Capillary-driven grain boundary motion and grain rotation in a tricrystal: A molecular dynamics study

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Received 8 June 2013; received in revised form 18 November 2013; accepted 20 November 2013

Abstract

We report on molecular dynamics (MD) simulations of a tricrystal composed of a cylindrical grain embedded at the center of a plane grain boundary (GB). The embedded grain shrinks by capillary forces and eventually vanishes. This process is often accompanied by rotation of the embedded grain in either a clockwise or counter-clockwise direction. Using the geometric theory of coupling between GB motion and grain translations, we propose a model capable of predicting the direction of the grain rotation depending on the crystallographic parameters of the three grains. Full agreement has been found between the model predictions and the MD simulation results for both spontaneous grain shrinkage and in the presence of applied shear stresses. The consequences of these results for grain rotation in polycrystalline materials and possible extensions of the model to multiple grains are discussed.

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Keywords: Modeling; Grain boundary; Rotation; Capillary

1. Introduction

Grain rotation is part of microstructure evolution in polycrystalline materials and has been observed experimentally during plastic deformation [1,2], recrystallization [3] and grain growth [4,5]. Rotation of an isolated cylindrical grain was predicted theoretically [6–8] and observed in computer simulations by molecular dynamics (MD) [6,9–12] and the phase-field crystal (PFC) method [13]. In most cases the grain rotation could be explained by the existence of coupling between grain boundary (GB) motion and shear deformation of the lattice [6–8,14,15].

In real materials, isolated grains are extremely rare. Most grains are surrounded by several other grains and are separated from those by multiple GBs and triple junctions. One could expect that this complexity would suppress the grain rotation, or at least make it a rare occurrence, for at least two reasons. First, the triple lines can be expected to act as pinning sites since the grain rotation requires accommodation of incompatibilities along the triple lines [16–18]. Second, assuming that the magnitude and sign of the grain rotation depend on geometric parameters of the GB, different GBs would try to rotate the grain in different directions, reducing or even blocking the net rotation. Nevertheless, in certain cases the individual GBs can exert forces acting in the same direction and collectively causing the grain to rotate. The ability to predict such cases requires a better understanding of the geometric conditions for grain rotation by individual GBs and the role of triple junctions in this process.

Because of the complexity of the general polycrystalline problem, it is strategically meaningful to start with a relatively simple configuration amenable to quantitative analysis and then continue to build up the complexity by adding more grains. As such, a perfect starting point is offered by a tricrystal consisting of a cylindrical grain sitting on a plane GB. This configuration involves two different GBs surrounding the embedded grain and two different triple lines. The embedded grain spontaneously shrinks by capillary forces and may or may not rotate in the process.
This tricrystalline structure was studied by MD simulations in a two-dimensional Lennard-Jones system with a triangular lattice \[11\]. No rotation was found when the two curved GBs were symmetrically equivalent, leading to cancellation of the rotational forces. When the symmetry was broken, rotation was observed in some cases but not in others, and was apparently influenced by the presence of open surfaces in the simulated models. The important observation was, however, that the presence of two different GBs and two triple lines did not prevent the grain rotation. More recently, Wu and Voorhees \[13\] reported on two-dimensional PFC simulations of a similar tricrystalline system with hexagonal lattices. The geometry was symmetrical with the curved GBs having opposing misorientations of \(\pm 5.2^\circ\). As expected from this symmetry, no grain rotation was observed, in agreement with the MD simulations \[11\]. Although interesting insights into the dislocation mechanisms of GB migration were obtained, grain rotation under asymmetric conditions was not tested.

In this work we address the same tricrystalline configuration but in a more systematic manner. Based on the analysis of geometric coupling factors, we propose a simple analytical model that permits predictions of the direction of grain rotation, depending on the misorientation angles of the GBs. We then conduct a series of MD simulations designed to test this model. All three GBs are chosen to be \([001]\) tilt type. The atomic interactions are modeled with an accurate atomistic potential to ensure realistic character of the results. As a further test of the model, we conduct simulations under a shear stress applied parallel to the plane GB and driving the collective motion of all three boundaries.

In Sections 2 and 3 we introduce our model and propose analytical expressions for the effective coupling factor governing the grain rotation. These two sections also serve to introduce a set of sign conventions for the angles and coupling factors; the nature of the problem makes it important to adopt and strictly follow such conventions in order to avoid confusion. In Section 4 we describe the simulation methodology and details of the GB geometries tested in this work. The results of the MD simulations for the spontaneous shrinkage of the embedded grain are presented in Section 5, followed by stress driven simulations in Section 6. We conclude the paper by discussing the comparison between the proposed model and the simulation results and outlining future work (Section 7).

2. Coupling model for curved grain boundaries

To set the stage for the discussion of curved GBs, we will first review the geometric theory of coupling for plane GBs. Fig. 1 illustrates the geometry of a plane asymmetrical \([001]\) tilt GB between two face-centered cubic (fcc) crystals, defining the tilt angle \(\theta\) and the inclination angle \(\phi\). Angle \(\theta\) measures the misorientation between \([100]\) directions in the grains, with \(\theta = 0\) for a single crystal and \(\theta > 0\) when the \([100]_U\) axis in the upper grain is rotated counter-clockwise relative to \([100]_L\) in the lower grain. The inclination is characterized by the angle \(\phi\) between the GB plane and the internal bisector between the \([100]\) directions in the grains, with \(\phi > 0\) if the bisector is rotated in the counter-clockwise direction relative to the GB plane. Due to the fourfold lattice symmetry around the \([001]\) axis, all distinct GB structures can be found in the angular domain \(\{0 \leq \theta < 90^\circ, -45^\circ \leq \phi < 45^\circ\}\). Symmetrical tilt GBs arise at \(\phi = 0\) and again at \(\phi = \pm 45^\circ\), all other boundaries being asymmetrical.

