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ABSTRACT

The structural characteristics of [111] twist boundaries in gold are investigated using a combination of x-ray diffraction and computer modeling techniques. Comparison of the measured scattering effects with those generated from EAM computer models reveals that the [111] boundary displacement field is weak, rotational in form and centered on 'O' lattice sites. Furthermore, the measured intensities of the strong 'O' lattice reflections decrease smoothly with increasing boundary angle up to 30° , as calculated from the model. The effect of double positioning on the diffraction pattern and the structural analysis is discussed.

INTRODUCTION

In a recent paper [1], the theoretical diffraction effects from a series of [111] twist boundaries in gold were calculated using an embedded atom model (EAM) to simulate the boundary displacements. In that study, the objective was to predict the strength and distribution of the kinematical scattering from individual [111] boundaries and also determine any additional effects introduced by a phenomenon known as double positioning. Double positioning is the coexistence, in the same crystal, of two distinct types of domains which are related by a simple twinning operation and differ in misorientation by 60° . Domains of doubly positioned (DP) boundaries are always found in (111) gold bicrystals prepared from epitaxially grown thin-films due to the presence of faults in the (111) stacking sequence and the formation of low-energy twin boundaries. It is clear that the presence of two distinct domain structures in a (111) bicrystal would complicate an x-ray diffraction study aimed at determining the atomic structure of the boundary. This is because, in general, each domain would be expected to diffract differently and the measurements would represent average intensities from both boundary types. In the calculations, however, it was found that a window of misorientation exists around 30° in which pairs of DP boundaries diffract with the same intensity and on the same reciprocal lattice section. Under these conditions, therefore, x-ray measurements could be made of the superimposed intensities and information derived concerning the structure (in projection) of each DP boundary. Outside this misorientation window, however, structure determination was predicted to be less reliable since the superimposed intensities were unequal and could not be separated.

In the present paper, the calculated scattering from [111] twist boundaries is compared to some quantitative x-ray measurements on the same boundaries in gold. Effects due to double positioning are discussed and, because the amount of available data is limited, emphasis is placed on reproducing trends and obtaining consistency rather than attempting detailed structure determinations.

COMPUTATIONAL

In this section, the principal results obtained from the computer simulations are summarized. Using molecular statics and an EAM potential for gold [2] the equilibrium atomic structures of 12 low- Σ [111] twist boundaries were determined. The details of the computational model and method of relaxation have been described previously [3] in an application to [001] twist boundaries. The boundaries studied spanned the angular range $9^\circ < \theta \leq 60^\circ$ and had Σ values equal to 3, 7, 19, 21, 31, 37, 39, 43, 57, 73 and 111. Several of these boundaries are DP pairs since their Σ 's are related by a factor of 3, e.g., $\Sigma 19$ and $\Sigma 57$, as discussed in [1]. After obtaining the relaxed coordinates of each boundary, the geometrical structure factor, $F(H,K,L)$, was computed on the appropriate boundary diffraction lattice which is the reciprocal of the CSL [4]. The value of $|F|^2/\Gamma^2$ was then calculated, where Γ is the planar density of coincidence sites, since this is a measure of the scattering power per unit area of the boundary.

The computed results indicate the following:

- (a) in the $L=0$ plane and when $20^\circ < \theta < 40^\circ$, the scattering from two separate DP boundaries is almost identical. However, when $L \neq 0$, or when $20^\circ > \theta > 40^\circ$, the scattering from the two boundaries is significantly different especially as θ approaches $\Sigma 1$ and $\Sigma 3$. It is concluded from this that, since the experimental scattering will contain superimposed information from both DP boundaries, only the projected atomic structure of [111] twist boundaries in the vicinity of 30° may be determined reliably.
- (b) the scattering power per unit area of symmetrically located 'O' lattice reflections varies smoothly with θ and passes through a minimum at 30° (see Fig. 1). This behavior is analogous to that exhibited by 'O' lattice reflections in [001] twist boundaries [3] and is directly related to the strength of the displacement field which is approximately rotational and centered on 'O' lattice sites [1].
- (c) the absolute magnitudes of the scattering power per unit area are calculated to be at least an order of magnitude smaller than the corresponding values found for [001] twist boundaries [3].
- (d) provided the coherence length of the x-rays is much smaller than the DP boundary domain size, interference effects from neighboring domains can be ignored. This is the case in the present work.

EXPERIMENTAL

I. Specimens: The required thin film bicrystals were prepared by welding two (111) single crystal gold films together using a variation of the technique first developed by Schober and Balluffi [5]. Specifically, $800\text{\AA}$ gold films were deposited epitaxially on about $3000\text{\AA}$ Ag films at a temperature of approximately 100°C (low enough to prevent Ag/Au interdiffusion) in a vacuum bell jar pumped down to $\sim 10^{-8}$ torr. The Ag film had previously been deposited on carefully cleaved mica substrates that had been annealed for a few hours at

~270°C at a base pressure of 10^{-7} torr. Films grown by this technique were found to contain extensive double positioning as determined by TEM and x-ray techniques. This result is consistent with earlier studies of (111) gold bicrystals [6,7]. The degree of double positioning was found in most cases to be distributed between 40-60% of the grains oriented parallel to one set of grains or the other. The bicrystal was manufactured by picking up one single crystal of gold floating on water (using HNO_3 to separate it from the Ag/mica substrate), by another Au film while still on its Ag/mica substrate. Details of the procedure will be given in a future publication.

