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Modeling of dislocation patterns of small- and high-angle grain boundaries in aluminum

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ABSTRACT

A molecular dynamics simulation approach to the investigation of grain boundary structures is presented. By the example of aluminum crystallization from the melt we demonstrate the formation of polycrystalline structures from coexisting nucleation seeds. The latter are used to induce specific crystallographic orientations and hence determine the tilting of the grains resulting from further crystal growth. This allows the systematic investigation of the evolution of grain boundary structures as a function of the tilt angle. On this basis, the transition from small- to high-angle grain boundaries in aluminum as obtained from rapid under-cooling of the melt is rationalized by arrays of two different sets of dislocation pairs.

1. Introduction

The study of polycrystallinity and the structure of grain boundaries represents a fundamental contribution to the understanding of materials properties at the atomic scale. In parallel to electron and atomic force microscopy, molecular simulation techniques promoted much of the in-depth knowledge available today [1,2]. Using quantum mechanics or suitable empirical potentials [3] it is in principle possible to study grain boundaries in atomistic models comprising of thousands to millions of atoms [4,5]. However, the main limitation of such models is given by the need of presuming much of the atomistic structure. While artificially prepared cuttings of differently oriented grains may be improved by optimization in favor of (local) energy minima, the formation of complex dislocation patterns and metastable configurations arising from kinetic trapping typically remain elusive. This issue is particularly problematic for small-angle grain boundaries which do not exhibit strong favoring of well-defined tilting planes related to specific coincidence angles which may be derived from geometric considerations [1].

In this contribution, we present an unprejudiced approach to the modeling of grain boundary structures. Therein, only the titling angle is to be fixed, while the atomic arrangement of the corresponding bicrystal model results from atomic self-organization during crystal formation from the undercooled melt. This approach paves a general way to the investigation of grain boundary structures as a function of the tilting angle which shall be demonstrated by the example of small- and high-angle ⟨100⟩ tilt grain boundary structures in aluminum.

2. Methods

While atomistic simulations of crystal nucleation from the melt reflect an intuitive way to the unbiased assessment of (poly)crystalline structures, the limited time- and length scales accessible to today's computing performances prevent straight-forward molecular dynamics simulations of such processes. To allow such simulations, here we promote crystallization (i) by rapid cooling far below the melting point and (ii) by introducing nucleation seeds. The latter are implemented as layers of solid aluminum embedded in the melt. Within these layers (highlighted in yellow in Fig. 1)[1] melting is avoided by restraining all atoms to follow the center-of-mass motion of the whole layer, only. In-between, a nano-

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[1] For interpretation of color in Fig. 1, the reader is referred to the web version of this article.

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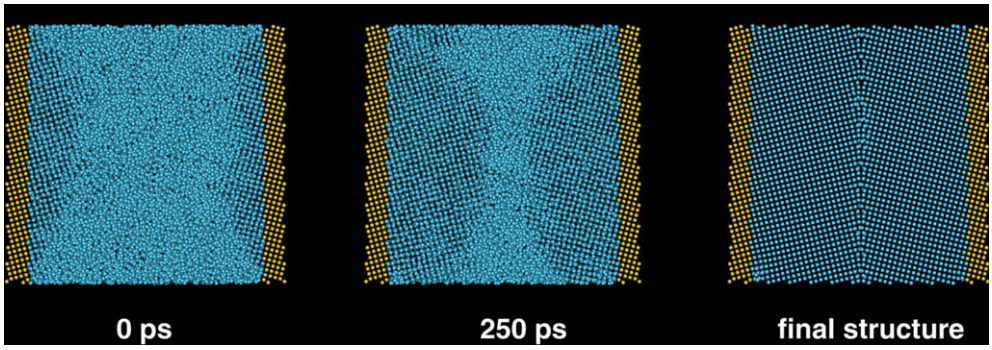

Fig. 1. Heterogeneous crystallization of aluminum from the melt (blue) promoted by fixed Al layers (yellow, here: tilted by 30°). Left: initially independent nuclei adopt the crystallographic orientation of the substrate layers acting as crystallization seeds. Upon further crystal growth (middle), the two nuclei merge into a bicrystal structure. Right: Final configuration after quenching to 0 K. 2D periodic boundary conditions are applied normal to the horizontal axis. In the layers embedding the melt the atoms are restraint to follow the layers center-of-mass motion, only. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

sized model of an aluminum melt is explored from unconstraint molecular dynamics simulations. Along this line, a thermostat is used to gradually reduce the simulation temperature from 1000 to 0 K. Cooling rates from 250 to 50 K ns^{-1} were explored, leading to qualitatively consistent bicrystal structures as final configurations (higher cooling rates give rise to serious artifacts, including partially glassy structures). The results discussed in the following were obtained from cooling by 50 K ns^{-1} (for which crystal nucleation is typically observed at around 500 K). In analogy to liquid phase epitaxy, the two nuclei adopt the crystallographic orientation of the substrate layers. Further crystal growth leads to the contact of the two nuclei and the self-organization of a bicrystalline structure. The tilt angle of the grain boundary is controlled by the different lattice orientations in the two substrate layers. For a given angle the atomistic structure of the grain boundary results from atomic self-organization during crystal growth and is hence free of prejudicing.