The coupling factor of the boundary is defined as the velocity ratio \(\beta = v_b/v_n\) (Fig. 1). Following the sign convention of \[8,14,15\], \(\beta > 0\) when the GB moves up while the upper grain translates to the right or when the GB moves down while the upper grain translates to the left. The coupling is called perfect, or pure, if \(\beta\) is a geometric constant depending only on the GB bicrystallography and not on the boundary velocity, driving force or any other physical parameter. An interesting feature of the coupling effect is that the coupling factor is a multi-valued function of the crystallographic angles. Owing to this multiplicity, physically the same GB can display different coupling factors and thus different mechanical responses to the same applied stress \[8,14,15\].

For symmetrical boundaries with \(\phi = 0\), the fourfold symmetry generates four possible coupling modes with the coupling factors \(\beta = 2\tan(\theta/2 - \pi k/4), k = 0, 1, 2, 3\). Two of them, corresponding to the smallest magnitude of \(\beta\), have been observed in simulations \[8,14,15,19,20\] and experiments \[21–25\], whereas the remaining two modes require exceedingly high stresses to activate and have never been reported. The two observed coupling modes are referred to as \([100]\) and \([110]\) type and have the coupling factors

\[\beta_{(100)} = 2\tan\left(\frac{\theta}{2}\right) > 0\]  \hfill (1)

and

\[\beta_{(110)} = 2\tan\left(\frac{\theta}{2} - \frac{\pi}{4}\right) < 0\]  \hfill (2)
respectively. The coupling factors measured in the MD simulations [8,14,15,19,20] and experiments [21–24,26] accurately follow the (100) mode at angles $\theta < \theta_c = 36^\circ$ and the (110) mode at angles $\theta > \theta_c$. At the critical angle $\theta_c$, the coupling factor abruptly switches from one mode to the other with a change of sign.

Analysis of the bicrystal symmetry [19] shows that, for symmetrical boundaries with inclinations $\phi = \pm45^\circ$, a similar discontinuity occurs at the critical angle of $90^\circ - \theta_c$. It was also suggested [8,15,19], and confirmed by simulations [19], that coupling continues to exist for asymmetrical GBs and exhibits the same two coupling modes with a discontinuous transition between them. It was shown that the discontinuity line plotted in the coordinates $(\theta, \phi)$ possesses certain symmetries reflecting the symmetry of the bicrystal and has an S-shape, as displayed in Figs. 2 and 22 of Ref. [19]. In particular, the line should pass through the points $[\theta = \theta_c, \phi = 0]$, $[\theta = 45^\circ, \phi = 22.5^\circ]$, $[\theta = 45^\circ, \phi = -22.5^\circ]$, $[\theta = 90^\circ - \theta_c, \phi = 45^\circ]$ and $[\theta = 90^\circ - \theta_c, \phi = -45^\circ]$. Other than this, the precise shape of the discontinuity line is specific to the material, and may additionally depend on the GB velocity, temperature and other factors.

For the purposes of this work, we will approximate the discontinuity line in the $(\theta, \phi)$ coordinates by the simplest possible shape satisfying the mentioned symmetry requirements. Namely, the line is composed of straight segments with either constant $\theta$ or constant $\phi$, as shown in Fig. 2a. We will additionally assume that the coupling factors corresponding to the (100) and (110) modes continue to follow Eqs. (1) and (2), respectively, for all inclination angles.

That the coupling factor for a given mode depends only on the tilt angle, but not the inclination, was suggested in Refs. [8,15] based on geometric considerations. Recent simulations [19] have revealed that $\beta$ can in fact depend on the inclination angle. Nevertheless, the sign and basic features of the coupling behavior were found to be qualitatively consistent with the above equations.

Having a model for the function $\beta(\theta, \phi)$ for a plane GB, we now consider a curved GB segment spanning the inclination angles $-45^\circ \leq \phi < 45^\circ$ (quarter of a loop). On the coupling diagram in Fig. 2a, such a segment could be represented by a vertical path at a fixed misorientation angle $\theta$ (not shown). If $\theta \leq \theta_c$ or $\theta \geq 90^\circ - \theta_c$, the coupling factor remains constant along this path and the entire GB segment can be described by a single coupling factor given by Eq. (1) or (2). If $\theta_c < \theta < 90^\circ - \theta_c$, the coupling factor is discontinuous along the segment and changes sign twice, at $\phi = -22.5^\circ$ and $\phi = 22.5^\circ$. In this case, the segment can be characterized by an effective coupling factor $\beta$ obtained by averaging the $\beta_{(100)}$ and $\beta_{(110)}$ values over the inclination angles. Thus, the misorientation dependence of the effective coupling factor of a curved GB predicted by this model is given by

$$
\beta(\theta) = \begin{cases} 
2 \tan \left( \frac{\theta}{2} \right), & 0 \leq \theta \leq \theta_c \\
\tan \left( \frac{\theta}{2} \right) + \tan \left( \frac{\theta - \theta_c}{2} \right), & \theta_c < \theta < 90^\circ - \theta_c \\
2 \tan \left( \frac{\theta - \theta_c}{2} \right), & 90^\circ - \theta_c \leq \theta < 90^\circ
\end{cases}
$$

and is plotted in Fig. 2b. Due to the fourfold symmetry of the lattice, this effective coupling factor remains valid for any GB segment whose inclination range comprises a multiple of $45^\circ$. This includes a half-loop ($-90^\circ \leq \phi < 90^\circ$) and even a circular GB ($-180^\circ \leq \phi < 180^\circ$).

