxed structures gave the first explicit insights into the

Fig 1. The four kinds of coordination that have been found so far by high resolution
microscopy of $\langle 110 \rangle$ tilt grain boundaries in gold. The large and small disks rep-
resent atoms in the separate $\{220\}$ planes of fcc stacking; (a) is an isosceles trigonal
prism as found in the $\Sigma=19$, 26.5° (331) boundary (b) is an equilateral trigonal
prism as found in the $\Sigma=11$, 50.5° (113) boundary (c) is the crystal coordination
found between isosceles trigonal prisms for $\theta \leq 26.5°$ and (d) is the twin config-
uration.

Fig 2. A diagram illustrating the core structure of a 26° $[110]$ tilt boundary in gold. Note
the two kinds of isosceles capped trigonal prism and the step with an associated
terminating (331) plane (in association with W. Krakow).

Fig 3. A dark field micrograph using the common $g=1\bar{1}1$ showing $\alpha$ fringes on ($\bar{1}12$)
oriented twins in gold (in association with M. F. Chisholm).
nature of the order at periodic tilt and twist boundaries in fcc and bcc metals [6-8]. In models for aluminum the importance of random close packing was soon appreciated. Certain of the Bernal groups were recognized as key elements of boundary structures [9] where the strict requirements for space filling and translational symmetry, which apply in crystals are relaxed but nevertheless high coordination numbers are favored by metallic bonding. Modelling with potentials designed for other metals suggested the creation of different groupings; for example in bcc metals the coordination shell consists of neighbors at $\frac{1}{2}a\sqrt{3}$ and a (where $a$ is the lattice parameter) [8]. The computer modelling was further extended to give an explicit and atomistic meaning to the structural unit concept posited by Bishop and Chalmers [10]. There was some early limited validation of the results obtained by atomistic modelling through the application of the $a$-fringe technique to asymmetrical twin boundaries [11]. This work established that the rigid lattice translations suggested by computer modelling were a real feature of the interface crystallography but did not reveal local atomistic features of the structure. The most satisfying validation of the conclusions derived from modelling came from direct imaging of particular boundaries for which the predictions from calculations could be directly compared with observations.

Observations

A range of symmetrical $<110>$ tilt boundaries in gold has been characterized by Ichinose and Ishida [13, 14] and Krakow, Wetzel and Smith [15, 16]. The rotation angles cover the range $0^\circ$ to $129^\circ$. Taken together these observations establish the existence of ordered structures in $<110>$ tilt boundaries over a wide range of angles. Figure 1 illustrates some of the structural units which have been observed in $<110>$ grain boundaries in gold. Isosceles capped trigonal prisms are found in the $10^\circ$ and $26^\circ$ boundaries; mirrored pairs of octahedra occur in the $70^\circ$ boundary ({$11\bar{1}$} twin); equilateral trigonal prisms occur in the $109^\circ$ and $129^\circ$ boundaries. Conventionally stacked octahedra occur between the isosceles trigonal prisms in the $10^\circ$ boundary. The $10^\circ$ boundary is intermediate in misorientation between the perfect crystal and the $\Sigma=19, 26.5^\circ / [110] (331)$ boundary. The observed structure of the $10^\circ$ boundary consists of distorted but topologically good crystal and isosceles capped trigonal prisms characteristic of the $\Sigma=19 (331)$ boundary which appears to be favored in the Sutton and Vitek sense [17]. This structure represents a clear example of the usefulness of the structural unit principle.

The complexity of grain boundary structure at the atomic level was first exemplified experimentally by the discovery of rigid lattice translations away from the coincidence site lattice position [11] Such translations decrease the symmetry of the bilattice and consequently a set of equivalent translations exists each of which defines a symmetry equivalent variant of the structure [18] Multiple structures have been observed in {$11\bar{2}$} twins in aluminum [12] silicon [19, 20] and GaAs [21] using the $a$-fringe technique.
A separate class of alternate structures corresponds to the occurrence of inequivalent minima in the function representing the variation of grain boundary energy with relative lattice translation or boundary plane topography. The two structures calculated and observed for the $\Sigma=19 / [110] 26.5^\circ / (331)$ symmetrical tilt boundary are an example of the occurrence of inequivalent stable structures which differ in translation state. The difference in translation state is $\sim \frac{1}{4} [110]$. The calculated internal energies of the two structures differ by about 4% [15].