The resulting bicrystal was generally free of water/air bubbles so that it could then be annealed at approximately 150-200°C without the film being ruined due to an occasional remaining bubble of water or air. Fairly extensive regions of good grain boundary could be obtained by this technique.

The gold bicrystals manufactured in this way were floated on to the surface of water and picked up onto stretched kapton films mounted on aluminum rings [3], in order to perform the x-ray scattering experiments.

II. X-ray Scattering Experiments: X-ray scattering experiments were performed on the above manufactured samples mounted on Huber 4-circle diffractometers, attached to both rotating anode and Synchrotron sources; due to the very weak nature of the scattering only a low-angle boundary (~8° twist angle) could be studied on the rotating anode; all other experiments were performed at the NSLS facility at Brookhaven National Laboratory using MIT/IBM beamlines X20A and X20C.

Briefly, a Ge (111) double crystal monochromator was used to select x-ray photons with an energy of 8keV and focussed using a Pt coated Si mirror, to a spot $\sim 1.0\text{mm}^2$, at the center of a Huber 4-circle diffractometer. The resulting resolution in reciprocal space around the 220 gold reflection was, $\Delta q_{11} \sim 0.001\text{\AA}^{-1}$, $\Delta q_{\perp} \sim 0.001\text{\AA}^{-1}$, $\Delta q_x \sim 0.03\text{\AA}^{-1}$, where Δq_{11} , $\Delta q_{\perp}$, Δq_x are defined in [8]. The absolute structure factor squared was measured using the technique described in [3], with changes appropriate for the synchrotron source. Due to the exponentially decaying nature of synchrotron radiation in a single run, a monitor was inserted so as to pick up diffuse scattering off a piece of Kapton placed in the path of the incident beam; all scans were then made with respect to a specified number of monitor counts (i.e., count/monitor counts) instead of scanning in real time (i.e., counts/sec). Since the measured reflections are very weak, significant signal/noise problems arise and fairly large counting times must be used. The background is estimated by fitting various polynomials to points away from the peak. The presence of double positioning in the bicrystal films significantly increases the likelihood of unwanted multiple scattering effects (double diffraction). As pointed out in previous work [9], these effects complicate the measurement of a large number of reflections and make structure determination more difficult. A thorough analysis has shown that the measurements presented below are not influenced by double diffraction.

III. Transmission Electron Microscopy: Following the x-ray experiment, the samples were floated off on water, sliced into several pieces, and analyzed

using transmission electron microscopy to determine what fraction of the boundary was welded, in the manner described in [3]. The measured x-ray intensities were adjusted to take account of this fraction which was determined to within an estimated uncertainty of 20%. In the angular range of boundaries studied, no intrinsic grain boundary dislocations were observed. In addition, no difference in general contrast effects was observed between the two DP boundary domains. It was thus not possible to determine the relative fraction of each DP boundary present and therefore the measured x-ray intensities represented an average effect from both types of boundary. In the present study, which focusses on structural characteristics, these average intensity measurements are sufficient. It is noted, however, that they could be used as part of a detailed structure determination provided the boundaries being investigated fall in the required angular range as discussed earlier.

DISCUSSION

Figure 1 compares the calculations and measurements for two families of symmetrically located '0' lattice reflections as a function of boundary misorientation θ . The two types of reflections, labelled A and B, are located immediately adjacent to the 220 region of reciprocal space as illustrated in Fig. 2 for the case of the $\Sigma 7$ boundary. They are typically the strongest grain boundary reflections closest to the origin. Since the measurements are averaged intensities from both DP boundaries (at θ and $60-\theta$), they are plotted symmetrically about 30° . The calculations, however, indicate that a slight asymmetry exists which becomes more pronounced as θ approaches 0° and 60° as discussed previously. (This is probably due to the different dislocation structures present in the simulated boundaries at these two angular extremes.) Despite this difference it is seen that the match between the calculations and measurements is quite good (to within the indicated experimental error) and that therefore several of the computational predictions are confirmed, namely: (i) the smooth variation of intensity with θ and the presence of a minimum at 30° , and (ii) the scale of the intensities which are considerably smaller than those found previously for [001] twist boundaries (for comparison, it is noted that the intensity level of the strongest measured $\Sigma 5(001)$ reflection in gold corresponds to a value of about 25 for $10^2 |F|^2 / r^2$). From these results, it may be concluded that the calculated displacement fields and structure factors are at least consistent with those being measured. Of course, much more data is required to perform a structure determination and this data will be presented in a forthcoming publication.

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FIG. 1. Measured and calculated (EAM) values of the scattering power per unit area of boundary ($|F|^2/\Gamma^2$) plotted as a function of θ . Values are for two types of reflection labelled A and B whose location near 220 is illustrated in Fig. 2. Experimental errors from all sources are estimated to be about 35%.

FIG.2. Boundary diffraction lattice projected onto the $L-O$ plane for a $\Sigma 7[111]$ twist boundary. Location of the A and B reflections near 220 is shown.

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