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Grain boundary migration and grain rotation studied by molecular dynamics

Z.T. Trautt, Y. Mishin

Department of Physics and Astronomy, MSN 3F3, George Mason University, Fairfax, VA 22030, USA

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Abstract

We report on molecular dynamics simulations of an isolated cylindrical grain in copper shrinking under capillary forces. At low temperatures, the coupling between grain boundary (GB) migration and grain translation induces rotation of the grain towards higher or lower misorientation angles, depending on the initial misorientation. The dynamics of the GB motion and grain rotation are studied as functions of the initial misorientation angle and temperature. The effects of imposed constraints blocking the grain rotation or exerting a cyclic torque are examined. The simulation results verify several predictions of the model proposed by Cahn and Taylor [Acta Mater 52, 4887 (2004)]. They also indicate that the GB motion is never perfectly coupled but instead involves at least some amount of sliding. This, in turn, requires continual changes (annihilation or nucleation) in the GB dislocation content. Dislocation mechanisms that can be responsible for the motion of curved GBs and dislocation annihilation in them are proposed.

Keywords: Molecular dynamics; Grain boundary; Dislocation; Grain rotation

1. Introduction

Grain rotation in polycrystalline materials is part of microstructure evolution, along with other processes such as phase transformations, grain coarsening and dislocation motion. Grain rotation was observed experimentally during plastic deformation [1,2], recrystallization [3] and grain growth [4,5]. It was also found to accompany stress-driven grain boundary (GB) motion in bicrystals subject to a tensile load [6]. Several theories have been proposed to explain the driving forces and mechanisms of grain rotation. In particular, it was suggested that grains rotate to decrease the GB free energy $\gamma$ [4]. This process was indeed observed in atomistic computer simulations [7,8]. It was also shown [9,10] that grain rotation can be induced by GB motion due to the coupling effect, which will be discussed below. In the latter case, grain rotation can lead to an increase in $\gamma$, provided that the GB area shrinks sufficiently fast to produce a net decrease in the total GB free energy [9]. As a mechanism of grain rotation, diffusion-controlled viscous flow along GBs was adopted in several models [3,11,12]. For low-angle GBs, models combining dislocation glide with climb were developed to explain subgrain rotation and coalescence during recovery and early stages of recrystallization [3,5,11,13,14].

Cahn and Taylor [10] proposed a unified approach to GB motion, grain translation and grain rotation. The underlying idea of their approach is the existence of coupling between GB migration and rigid translations of the grains parallel to the boundary. A coupled GB produces plastic shear deformation of the lattice regions swept by its motion, which in turn causes grain translation. Conversely, a shear stress applied parallel to a coupled GB induces its normal motion. Stress-induced GB motion was observed experimentally [6,15–25] and by atomistic simulations [9,26–30], and is considered to be responsible for stress-induced grain growth in nanocrystalline materials [19,31–34]. The reader is referred to [35] for a recent review of the coupling effect.

An interesting consequence of the coupling effect, pointed out in [9,10], is that coupled motion of a curved
GB must induce grain rotation. Cahn and Taylor [10] proposed a model of grain rotation based on a linear dissipation approximation. They applied this model to describe evolution of an isolated cylindrical grain with a circular cross-section shrinking by capillary forces. A number of possible scenarios were discussed, depending on the presence or absence of coupling, GB sliding, applied shear stresses and other factors [10]. In particular, the model predicts that a low-angle GB induces grain rotation towards higher angles and thus larger $\gamma$. Such a rotation was indeed observed in molecular dynamics (MD) simulations of a cylindrical grain in a Lennard–Jones system [9]. More recently, Wu and Voorhees [36] applied the phase field crystal (PFC) method to model the shrinkage of an isolated circular grain in a two-dimension hexagonal system. At misorientation angles below $10^\circ$, the grain shrinkage produced grain rotation towards increasing angles.