3. Effective coupling factor and rotation of an embedded grain

As a test of the proposed model, we first apply it to the case of an isolated cylindrical grain with a circular cross-section shrinking by capillary forces [6,7,11–13]. The dynamics of the shrinking process are described by the equations [7,12]
\[ v_n = -\dot{R} = M\gamma/R \]  
\[ v_\parallel = R\dot{\theta} = \beta v_n \]  
where \( R \) is the grain radius, \( \gamma \) is the GB free energy, which is assumed to be constant, \( M \) is the GB mobility coefficient and the dot denotes the time derivative. A positive coupling factor causes the grain to rotate towards larger misorientation angles, while a negative coupling factor causes rotation towards smaller angles. Eq. (3) and Fig. 2b predict that grains with \( \theta < \theta_c \) will rotate to larger angles while grains with \( \theta > \theta_c \) will rotate in the opposite direction. This is exactly what was observed in the MD simulations [12], where the grains with the initial misorientations of 16° and 28° rotated towards higher angles while the grains with the initial misorientations of 37° and 44° rotated towards lower angles. In all initial misorientations, the rotation stopped when the angle approached the critical value \( \theta_c = 36° \). It was found that the discontinuity of the coupling factor arising at \( \theta = \theta_c \) creates a “frustrated” state of the boundary and prevents grain rotation. This behavior of the isolated grain is fully consistent with the present model.

Attention will now be turned to an embedded grain placed on a plane GB. Fig. 3 explains our convention for the misorientation angles, \( \theta_U \) and \( \theta_L \), of the upper and lower curved GBs. In both cases, the misorientation angle is considered positive if the \([100]\) direction in the embedded grain is obtained by counter-clockwise rotation of the \([100]\) direction in the neighboring grain, otherwise it is negative. The following sign convention is used for the coupling factors. The coupling factor \( \beta_U \) of the upper GB is considered positive if it produces translation of the upper grain to the right when the GB moves down. Accordingly, the coupling factor \( \beta_L \) of the lower GB is considered positive if it produces local translation of the lower grain to the left when the GB moves up. This convention is consistent with that of the previous work \([8,15]\) applied locally to each part of the embedded grain by orienting the observer so that the GB would look horizontal with the embedded grain above and the neighboring grain below.

If both \( \beta_U \) and \( \beta_L \) are positive, both GBs try to rotate the embedded grain in the counter-clockwise (i.e. positive) direction; if both are negative, they try to rotate the grain in the clockwise (negative) direction. Different signs of \( \beta_U \) and \( \beta_L \) lead to competition, with the outcome dependent on the magnitudes of the coupling factors. When the crystal symmetry dictates \( \beta_U = -\beta_L \), we expect that the rotational forces will cancel each other and there will be no net rotation of the grain. As a crude approximation, we will assume that the grain rotation is governed by the effective coupling factor

\[ \beta_e = \frac{\beta_U + \beta_L}{2} \]  
In other words, to predict the rotation we mentally replace the embedded grain sitting on the plane GB by an isolated cylindrical grain \([6,12]\) with the effective coupling factor \( \beta_e \).

Applying Eq. (3) to both the upper and lower boundaries, we have \( \beta_U = \beta(\theta_U) \) and \( \beta_L = \beta(\theta_L) \). Thus \( \beta_e \) defined by Eq. (6) is a function of two misorientation angles, \( \theta_U \) and \( \theta_L \). This allows us to make predictions of the embedded grain rotation for any given set of crystallographic orientations of the three grains.

Given the approximate character of the model, it is unlikely to provide quantitatively accurate results. However, we expect that it will be capable of predicting at least the correct sign of \( \beta_e \) and thus the direction of the grain rotation. When testing this capability of the model, we will use the \((\theta_U, \theta_L)\) diagram, which shows the domains of different signs of \( \beta_e \) (Fig. 4). The white and gray areas in the diagram designate the positive and negative signs of \( \beta_e \), whereas the boundaries between them indicate the cancellation of rotation when \( \beta_e \) goes through zero. The points and numbers appearing on this diagram will be explained later.

Fig. 3. Crystallography of an isolated grain placed on a plane GB. All three grains are obtained by rotation around the common [001] axis. For the upper and lower curved GBs, the misorientation angles \( \theta_U \) and \( \theta_L \) are defined using the convention shown in the top right corner.

![Fig. 3](image_url)

Fig. 4. Diagram of the effective coupling factor \( \beta_e \) of an embedded grain, with white and gray colors representing the domains of \( \beta_e > 0 \) and \( \beta_e < 0 \), respectively. The coupling factor is discontinuous along the dashed lines. The points represent the crystallographic orientations implemented in the MD simulations with the numbers corresponding to the first column in Table 1. The angles are measured in degrees.

