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ABSTRACT

High-resolution electron microscopy (HREM) in combination with computer simulations of the fully relaxed atomic structures and energies of symmetric and asymmetric grain boundaries (GBs) in Au $<1\bar{1}0>$ tilt bicrystals has been applied to a study of atomic relaxation modes. These investigations indicate that: (i) Atomic relaxations are typically dominated by short-range interactions and the tendency of the solid to assume a local atomic environment similar to the bulk. (ii) Misfit localizations are likely within a structural unit whenever the GB unit cell is large. (iii) When there is a length mismatch along the GB the atomic relaxations can assume quasiperiodic character, generating densely spaced regions of structural disorder, akin to misfit dislocations. (iv) Atomic relaxations can take the form of stacking disorder.

1. INTRODUCTION

The study of atomic-scale features of grain boundaries (GBs) has within the past decade become a practical reality due to (i) the development of computational techniques for atomistic GB simulations and (ii) the experimental observation of GBs by modern HREM instruments. The combination of these two techniques provides a powerful tool for investigating GBs and developing an understanding of the connection between properties of GBs and their atomic-scale structure [1].

In the past, many of the views about the atomic-scale structure of GBs were based on rigid atomic models, which are typically constructed for periodic bicrystals, such as the coincident-site lattice (CSL) [2]. The GB periodicities, and size of structural units, etc. can within this framework be derived from simple geometrical arguments. Quasiperiodic GB structures have been discussed, based on GBs formed on irrational planes or in aperiodic bicrystals [3, 4]. However, as will be shown below, quasiperiodic structural units can also form as a consequence of the short-range nature of atomic relaxations within periodic GB unit cells.

Atomic relaxations, including rigid-body translations normal and parallel to the GB plane, have long been recognized to play an essential role in determining the detailed atomic structure of GBs, as well as the GB energy and many other GB properties. In addition, possible GB reconstructions may be of importance in the formation of GBs.

In the present work we examine several important features of possible atomic relaxation modes in fcc metals. Since the majority of high-angle GBs of interest do not have very small planar unit cells we have chosen the tilt boundaries about $<1\bar{1}0>$ with a misorientation angle $\psi=55°$ as a model system for studying atomic relaxations that occur within the relatively large GB planar unit cells in this geometry.

2. HIGH-RESOLUTION ELECTRON MICROSCOPY OF TILT GRAIN BOUNDARIES

Thin (110) gold films were grown epitaxially on NaCl in a UHV system and then pairwise sintered together at the appropriate misorientation angle. Columnar grains with the desired tilt GBs were obtained by further processing, as described previously [5]. High-resolution images were taken under axial illumination for several defocus values near optimum defocus, utilizing a H9000 high-resolution electron microscope, operated at 300 kV.

3. GRAIN BOUNDARY GEOMETRY

In the present experiments the symmetric (338)(338) and (443)(443) GBs as well as the asymmetric (111)(001) GB were examined. These GBs are formed in bicrystals near $\Sigma = 41$ (where Σ is the reciprocal coincident-site-lattice density). The exact CSL misorientation about the $<1\bar{1}0>$ axis is $\psi = 55.88°$. Within about 1° of this value, at ψ=54.74° (001) planes within one crystal are parallel to a set of (111) planes in the other crystal and an (incommensurate) (111)(001) GB can be formed. The corresponding periodic GB at Σ=41 is (24,24,23)(001) which has a rather large planar unit cell of length 11.8 nm. Figure 1 shows a TEM image of (111)(001) and (338)(338) facets with their underlying structural periodicities indicated by the closely-spaced strain contrast features that can be observed in fig. 1.

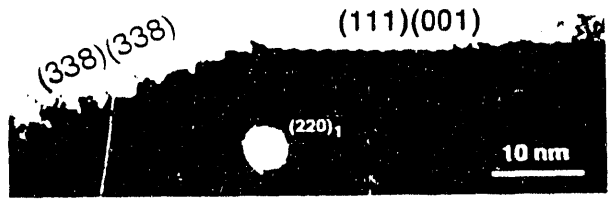

Fig. 1. TEM image shows strain contrast features due to structural periodicities in (111)(001) and (338)(338) GBs.

4. GRAIN BOUNDARY SIMULATIONS

A zero-temperature iterative energy-minimization algorithm ("lattice statics") was used to compute the fully relaxed atomic structures of GBs. To enable the GB to expand or contract, the unit-cell volume was allowed to increase or decrease in response to the internal pressure [6]. Also, by computing the forces which the two halves of the bicrystal exert on each-other, translations parallel to the GB plane were allowed while the atoms relax. By starting from a variety of initial rigid-body translational configurations, the GB energy is thus minimized with respect to both the atomic positions and the three components of the rigid-body translation of the GB.

A many-body potential of the embedded-atom-method (EAM) type, fitted for Au [7], was employed. A comparison of these simulations to results obtained with a much simpler Lennard-Jones potential suggests that qualitatively, the main structural features observed are typical of fcc metals.