Figure 2 is a plot of the atom positions derived from a high resolution transmission electron micrograph of a $\Sigma=19, 26.5^\circ / [110] (331)$ tilt grain boundary in gold. This section of boundary illustrates several key points. The core structure for most of the field of view consists of isosceles capped trigonal prisms arranged so that the boundary has mirror symmetry, the $\alpha$ structure. The capped trigonal prisms are the cores of $90^\circ, \frac{1}{2} [110]$ Lomer type primary grain boundary dislocations. There is one unit of an alternate configuration, the $\beta$ structure. The $\beta$ structure no longer has the mirror symmetry characteristic of the $\alpha$ structure. The core structure consists of isosceles trigonal prisms which are asymmetrically arranged relative to the boundary plane. The $\alpha$ and $\beta$ structures are separated by a dislocation with the approximate Burgers vector $\frac{1}{4} [110]$, i.e. the translation states differ by this vector. The electron beam is parallel to this direction and the presence of the dislocation is inferred from the change of coordination. A single terminating $(331)$ plane at $P$ indicates the location of a $\frac{1}{19} [331]$ DSC dislocation. There is a grain boundary step at the core of this DSC dislocation and the height is two $(331)$ interplanar spacings. There is also a perturbation to the spacing of the primary dislocations which illustrates directly the association of secondary dislocations with irregularities in the spacing of primary dislocations.

Even the $10^\circ$ boundary which has low angle character in the sense that it consists of perfect lattice dislocations together with the kind of imperfect dislocations which are required at the edges of $(111)$ stacking faults exhibits a not entirely anticipated richness. The Burgers vector density is imposed by the angle of rotation and the boundary plane and can be deduced from Frank's formula [22] and the Read-Shockley [23] model and its developments. However, the actual distribution of this Burger vector is a quite separate issue. Asymmetrical $[110]$ tilt boundaries in gold do not consist of an appropriate combination of the dislocations in $(h\bar{h}i)$ and $(i\bar{i},2\bar{h}i)$ symmetrical tilt boundaries. A remarkable result is that the symmetric $(110)$ boundary appears to have two possible structures. The first is the ex-
pected wall of 90° $\frac{1}{2} [\overline{1}1\overline{0}]$ edge dislocations. The alternate structure amounts to a
dissociation of the $\frac{1}{2} [\overline{1}1\overline{0}]$ dislocations into stair-rods as follows:

$$\frac{1}{2} [\overline{1}1\overline{0}] \rightarrow \frac{1}{6} [1\overline{1}0] + \frac{1}{3} [1\overline{1}0]$$

A zig-zag configuration of stacking faults on $(\overline{1}1\overline{1})$ and $(1\overline{1}1)$ is formed with a $\frac{1}{6} [1\overline{1}0]$ and
$\frac{1}{3} [1\overline{1}0]$ stair-rod dislocations at each junction. The dislocation configurations observed in
a 10° [110] tilt grain boundary as the azimuthal orientation of the boundary plane varied
are reported in [24]. From a geometrical standpoint all [110] tilt grain boundaries in an fcc
material can be generated from $\frac{1}{2} <110>$ dislocations. It appears that the low value of
stacking fault energy in gold is a significant factor and the total energy of a dislocation array
which contains redundant Burgers vector can be reduced by dissociation below that of the
most efficient dislocation configuration.

The core width of grain boundary dislocations has been a controversial issue for some
time [25, 26] but the arguments have been based on indirect evidence some of which is de-

erived from the width of the image of a dislocation under a variety of two beam conditions.