![Fig. 4](image_url)
4. Methodology of atomistic simulations

Copper was chosen as the model material to enable comparison with previous simulation studies of coupled GB motion and grain rotation [8,12,14,15,19,20,27]. Atomic interactions in Cu were modeled with an embedded-atom potential fit to experimental and first-principles data [28]. The potential predicts that the melting temperature of $T_m = 1327$ K in good agreement with the experimental value of 1356 K. The MD simulations employed the ITAP Molecular Dynamics (IMD) program [29] implementing the isothermo-isobaric (NPT) ensemble with zero pressure maintained independently in each Cartesian direction. This statistical ensemble ensured that, as the embedded grain was shrinking, the loss of GB free volume did not generate significant internal stresses. Multiple snapshots of the simulation block containing atomic coordinates and other relevant information were saved during the simulations and used for post-processing.

The simulation block represented a slab with approximate dimensions 300 by 300 by 36 Å and periodic boundary conditions in all three Cartesian directions. It contained three grains: two grains were separated by a plane GB and the third, cylindrical grain, with an initial diameter of 200 Å, was embedded at the center of the plane GB (Fig. 5). The construction of this simulation block included several steps. First, a periodic bicrystal with a fully relaxed plane [001] tilt GB was created using the procedures described in Refs. [8,30]. The GB plane was aligned parallel to the laboratory coordinate plane (x, z) and normal to the y axis. The lattice orientations of the upper and lower grains were represented by rotation angles $\psi_U$ and $\psi_L$, respectively, of the [001] directions in the grains around the common [001] axis. These angles are measured relative to the laboratory coordinate system and considered positive in the counter-clockwise direction.

The embedded grain was created by carving out a cylindrical void and filling it with a grain whose [001] axis was rotated by an angle $\psi_0$ around the [001] direction. The grain radius was slightly larger than the radius of the void, creating a 1.2 Å overlap between the lattices. This overlap was needed to permit the removal of excess atoms, as explained below (insertion of atoms would be much more difficult than removal). The energy of the system was then minimized to produce two curved GBs with structures close to equilibrium. To this end, the initial curved GBs were mentally divided into small portions and the energy was minimized by removing atoms and varying atomic positions within each portion and its vicinity while keeping all other atomic positions fixed. After all the portions had been relaxed by this procedure one at a time, all of the atoms of the simulation block were made dynamic and the energy was again minimized with respect to all atomic positions. Finally, the block was heated up to the temperature of 300 K and annealed by running NPT MD for 200 ps.

This multi-step process created a partially equilibrated structure suitable for the subsequent MD simulations at high temperatures. Without this pre-equilibration, heating the system to high temperatures would produce a highly non-equilibrium structure, causing local plastic deformation and melting. The MD simulations were performed by raising the temperature to the desired level over a period of 200 ps, followed by an isothermal anneal in the NPT ensemble. The isothermal anneals were conducted at the temperatures of 800 and 1000 K.

For stress-driven simulations, the block size in the y direction was increased to 1400 Å to create room for GB migration in that direction. The boundary conditions were modified by creating fixed surface layers normal to the y-direction, as in our previous work [19]. To apply a shear, the lower surface layer was moved as a rigid body parallel to the positive x direction with a constant velocity of $v_x = 1$ m s$^{-1}$ relative to the upper surface layer.

The crystallographic orientations studied in this work are listed in Table 1. Knowing the angles $\psi_U$, $\psi_L$ and $\psi_0$ relative to the coordinate system, we determined the lattice misorientation, $\theta_p = \psi_U - \psi_L$, across the plane GB and the lattice misorientations, $\theta_U = \psi_0 - \psi_U$ and $\theta_L = \psi_0 - \psi_L$, across the upper and lower curved GBs, respectively. If the angle came out negative, it was shifted by 90$^\circ$ to obtain the equivalent misorientation within the studied angular range $\{0 \leq \theta < 90^\circ\}$.

The following procedure was devised to continuously monitor the size and crystallographic orientation of the embedded grain during the simulations. First, the individual grains were automatically identified in every snapshot. To this end, local environments of all atoms were divided into perfect and defected using local rotational distortions of nearest-neighbor bonds as the criterion of perfection. High-angle GBs were identified as continuous defected layers separating adjacent grains. For low-angle GBs composed of discrete dislocations, the defect-selection criterion was adjusted so that the dislocation cores would appear to be overlapped, creating a continuous layer with a thickness larger than the lattice spacing. This adjustment was made only once for the boundary with the smallest tilt angle studied here, after which the defect detection parameters were fixed. This ensured that all other GBs were also represented by relatively thick continuous layers.

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Fig. 5. Simulation block used in this study. The grains are colored by lattice orientation relative to the laboratory coordinate system (x, y, z). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
The grain interiors were then identified as topologically disconnected perfect-lattice regions separated by GB layers, as illustrated in Fig. 6a. The atoms belonging to each grain were found by applying a cluster-identification algorithm. Next, 12 vectors pointing to nearest neighbors of every perfect-lattice atom were averaged over each grain, producing three reference vector sets representing the lattice orientations in the grains. The orientation angle $\psi$ of the embedded grain was determined as the Euler angle of the respective vector set relative to the axes of the laboratory system. To avoid possible complications due to the fourfold symmetry of the lattice, we used two laboratory systems rotated relative to each other by $45^\circ$ around the [001] axis and the calculated angles were cross-checked for consistency. This algorithm was tested by recomputing the lattice orientations in the snapshots of the isolated cylindrical grain studied in Ref. [12]. The computed angles $\psi$ were found to agree with the angles determined by the independent method [12] within tenths of a degree.