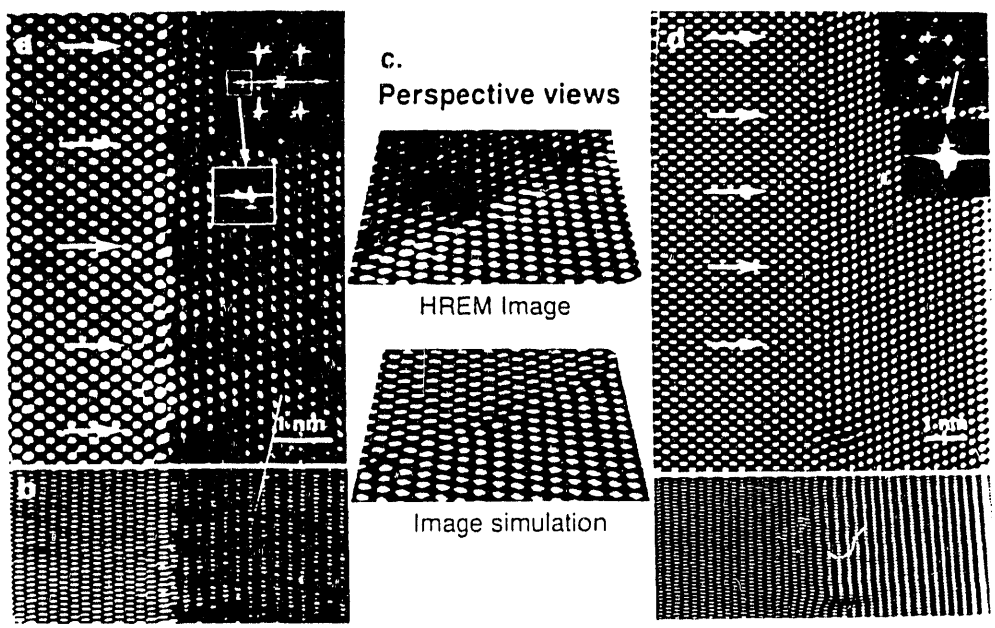

Fig. 2. (a) HREM image of (111)(001) GB. (b) Same as (a), but with vertical image compression. (c) Perspective view along $<110>_1$,$<112>_2$ directions for experimental and simulated images. (d) Simulated image of (24,24,23)(001) GB unit cell, based on fully relaxed GB model, calculated using embedded atom potential.

However differences may well occur from one material to another, for example, in features which are sensitive to the stacking fault energy, as discussed below.

5. THE (111)(001) TILT GRAIN BOUNDARY

The observation of extended facets of the (111)(001) GB is in agreement with previous work that suggested that the combination of relatively dense-packed planes results in the formation of low-energy boundaries [5, 8-10]. Figure 2a shows an HREM image of a section of the (111)(001) GB. The compressed image in fig. 2b clearly indicates the subtle image variations which are due to misfit localizations along this GB. These atomic relaxations are misfit-dislocation-like, as can be seen in fig.2c , which shows a perspective view of fig. 2a, emphasizing the arrangement of planes crossing the interface. For a rigid model the misfit between atomic planes along the <110> and <112> directions is 14% and corresponds to a period of 1.86 nm, which is close to the observed value of 1.8 nm. The spacing of these relaxations is the same as for the strain-contrast features in fig.1, and is also identical to the periodicities implied by relrod features in reciprocal space from the diffraction pattern as well as the corresponding features in the Fourier transform of the lattice image. To answer the question whether relaxations that are caused by the incommensurability of the (111) and (001) planes also occur in a perfectly periodic GB, the (24,24,23)(001) boundary was simulated described above. Figure 2d shows one structural repeat unit of the (24,24,23)(001) GB as the result of a calculation of atomic relaxations, followed by a multislice calculation of the GB HREM image. The quasiperiodic structural units within the periodic GB supercell can be clearly recognized and the signature of these quasiperiodic relaxations is also visible in the Fourier transform of this image (see inset).

6. THE (338)(338) TILT GRAIN BOUNDARY

This GB, as seen in figure 3, has an interesting structure which is characterized by the extension of localized regions of stacking disorder at an angle of 82° relative to the GB plane. This boundary can be considered vicinal to the (113)(113), $\Sigma = 11$, GB ($\psi = 50.5°$), which is associated with a deep cusp in energy [8] and has an atomically well-matched structure, characterized by a coherent continuation of (111) planes across the interface. The stacking faults apparently are formed to accommodate the additional misorientation of approximately 5°. The partial dislocations terminating the stacking faults and running parallel to the misorientation axis give rise to extended lattice strain, which can be recognized in fig. 1. The lowest energy structures obtained by computer simulation of this GB also show the stacking fault nature of the experimental structure (see fig. 3b). However, at higher energy a reconstruction of this boundary into microfacets of the (111)(001) type has been observed in simulations based on the embedded atom as well as the Lennard-Jones potential (fig.3c). This suggests that this mode of GB relaxation may be preferred in materials of high stacking-fault energy.