High resolution images of dislocations permit the core width of a dislocation to be estab-
lished directly. One significance of the core width is that it gives an indication of the shape
of the potential energy minimum in which the atoms at a grain boundary reside. High re-
solution transmission electron microscopy of twins has confirmed that the translation, $T$ is
zero for the coherent $\{111\}$ orientation and shown that $T$ has a component $\frac{1}{9} <111>$ for
extended $\{211\}$ oriented facets. The difference in translation state requires a partial grain
boundary dislocation in the region of the junction between $\{111\}$ and $\{211\}$ facets. Such
dislocations have been observed [27] and appear to have wide cores which are extended
about 30Å in the glide plane. This observation suggests a relatively broad and shallow
minimum in the $\gamma$-surface for the [211] twin, i.e. the interface energy is not very sensitive
to translation parallel to the boundary. Additional corroboration for this interpretation is
drawn from the observation that $T=0$ for small segments of [211] twin such as those found
at the ends of growth twins in thin films [27]. The translation $T$ on $\{111\}$ twins is zero; on
the perpendicular [211] twin the translation is zero when the [211] facet is small and $\sim \frac{1}{9}$
$<111>$ when the facet is large. In this case a $\frac{1}{9} <111>$ dislocation is required at the
junction between $\{111\}$ and $\{211\}$ A $\{211\}$ step of height $h$ between two parallel segments
of \{111\} orientation is bounded by a dipole. In equilibrium the elastic energy of the dipole must be less than the energy \( h \Delta \gamma \) gained by changing the translation from zero to \( \frac{1}{9} <111> \). If a step of height \( h \) is observed with \( T=0 \) it can be deduced that for that \{211\} twin:

\[
h \Delta \gamma \leq G \frac{b^2}{4} \ln \left( \frac{h}{2b} \right);
\]

for the case of gold \( \Delta \gamma \leq 10 \text{mJm}^{-2} \).

High resolution microscopy with the 2.3\(\text{Å} \) point resolution characteristic of 200kV instruments is for most structures limited to resolving the structure of only one projection. The discussion above concerning the translation state of \{211\} twins and the invisibility of the intervariant dislocation in the 26.5° tilt boundary emphasize this limitation. The \( \alpha \)-fringe technique [11] is potentially able to give the complementary information needed to define fully the translation state. Figure 3 is an example of the application of the \( \alpha \)-fringe technique to detecting the translation on \{211\} facets of growth twins in gold. There are four sets of annealing twins corresponding to the four different \{111\} planes. Figure 3 is a (110) oriented single crystal film which has been tilted approximately 30° about \{111\}. For the arrowed twins in fig. 3 the coherent (111) plane is edge on and the perpendicular (112) plane is inclined to the electron beam. The \( \alpha \)-fringes produced with the common \( g \overline{1}1 \) excited (symmetrical profile in bright field, antisymmetrical in dark field) are evidence that \( g \cdot T \) (modulo one) \( \neq 0 \). From the high resolution transmission electron microscopy \( T \) includes a component \( \frac{1}{9} \{111\} \) so that with \( g=\overline{1}1 \), \( g \cdot T= \frac{1}{3} \) which is consistent with the relatively strong contrast of the \( \alpha \)-fringes.

![Fig 4](image-url) Field ion micrographs showing (a) the narrow substrate-deposit interface similar in appearance to a high angle grain boundary and (b) a wide intercolumnar boundary in sputtered tungsten.
It is worth mentioning the contribution of field-ion microscopy to the understanding of the atomic structure of grain boundaries in metals. Field-ion microscopy supports the view that the cores of grain boundaries [28, 29] and grain boundary dislocations [30] are narrow in general. These observations relate to grain boundaries which are not all accessible to high resolution transmission electron microscopy. The field-ion microscope also gives additional insight into the inter-columnar boundaries which are characteristic of metallic films sputtered onto substrates maintained at low homologous temperatures. Fig. 4 illustrates the similarity between (a) the substrate-deposit interface and a conventionally formed grain boundary and (b) the difference from an inter-columnar boundary for the case of tungsten sputtered at room temperature onto a tungsten FIM specimen substrate.

This paper has been exclusively concerned with the structure of static grain boundaries. It may be anticipated that future work will address the issues of the dynamics of grain boundaries together with the sites occupied and structural changes resulting from segregation [31].

Conclusions

High resolution microscopy has verified the prediction of computer modelling that grain boundaries in fcc and bcc materials are ordered. The interrelationship of the primary and secondary dislocation models with each other and with the structural unit concept is experimentally validated. There is some evidence for the coexistence of multiple structures.

References