Finally, the nearest-neighbor environments of all atoms, perfect or defected, were analyzed and the reference vector set representing the grain with the smallest mean-squared deviation from the local set was identified for each atom. This allowed us to partition all of the atoms between the three grains and outline atomically sharp boundaries between the grains (Fig. 6b). Using this partitioning procedure, the instantaneous area $A$ of the embedded grain was calculated by $A = A_0 N / N_0$, where $N$ is the number of atoms assigned to the grain, $N_0$ is the total number of atoms in the simulation block and $A_0$ is the total cross-sectional area of the block normal to [001].

In the structural images presented below, we show either the color-coded grain orientations or defected regions identified by local rotational distortions adjusted to best reveal the dislocations and the GB structure.

5. MD results for spontaneous grain shrinkage

At all temperatures and geometries tested, the embedded grain shrank spontaneously until the simulation block became a bicrystal. In most cases the remaining GB was flat. In some cases a kink was observed immediately after the embedded grain had vanished. We expect that, given enough time, the kink would disappear, but a proof would be shown later, most of the grain rotation occurs towards the end of the simulation. Thus, any artifacts that might have been created by the initially non-equilibrium dihedral angles were unlikely to affect the observed rotation. Likewise, during the long anneals at the elevated temperatures, the GBs formed new structures that were independent of their initial structures equilibrated at 300 K.

Table 1
Crystalllographic parameters of the GBs studied in this work and the coupling factors computed from the geometric model (the last column indicates the direction of the embedded grain rotation observed in the MD simulations).

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<th>$\psi_U$ (°)</th>
<th>$\psi_0$ (°)</th>
<th>$\theta_p$ (°)</th>
<th>$\theta_L$ (°)</th>
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<td>41.13</td>
<td>24.87</td>
<td>-0.079</td>
<td>0.441</td>
<td>0.181</td>
<td>Positive</td>
</tr>
</tbody>
</table>

Fig. 6. Illustration of the procedure for automatic identification of grains and calculation of the lattice orientations. (a) Identification of individual grains as areas separated by thick “defected” regions outlining GBs. (b) Partitioning of atoms between the grains by best-matching lattice orientation to produce sharp GBs.
The initial misorientations of the upper and lower GBs modeled in this work are represented by points on the diagram in Fig. 4. Twelve different cases were tested, which are labeled by the numbers indicated in the first column of Table 1. The MD results will be presented below in the order of the cases.

Case 1: The plane GB has a high-angle misorientation, \( \theta_p = 57.48^\circ \), whereas the curved GBs have low-angle misorientations, \( \theta_U = 16.26^\circ \) and \( \theta_L = 73.74^\circ \). (16.26° away from the perfect lattice arising at 90°). We thus have a low-angle grain sitting on a high-angle GB. Fig. 7 shows the dislocation structure that can be resolved in the low-angle GBs. Due to the symmetry of this geometry, it is expected that the upper and lower GBs will try to rotate the embedded grain in opposite directions. The coupling forces are expected to balance and no rotation should be observed. This prediction also follows from Eq. (6) and Fig. 4: the coupling factors of the curved GBs are \( \beta_U = -\beta_L = 0.286 \) with the effective value \( \beta_e = 0 \) (Table 1). This prediction is confirmed by the MD simulations, which do not detect any significant rotation during the grain shrinkage (Fig. 8a). A similar geometry was previously studied in the PFC simulations [13], where it was also found that the grain did not rotate. Because the curved GBs have low-angle misorientations, they are expected to have a small reduced mobility [12]. This is indeed observed in Fig. 8b; for example, at the temperature of 1000 K the embedded grain does not disappear after a 20 ns long anneal.

Case 2: The curved GBs now have high-angle misorientations, \( \theta_U = 53.13^\circ \) and \( \theta_L = 36.87^\circ \), corresponding to \( \Sigma = 5 \) (reciprocal density of coincidence sites), whereas the plane GB has a low-angle misorientation, \( \theta_p = 73.74^\circ \). The plane boundary is composed of discrete dislocations, as illustrated in Fig. 9. Again, the symmetry dictates that the curved GBs have opposing coupling factors, \( \beta_U = -\beta_L = 0.167 \), and are not expected to cause grain rotation. Accordingly, the MD simulations do not reveal any significant grain rotation (Fig. 10a). Because the high-angle GBs have a large reduced mobility [12], the embedded grain disappears fast (e.g. in approximately 0.8 ns at 1000 K; see Fig. 10b).

Fig. 11 shows a set of snapshots during the grain shrinkage at 800 K. Because the two curved GBs have equal mobilities, the shape of the embedded grain remains symmetric with respect to the plane GB. At this temperature it takes 1.8–4.2 ns for the embedded grain to disappear.