7. THE (443)(443) GRAIN BOUNDARY

Within the GB unit cell of this boundary one can clearly recognize in fig.4 regions of misfit

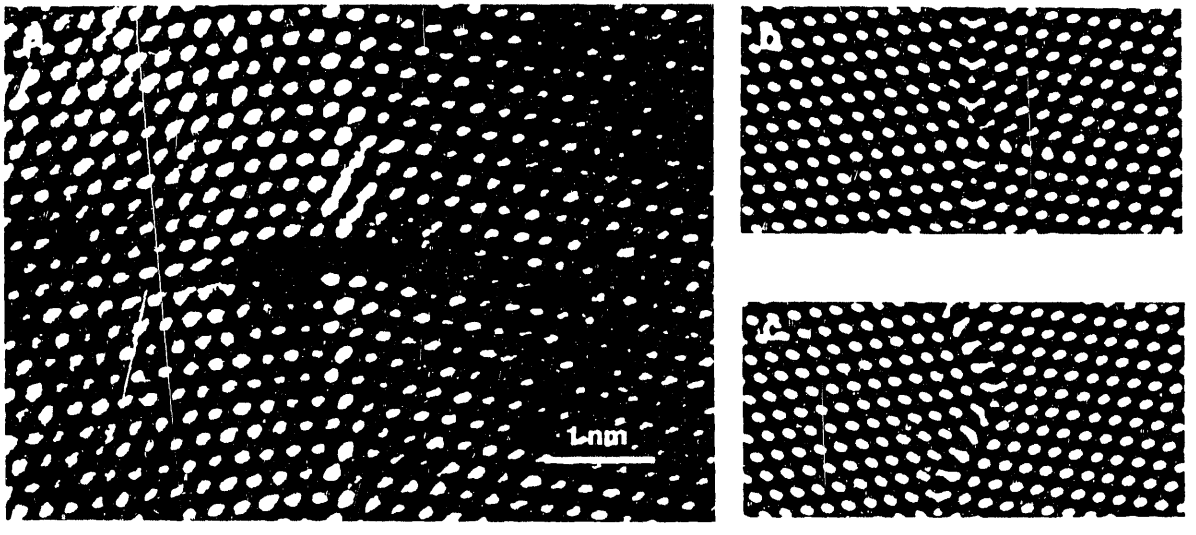

Fig. 3. (a) HREM image of (338)(338) GB with stacking fault extending along (111) planes crossimg the interface. (b) stacking disorder in simulated GB. E = 327 mJ/m^2. (c) Microfaceting in calculated GB. E = 392 mJ/m^2.

localization. This boundary also displays stacking-fault type relaxations. However the translations between the (111) planes can be clearly recognized on only one side of the GB, in contrast to the calculated structure, which has a more symmetric distribution of the stacking disorder. Therefore, although the calculated lowest-energy core-structure of this GB roughly agrees with the HREM observations, small deviations do exist between calculated and observed structures.

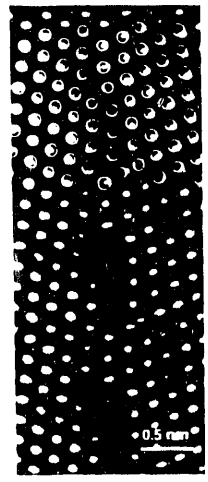

Fig. 4. Digitally averaged HREM image of (443)(443) GB. Calculated GB model is superimposed near top.

8. DISCUSSION AND SUMMARY

The atomic relaxations observed here and elsewhere [10-12] in high-angle GBs can be discussed in view of the concept of minimizing the overall atomic disorder per GB unit area, and therefore its energy. This is reflected in the tendency of the solid to maintain, as much as possible, the local atomic coordination present in the bulk.

In GBs with a large geometric repeat unit, one consequence of this tendency to maintain ideal coordination is *misfit localization*. In this manner regions of well coordinated material alternate with regions of misfit [13]. The connection between GB energy and the atomic coordination per GB unit area has recently been investigated via a broken-bond model for a wide range of GB geometries [14]. In an extension of the present work we have used this model to localize regions of misfit within the GB unit cell.

Another consequence of the tendency toward atomically well-coordinated regions is the formation of stacking faults, which results in unusual high-angle GB structures, which include relatively far-reaching strain fields, associated with line defects at the termination of the stacking faults.

Finally, we have shown that quasiperiodic relaxations are present in the (111)(001) GB. The quasiperiodic nature of such misfit localizations is to be expected for an incommensurate interface. The associated strain fields of such misfit-like dislocations have recently been analyzed by Bonnet and co-workers, who have named them "Somigliani" dislocations [15]. The physical reason for such relaxations must be sought in the short-range nature of the interatomic interactions. Therefore such relaxations occur even in a perfectly periodic GB where they appear as quasiperiodic subunits within the GB unit cell, as has been shown here for the (24,24,23)(001) GB. In real crystals the GB translational symmetry on the atomic scale will be broken by this atomic relaxation mode.

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