Since the publication of [9], significant progress has been achieved in understanding the geometric rules and atomic mechanisms of coupled GB motion through a series of experimental studies [6,15–25] and MD simulations [26–30]. It seems timely to revisit the grain rotation problem, building on the new knowledge combined with recent improvements in computational methodologies and power. The goals of this work are to conduct a systematic MD study of a cylindrical grain similar to [9], compare the results with the Cahn–Taylor model [10], and gain insights into microscopic mechanisms of GB motion and grain rotation. Our simulations are based on a realistic interatomic potential for copper, which was employed in previous work on coupled GBs [26–29]. We study a wide range of temperatures and examine the effects of imposed constraints and applied torque on GB dynamics.

To make the paper self-contained, we start with a brief overview of the Cahn–Taylor model (Section 2) and point to several predictions that can be directly compared with atomistic simulations. Our simulation methodology is introduced in Section 3. In Sections 4–7 we present our simulation results of the dynamics of GB motion and grain rotation, including simulations with prohibited grain rotation and applied torque. In Section 8 we compare our simulation results with the Cahn–Taylor model and discuss possible dislocation mechanisms that could be responsible for the coupled GB migration, GB sliding and grain rotation. Our conclusions are summarized in Section 9.

2. The Cahn–Taylor model of grain shrinkage and rotation

Fig. 1 illustrates the geometry and conventions adopted in the Cahn–Taylor model [10]. We consider an isolated cylindrical grain with a circular cross-section and an instantaneous radius $R$. The grain is embedded in an outer environment: $f = 2\pi R^2 p$. The excess $p$ can be due to a defect concentration, magnetic energy or other factors. This excess is assumed to be homogeneous and independent of time. The GB free energy $\gamma$ is assumed to be uniform over the boundary and depend on $\theta$ only: $\gamma = \gamma(\theta)$. Thus,

\begin{equation}
\dot{\gamma} = 2\pi R^2 \gamma + 2\pi R^2 \dot{\theta} + 2\pi R \dot{p} R
\end{equation}

where $\gamma' := \partial \gamma / \partial \theta$. Also,

\begin{equation}
\dot{\omega} = 2\pi R \sigma v_n
\end{equation}

Combining the above equations,

\begin{equation}
\dot{f}_{\text{tot}} = -2\pi R (P_n v_n + P_\| v_\parallel)
\end{equation}
where
\[ P_a := p + \frac{\gamma - \beta \gamma'}{R} + \beta \sigma \] (8)
and
\[ P_\parallel := \sigma - \frac{\gamma'}{R} \] (9)
are interpreted as driving forces causing the normal GB motion and GB sliding, respectively.

The next step is to postulate linear-response relationships between the driving forces and the respective velocities:

\[ v_n = M_a P_a \] (10)
\[ \tilde{v}_\parallel = M_\parallel P_\parallel \] (11)
where \( M_a \) and \( M_\parallel \) are phenomenological mobility coefficients corresponding to GB migration and GB sliding, respectively. After some rearrangements, we finally arrive at the kinetic equations [10]

\[ v_n = -\dot{R} = M_a \left( p + \frac{\gamma - \beta \gamma'}{R} + \beta \sigma \right) \] (12)
\[ \tilde{v}_\parallel = R \dot{\theta} = \beta M_a \left( p + \frac{\gamma - \beta \gamma'}{R} + \beta \sigma \right) + M_\parallel \left( \sigma - \frac{\gamma'}{R} \right) \] (13)
Dividing Eq. (13) by Eq. (12), we obtain the differential equation [10]

\[ -\frac{d\theta}{d \ln R} = \beta + \frac{M_\parallel \left( \sigma - \frac{\gamma'}{R} \right)}{M_a \left( p + \frac{\gamma - \beta \gamma'}{R} + \beta \sigma \right)} \] (14)
Eqs. (12)–(14) constitute the basic equations of the Cahn–Taylor model and permit predictions of the grain evolution. We will now analyze several particular cases that are most relevant to our simulation conditions. For simplicity, it will be assumed that \( p = 0 \) throughout this discussion.