Case 3: This case is similar to the previous one but all three lattices have been rotated by the same angle of 45°. This rotation does not change the misorientation angles between the grains but results in a 45° shift of the inclination angles of all three GBs. According to the model discussed in Section 3, we expect that this shift will not affect the coupling factors (Table 1) and that the symmetry preventing the grain rotation must be preserved. This prediction is indeed confirmed by the MD simulations, which show that the embedded grain shrinks without rotation.
The kinetics of shrinkage (Fig. 12b) are very similar to those in case 2, demonstrating that the mobility of these GBs does not depend sensitively on the inclination. For example, at 1000 K the embedded grain disappears in about 0.9 ns (compared with 0.8 ns in the previous case).

Case 4: This case demonstrates the effect of broken symmetry. The embedded grain is sitting on a high-angle GB ($\psi_p = 30.51^\circ$) and is bounded by a low-angle boundary ($\theta_U = 14.25^\circ$) on top and a high-angle boundary ($\theta_L = 44.76^\circ$) at the bottom. The initial structure of this grain is illustrated in Fig. 13. Our model predicts that the upper GB has a positive coupling factor ($\beta_U = 0.250$) and is trying to rotate the embedded grain in the positive direction, whereas the lower GB has a negative coupling factor with a smaller magnitude ($\beta_L = -0.005$) and is trying to rotate the grain in the negative direction (Table 1). By Eq. (6), the upper grain should win and cause a positive rotation, which is indeed confirmed by the MD simulations (Fig. 8a). The amount of rotation ($\sim 2^\circ$) is small in comparison with other cases considered below, which is consistent with the relatively small magnitude of the effective coupling factor $\beta_L$.

Selected snapshots of the shrinking grain are shown in Fig. 14. Because the upper curved GB has a much smaller reduced mobility than the lower one, the grain shrinkage occurs predominantly by migration of the lower GB upwards. Eventually the lower GB becomes nearly plane and then continues to migrate upwards inside the embedded grain, gradually eliminating the dislocations forming the upper GB. This process requires upward motion of
the triple lines and causes the plane GB to bow out towards the upper tip of the embedded grain. At the point when the embedded grain disappears, the formerly plane GB has developed a significant protrusion, which then recedes and the boundary eventually returns to its plane state. This process resembles the grain growth during primary recrystallization of metals, in which a moving GB consumes dislocations in the deformed grain driven by the release of strain energy.

Case 5: The situation is different from the previous case in two ways. First, both the plane GB and the lower curved GB now have low-angle misorientations and are composed of discrete dislocations, as illustrated in Fig. 15. Second, both the upper and lower curved GBs have positive coupling factors, which amplify each other and create a large driving force for grain rotation (Table 1).

The results of the MD simulations are consistent with the model predictions: the embedded grain shrinks very quickly and rotates in the positive direction. At the temperature of 1000 K it vanishes within 2 ns (Fig. 8). The time evolution of the grains illustrated in Fig. 16 reflects the large difference in the mobilities of the low- and high-angle GBs. The structural changes can be understood by imagining for a moment that the plane GB and the lower curved GB are completely immobile. The upper GB then shrinks by driving the triple lines along the landscape formed by the two low-angle GBs. As a result, the entire embedded grain moves down and ends up at the bottom of the “crafter” formed by the lower GB, where it eventually disappears, leaving behind a significant depression in the shape.
of the formerly plane GB. Because the mobilities of the two low-angle GBs are in fact non-zero, the shape of the “crafter” smooths to some extent. However, the depression is still significant. Some of it remained for as long as the MD simulations could be run, suggesting that complete flattening of the GB would require longer time scales.

Cases 6–11: Starting from the symmetrical case 2, we produce new initial configurations by rotating the embedded grain in the positive direction in 2° increments. The plane GB remains unchanged ($\theta_p = 73.74°$) but the misorientation angles of the upper and lower curved GBs increase and decrease, respectively, while remaining in the high-angle range. This breaks the symmetry and creates an increasing disparity between the coupling factors of the upper and lower GBs. The model predicts that the embedded grain should rotate in the positive direction. Interestingly, at $\psi_0$ between 8° and 10° the misorientation angle of the lower GB goes through 45° and the coupling factor $\beta_L$ changes sign. Nevertheless, $\beta_U$ remains negative and is predicted to overpower the positive sign of $\beta_L$, producing net rotation in the negative direction.

The latter prediction is confirmed by the MD simulations, which show negative rotation of the embedded grain for all cases 6–11 (Fig. 10a). The rate of the grain shrinkage tends to decrease with $\psi_0$ (Fig. 10b).

Cases 12–16: Again, we start from the symmetrical case 3 and create new initial configurations by rotating the embedded grain away from $\psi_0 = 45°$. This breaks the symmetry and activates grain rotation in the positive direction. While $\beta_L$ remains positive, $\beta_U$ decreases and reaches the critical angle $\theta_e$, at which it changes sign. The model predicts, however, that the effective $\beta_e$ should remain positive. Positive rotation is indeed observed in the MD simulations in all these cases (Fig. 12a). As in the previous cases 6–11, the activation of rotation is accompanied by a decrease in the rate of grain shrinkage (Fig. 12b).

6. MD simulations under applied shear

The stress-driven simulations were conducted on the embedded grain with $\theta_U = 16.26°$ and $\theta_L = 73.74°$, corresponding to case 1 in Table 1. The low-angle grain is sitting on a high-angle GB with $\theta_p = 57.48°$, as shown in Fig. 7. Recall that, by the crystal symmetry, $\beta_p = 0$ and the embedded grain spontaneously shrinks without rotation.