The atomic interactions are modeled by the EAM potentials [3] of Sutton and Chen [6]. This approach was recently demonstrated to be suitable for modeling high-angle tilt boundary structures in agreement with HRTEM of aluminum bicrystals [7]. For the molecular dynamics simulations a time step of 2fs was found to be appropriate. Each of the investigated simulation systems comprises of 17,500 Al atoms which upon solidification corresponds to a $25 \times 25 \times 7$ single crystal if the tilting angle is chosen as zero. To introduce a desired tilting angle α, the atomic lattices of both layers are rotated by $\pm\alpha/2$, respectively. The approach is not confined to coincidence angles (for which – in case of certain low CSL numbers – grain boundary structures may be obtained directly from geometric modeling). This feature is particularly useful for the study of the interplay of dislocation patterns and lattice distortions in polycrystals prior to long-termed annealing of the microstructure.

3. Results

We scanned a series of $\langle 100 \rangle$ tilt grain boundary structures in pure aluminum as a function of the tilting angle ($\alpha = 0°$, 2.5°, 5°, 7.5°, 10°, 12.5°, 15°, 20°, 30° and 45°). The formation of dislocations was observed to occur only to the extend which is required geometrically to accommodate the related tilting angle – in particular the $\alpha = 0°$ run lead to a perfect single crystal illustrating that no extra dislocations arise from kinetic traps and short simulation time scales. Indeed, we suggest the bicrystal growth run with $\alpha = 0°$ as a testing procedure to help avoiding artifacts from excessive driving of the solidification process. For $\alpha > 0°$ two types of dislocation pairs were found to accommodate lattice distortions and grain interfaces: In small-angle grain boundary structures, pairs of perpendicular dislocations as illustrated in Fig. 2a are observed. With increasing tilting, pairs of parallel dislocations become of increasing importance and are fully dominant for $\alpha > 10°$ (Fig. 2b). This evolution is accompanied by the formation of $\langle 001 \rangle$ tilt planes and hence *reflects the transition from small- to high-angle grain boundary structures* as discussed in the following. From $\alpha \geqslant 20°$ the centers of the pairs of parallel dislocations merge and the dislocation patterns collapse into sharp planes acting as grain interfaces. Along this line, the diameter of the interfacial region changes from several nanometers to atomic layers constituting mirror planes as grain boundaries. The small-angle grain boundary structures were hence found to be less sharp then commonly assumed.

Our simulations help to rationalize the transition from small- to high-angle grain boundary characteristics by means of structural motifs: with increasing tilting angle the dislocation patterns evolve from pairs exhibiting perpendicular Burgers vectors $\left(\frac{a}{2}[100], \frac{a}{2}[010]\right)$ to pairs of parallel dislocations $\left(2 \times \frac{a}{2}[100]\right)$. It is noteworthy that for each pair of perpendicular dislocations observed for the small-angle grain boundary structures we always found a corresponding pair which in principle allows dislocation recombination to annihilate two dislocations while two parallel dislocations would remain. To explore putative annealing processes, the final configuration involving the 5° grain boundary was subjected to additional relaxation studies. However, from a 25 ns annealing simulation at 500 K no changes in the dislocation patterns could be observed. To promote dislocation motion, we then introduced periodic ±2.5° shearing by smoothly shifting the substrate layers in an anti-parallel manner along the [001] direction. Three cycles each involving a 10 ns run were explored. While the dislocation pairs moved by up to several nanometers during the shearing simulations, again no recombination could be observed, hinting at the metastability of the dislocation patterns illustrated in Fig. 2.

A possible explanation for the preferential formation of pairs of perpendicular dislocations during the intergrowth of bicrystals of small-tilting angles may be suggested on the basis of the local lattice distortion next to the dislocation pairs. While a pair of perpendicular dislocations is associated to the cutting of a single layer of atoms from the ideal fcc-lattice (Fig. 2a), the incorporation of a pair of parallel dislocations requires the cutting of two parallel layers (Fig. 2b). Indeed, the pairs of perpendicular dislocations give rise to smaller lattice distortion and hence appear more favorable to accommodate the intergrowth of bicrystals of very small tilting angles. Moreover, the net Burgers vector of $\frac{a}{2}[100] + \frac{a}{2}[010] = \frac{a}{2}[110]$ corresponds to the shearing direction of the most favorable Peierls