2.1. Case A: free grain, no sliding

Consider the evolution of a free grain (no applied stresses, \( \sigma = 0 \)) in the absence of GB sliding (\( M_\parallel = 0 \)). Eqs. (12)–(14) become

\[ v_n = -\dot{R} = M_a \frac{\gamma - \beta \gamma'}{R} \] (15)
\[ \tilde{v}_\parallel = \beta v_n \] (16)
\[ -\frac{d\theta}{d \ln R} = \beta \] (17)

Two cases are possible.

Case A1: no coupling. Suppose the boundary is not coupled, i.e. \( \beta = 0 \), which is the case for high-angle GBs at high temperatures [28]. Then the grain is shrinking by the capillary force \( \gamma / R \) without rotation (\( v_\parallel = 0 \)). Eq. (15) gives

\[ \dot{R} = -M_a \frac{\gamma}{R} \] (18)
Since the misorientation angle does not vary during this process, we can treat \( M_a \) and \( \gamma \) as constants and integrate Eq. (18) to obtain

\[ R^2(t) = R_0^2 - 2M_a \gamma \theta t \] (19)
where \( t \) is the elapsed time. This is a trivial case of curvature driven GB migration obeying parabolic kinetics.

Case A2: coupled motion. Suppose the GB is coupled whereas sliding is still prohibited. The driving force for GB motion now consists of two competing terms: \( \gamma / R \), leading to reduction in the GB area, and \( -\beta \gamma / R \), driving the misorientation angle towards a nearby minimum of \( \gamma \). These terms can have opposite signs and, hypothetically, the second one can even win over the first, producing grain growth (\( v_n < 0 \)). This interesting possibility was pointed out by Cahn and Taylor [10], but has not been seen in experiments or simulations so far, including the present work.

Whether the grain shrinks or grows, it simultaneously rotates with the angular velocity \( \dot{\theta} = \beta v_n / R \). The kinetics of GB motion and grain rotation can be rather complex because \( \beta \), \( \gamma \) and \( M_a \) generally depend on \( \theta \). As a simple example, consider a low-angle GB with \( 0 \leq \theta \ll 1 \). Knowing that \( \beta \approx \theta \) [10,28] and adopting the Read–Shockley approximation [37],

\[ \gamma \approx \gamma_0 \theta(a - \ln \theta) \] (20)
where \( \gamma_0 \) and \( a \) are constants, Eq. (15) gives

\[ \dot{R} = -M_a \frac{\gamma_0 \theta}{R} \] (21)
For a GB composed of discrete dislocations, the approximation \( M_a \approx M_b \theta / \theta \) can be invoked [10], where \( M_b \) is the glide mobility of a single dislocation and \( b \) is the magnitude of the Burgers vector. With these approximations, Eq. (15) gives the parabolic law of grain shrinkage,

\[ R^2(t) = R_0^2 - 2M_b \gamma_0 \theta t \] (22)
which looks similar to Eq. (19) but has a different kinetic coefficient.

Integrating Eq. (17) with \( \beta \approx \theta \), we obtain

\[ R \theta = R_0 \theta_0 \] (23)
This relation shows that the shrinking grain rotates towards higher misorientation angles and thus larger \( \gamma \). As was discussed in Ref. [9], this increase in \( \gamma \) does not contradict the thermodynamics because the GB area decreases faster, leading to a reduction in the total GB free energy \( 2\pi R \gamma \).

A more accurate approximation for \( \beta(\theta) \) is

\[ \beta = 2 \tan \frac{\theta}{2} \] (24)
This relation was derived geometrically for symmetrical tilt GBs with arbitrary angles \([27,28]\), and was verified by atomistic simulations \([26–28,30,38]\) and experimental measurements \([22,25]\). Using this expression for \(\theta(\theta)\), Eq. (17) is readily integrated to give
\[
R \sin \frac{\theta}{2} = R_0 \sin \frac{\theta_0}{2} = \text{const}.
\] (25)

This solution has a simple interpretation for low-angle boundaries. According to the Frank formula \([39,40]\), \(2\sin(\theta/2) = b/d\), where \(d\) is the spacing between GB dislocations. Thus, \(R \sin(\theta/2) = Nb/2\), where \(N\) is the number of dislocations and \(Nb\) is the total magnitude of their Burgers vector. In other words, Eq. (25) expresses the conservation of the dislocation content of the boundary when it moves in a perfectly coupled mode.