The applied shear stress breaks the symmetry and is expected to induce both GB motion and grain rotation. Specifically, for the lower curved GB we have $\beta_L < 0$ and expect its coupled motion in the upward direction. This motion, in turn, should create a force for the grain rotation in the negative direction. For the upper curved GB we have $\beta_U > 0$ and, by our sign convention for coupling factors, we expect the applied shear to cause its coupled motion also in the upward direction. This motion should rotate the embedded grain in the negative direction as well. For the plane GB we have $\theta_p > 90° - \theta_e$, corresponding to the (110) coupling mode. The coupling factor computed from Eq. (2) is negative, $\beta_{(110)} = -0.583$, predicting coupled motion upwards. Thus, the applied shear is expected to cause upward motion of all three GBs and rotation of the embedded grain in the negative direction.

This prediction can be rationalized by examining the dislocation structure of the GBs, as shown schematically in Fig. 17. Although the plane GB with $\theta_p = 57.48°$ cannot be treated as a dislocation array, this schematic is only conceptual and assumes that all three boundaries have low-angle misorientations. Furthermore, the “curved”
boundaries are assumed to form facets composed of (100) and (1/2)(110) dislocations. The important observation is that all (100) dislocations present in the system have the same orientation of the Burgers vector. Thus, under the shear stress applied as indicated on the diagram, these dislocations experience a Peach–Koehler force driving them in the upward direction. By contrast, the (1/2)(110) dislocations do not see any Peach–Koehler force from the applied shear. Thus, the net force acting to the dislocations should drive all three GBs in the upward direction.

Selected snapshots of the MD simulation are shown in Fig. 18. Exactly as predicted, the plane GB and the embedded grain are moving up. Note the asymmetry in the shape of the embedded grain developing at later stages of the simulation. This asymmetry is to be expected from the system geometry. As evident from the schematic in Fig. 17, clockwise rotation of the embedding grain decreases the misorientation angle of the upper curved GB and increases the misorientation angle of the lower curved GB. As a result, the mobility of the lower GB increases while that of the upper GB decreases. The faster motion of the lower GB is manifested in its smaller curvature in comparison with the slower-moving upper GB, an effect which becomes especially pronounced towards the end of the simulation (Fig. 17c). For the same reason, the initially plane GB develops a protrusion (Fig. 17d), which eventually smooths out after the embedded grain disappears. The situation is somewhat reminiscent of case 4 shown in Fig. 13. Besides the confirmation of the model predictions, these snapshots demonstrate the interesting effect of stress-driven motion of an entire grain sitting on a GB.

The predicted lattice rotation in the negative direction is confirmed by the time dependence of the angle ψ displayed in Fig. 19. Furthermore, the simulation was repeated by reversing the shear direction. As expected, both the plane GB and the embedded grain were observed to move down.

Fig. 17. Schematic illustration of the dislocation structure of a tricrystal with misorientation angles similar to case 1 in Table 1. The arrows indicate the directions of the applied shear. The vertical arrows show the direction of the Peach–Koehler forces acting on the dislocations. As a guide to the eye, the dashed lines indicate selected {100} planes in the grains.

Fig. 18. Simulation snapshots for the lattice orientations ψ_U = 73.74°, ψ_L = 16.26° and ψ_0 = 0°, corresponding to case 1 in Table 1 under an applied shear at the temperature of 1000 K. The shear direction is indicated in Fig. 17.

Fig. 19. Time dependencies of the embedded grain orientation ψ in the presence of applied shears with two different directions at 1000 K. The lattice orientations are ψ_U = 73.74°, ψ_L = 16.26° and ψ_0 = 0°, corresponding to case 1 in Table 1.

Accordingly, the lattice of the embedded grain now rotated in the positive direction by about the same amount. This consistency with the MD results further confirms the predictive capability of the proposed model.

7. Discussion and conclusions

We have investigated the process of capillary-driven shrinkage of an embedded cylindrical grain initially centered on a plane GB. Although this tricrystalline structure
is highly idealistic, it captures some of the key features of polycrystalline materials: namely, the multiplicity of GBs surrounding a given grain and the presence of triple junctions. This structure is ideal for an initial study of GB motion and grain rotation in the presence of multiple GBs and triple junctions.

We find that grain rotation is a common feature of this structure during both spontaneous grain shrinkage and under applied shear stresses. Grain rotation was not observed only in cases where symmetry led to opposing coupling factors of equal magnitude. By contrast to an isolated cylindrical grain whose rotation stops as soon as the misorientation angle reaches the angle of discontinuity of the coupling factor [12], in the tricrystalline structure such a discontinuity in one of the GBs does not necessarily prevent continuing grain rotation. For example, the upper GB goes through a discontinuity and switches the sign of the coupling factor at \( \theta = 36^\circ \) between cases 5 and 16 (Fig. 4). Nevertheless, the coupling factor of the lower GB remains positive and ensures that the grain continues to rotate in the positive direction.

The MD simulations demonstrate that the presence of triple junctions does not block the grain rotation as one might expect. However, they do affect the dynamics of grain shrinkage by making it significantly slower. For example, in case 1 (Table 1), where the plane GB does not rotate due to the symmetry constraint, it takes approximately 23 ns for it to vanish at the temperature of 1000 K. For comparison, the circular isolated grain with a similar cross-sectional area, the same misorientation angle of 16.26° and a fixed center preventing its rotation vanishes in only 2 ns at the same temperature [12].

Part of this retardation can be attributed to the smaller driving force due to the additional tension of the plane GB opposing the shrinkage of the embedded grain in the tricrystal. However, the large magnitude of the effect suggests additional factors that could be related to the triple junction drag [17,18,31,32] and/or the effect of the triple lines on dislocation reactions. As discussed in Ref. [12], grain shrinkage must be accompanied by annihilation of part of the dislocation content of the GBs. It was found that this annihilation occurs by propagation of the dislocation content along the GB by a chain of dislocation reactions. Dislocations with opposite Burgers vectors propagate along the GB by this mechanism, meet and annihilate. Such chains of reactions can be disrupted by the triple lines. In addition, the dislocation reactions occurring at the triple line itself could form a kinetic bottleneck of the process. In this particular case (case 1 in Table 1), the reaction consists of the recombination of two perpendicular \((1/2)(11 \bar{0})\) dislocations into one \((100)\), which then attaches to the dislocation array forming the plane GB and increases its length (cf. Fig. 17). In other cases, however, the reactions can be more complex, and deserve a separate detailed study in the future.

We proposed a simple analytical model describing the embedded grain by a single effective coupling factor \( \beta_c(\theta_U, \theta_L) \) defined by Eqs. (3) and (6). Based on crude approximations, this model is not intended to be fully quantitative, but is expected to be capable of predicting at least the correct direction of grain rotation (or lack thereof), depending on the lattice orientations in the three grains. It should be remembered that this model is specific to the [001] texture and would have to be re-derived for other choices of the tilt axis. The model predictions are summarized in the \((\theta_U, \theta_L)\) diagram in Fig. 4, which shows the domains of positive and negative rotation. (By the symmetry of the problem, only a quarter of these domains are distinct; all others are symmetrically equivalent.) The cases tested in this work are shown by points in the diagram and sample several of the distinct domains and their boundaries. Note that these points designate the initial misorientations of the GBs; as the embedded grain rotates, each point moves along a line with \( \theta_U - \theta_L = \text{const.} \)

As discussed in Section 5 and evident from Table 1, the MD results are in full agreement with the model predictions. Specifically, the tricrystals with predicted \( \beta_c = 0 \) do not show any grain rotation. When rotation is observed, its sign always matches the model prediction. This agreement suggests that, despite its approximate character, this model captures the essential physics of the process and can be used in future applications.

The results of the shear-driven MD simulations are also consistent with the proposed model. As predicted from the coupling factors, all three GBs move in the same, upward direction while the embedded grain rotates towards smaller angles. Changing the sign of the applied shear stress reverses the directions of the GB motion and grain rotation. These simulations provide an additional validation of the model. They also demonstrate the interesting effect of stress-driven motion of an entire grain. Indeed, a shear stress applied to an isolated cylindrical grain would only distort its cross-section to an oval but would not cause the motion of its center of mass. In the tricrystal case, the opposite signs of the coupling factors of the upper and lower GBs lead to their correlated motion in the same direction. In other words, the coupling effect is capable of inducing not only stress-driven GB migration [8] but also stress-driven grain motion.

At the same time, the fact that, even for a simple configuration, such as the tricrystal the rotation diagram (Fig. 4) contains a significant number of domains suggests that possible generalizations of our model to multiple grains will quickly increase the complexity. For example, imagine a cylindrical grain placed at a triple junction parallel to the cylinder axis. This creates a four-crystal structure in which the embedded grain is bounded by three curved GBs and three triple junctions. As a generalization of Eq. (6), one could describe this grain by an effective coupling factor obtained by averaging the \( f_b \) of the three GBs, each given by an equation similar to Eq. (3) with the respective misorientation angle. Fig. 4 would then be replaced by a three-dimensional diagram in the three misorientation angles, with the domains of positive and negative rotation.
represented by isolated or joint pyramids and prisms. Adding another grain would take us to a four-dimensional space of variables, which is less amenable to graphical presentation. While the model itself could remain meaningful and might still give correct sign predictions, its testing by simulations becomes a daunting task. Nevertheless, such models can be worth further exploration, especially in the context of incorporation in statistical models of polycrystalline evolution.

Returning to the tricrystal case, another path towards the reality of polycrystalline materials would be a study of tilt axes other than [001], especially if such axes are different in different grains. The next step would be a full three-dimensional simulation, something that has not been done in the context of grain rotation, even for an isolated grain. In terms of analytical models, the challenge lies in the fact that the angular dependence of the coupling factor given by Eq. (3) is specific to [001] tilt boundaries. Geometric models of coupling for other tilt axes, or more general types of GBs, are yet to be developed.

Another limitation of the proposed model is the assumption that each GB is perfectly coupled and follows geometric rules. It is known from simulations [8,19,20] as well as bicrystal experiments [33] that coupling disappears at high temperatures. Impurity segregation can also influence the degree of coupling [34–36] and reduce the amount of grain rotation. Given the multitude of factors that may affect the coupled state of GBs in polycrystalline materials, it should not be surprising that grain rotation was observed in some experiments [2,4,5,37] but not in others [38]. It is hoped that the present work takes a step towards a better understanding of at least the crystallographic aspect of this complex problem and may motivate further studies of the temperature dependence, segregation effect and other factors.

Acknowledgements

This work was supported by the U.S. Department of Energy, the Physical Behavior of Materials Program, through Grant No. DE-FG02-01ER45871.

References