Eq. (25) can be used as a test of coupled GB motion in simulations and experiments. Such tests should be taken with caution, however, because the underlying Eq. (24) may not remain accurate after averaging over all inclination angles sampled by the boundary. In addition, the right-hand side of Eq. (25) can remain constant only between the radii \(R_0\) and \(R_0\sin(\theta_0/2)\).

### 2.2. Case B: free grain, no coupling

Suppose the GB is not coupled (\(\beta = 0\)) but sliding is now allowed. The normal GB motion is described by Eq. (18), but its integration leading the parabolic relation Eq. (19) is now only justified in the absence of grain rotation. Generally, however, the grain can rotate and \(M_n\) may not remain constant. The rotation equation,
\[
v_\parallel = R \dot{\theta} = -M_n \frac{\gamma}{R}
\] (26)

shows that the grain rotates by GB sliding towards a nearest minimum of \(\gamma\). Eq. (14) becomes \([10]\)
\[
\frac{d \theta}{d \ln R} = M \frac{\gamma}{M_n}.
\] (27)

Integration of Eqs. (26) and (27) requires knowledge of the angular dependencies of \(\gamma\), \(M_n\) and \(M_1\).

Consider a particular case of a low-angle GB (\(0 \leq \theta \ll 1\)), Using the Read–Shockley equation, Eq. (20), we have \(\gamma/\gamma \approx 1/\theta\). As before, we can assume \(M_n \approx M_\parallel b/\theta\) \([10]\). Because little is known about \(M_\parallel\), keeping it constant appears reasonable. Then integration of Eq. (27) gives
\[
\theta = \theta_0 + \frac{M_\parallel}{M_n b} \ln \frac{R}{R_0}
\] (28)

This solution predicts that the shrinking grain rotates towards smaller angles until the two grains merge (\(\theta \to 0\)) at a finite radius \(R^* = R_0 \exp(-M_\parallel b/\theta /M_\parallel)\). If we keep both \(M_n\) and \(M_\parallel\) constant, then
\[
\theta^2 = \theta_0^2 + \frac{2M_\parallel}{M_n} \ln \frac{R}{R_0}
\] (29)

and \(R^* = R_0 \exp(-M_\parallel \theta_0^2/2M_\parallel)\). Cahn and Taylor \([10]\) assumed that \(M_n \approx M_\parallel b/\theta\) while \(M_\parallel\) goes to zero as \(\theta^2\) and obtained a different solution, by which \(\theta \to 0\) at \(R \to 0\). Despite these differences, the important common feature of all these solutions is that the grain rotates towards smaller angles, which is evident from Eq. (27) and the fact that for low-angle GBs \(\gamma > 0\). This behavior can be contrasted to the case of coupled GB motion, Case A2 above, in which the grain rotates towards higher angles. Thus, for low-angle GBs the direction of grain rotation can be used as an indicator of predominance of either coupling or sliding.

Another property of Eqs. (28) and (29), as well as other solutions based on different assumptions regarding the mobilities \([10]\), is that they do not satisfy the dislocation conservation condition equation, Eq. (25). Mathematically, \(R \sin(\theta/2)\) cannot remain constant when both \(R\) and \(\theta\) decrease with time. Physically, the reduction in GB area and rotation to smaller angles both require a decrease in the total number of geometrically necessary dislocations accommodating the grain misorientation. This process cannot occur without a mechanism for annihilation of GB dislocations.

### 2.3. Case C: grain rotation prohibited

Now consider a situation when the grain is subject to imposed constraints that prohibit grain rotation. The constraints give rise to shear stresses \(\sigma\) acting along the GB and canceling the rotation caused by all other driving forces. The situation is described by setting \(\epsilon_\parallel = 0\) in Eq. (13), solving this equation for \(\sigma\) and inserting the solution in Eq. (12). This gives
\[
v_\parallel = -\dot{R} = -\frac{M_\parallel v_\parallel}{R(\beta^2 M_n + M_\parallel)}
\] (30)

Because there is no rotation, all parameters remain constant and this equation is readily integrated to give the familiar parabolic law of GB shrinkage:
\[
R^2(t) = R_0^2 - \frac{M_\parallel v_\parallel /\beta^2 M_n + M_\parallel}{t}
\] (31)

In the absence of coupling (\(\beta = 0\)), \(M_\parallel\) cancels out and this equation correctly reduces to the previously derived Eq. (19). Coupling decreases the kinetic coefficient in Eq. (31), resulting in slower GB motion.

If the boundary is coupled (\(\beta \neq 0\)) while sliding is difficult (small \(M_\parallel\)), Eq. (31) becomes
\[
R^2(t) \approx R_0^2 - \frac{M_\parallel v_\parallel /\beta^2}{t}
\] (32)

showing that the grain shrinkage rate is controlled by sliding. In the absence of sliding (\(M_\parallel \to 0\)), the kinetic coefficient vanishes and the GB cannot move. In other words, when grain rotation is prohibited by constraints, coupled GB motion must be accompanied by sliding. This can be easily understood: the coupled GB motion “wants” to cause grain rotation, as we saw in Case A2 above. To
prevent the net rotation, back-stresses must be generated by the constraints so that to induce GB sliding in the opposite direction and exactly cancel the rotation due to coupling. If this “reverse sliding” process is difficult or impossible, the GB moves slowly or not at all.

For a small-angle grain, the presence of sliding destroys the conservation of GB dislocations. This is clear from the fact that, in the absence of grain rotation, the density of geometrically necessary dislocations must remain constant and satisfy the Frank relation for the given θ. Since the GB area decreases due to the grain shrinkage, part of the dislocations must be eliminated in order to maintain the required constant density.

3. Methodology of atomistic simulations

Copper was used as a model material representing a typical face-centered cubic (fcc) metal. Atomic interactions in Cu were modeled with an embedded-atom potential fit to experimental and first-principles data [41]. The melting temperature of Cu predicted by this potential is \( T_m = 1327 \text{ K} \), in good agreement with the experimental value 1356 K. The MD simulations employed the ITAP Molecular Dynamics (IMD) program [42] with temperature controlled by a Nose–Hoover thermostat. The simulation block represented a slab with approximate dimensions 300 by 300 by 36 Å and periodic boundary conditions in all three Cartesian directions. The block initially contained a perfect fcc lattice, with crystallographic directions parallel to its edges. A new grain was created by rotating a cylindrical region of approximately 200 Å in diameter by an angle θ around the [001] direction normal to the surface of the slab (Fig. 2). The new grain was surrounded by a [001] tilt GB, with the tilt angle θ and the inclination angle varying over the 360° interval.

Special efforts were taken to create an initial GB structure that would be as close to equilibrium as possible. Immediately after the rotation, the GB structure was highly non-equilibrium and contained pairs of atoms that were either too close or too far apart in comparison with the equilibrium atomic spacing. An immediate application of MD would produce a shock that would cause local melting and re-solidification in the GB region. To perform a smoother relaxation of the GB structure to a more stable atomic configuration, we applied a thermodynamic integration procedure similar to the cleavage technique known in interface thermodynamics [43,44]. The method implements a transition from a set of two non-interacting single crystals, grain A and grain B, to two non-interacting bicrystals: one with a cylindrical grain A embedded in grain B and the other with a cylindrical grain B embedded in grain A [45]. This transition is achieved by gradually increasing an atomic interaction parameter \( \lambda \) between the initial (\( \lambda = 0 \)) and final (\( \lambda = 1 \)) states during an MD simulation run. The MD temperature (300 K) and time (a few hundreds of picoseconds) were chosen to be small enough to avoid a significant shrinkage of the grain during the equilibration process. The benefit of this method is that the atomic overlaps and the GB free volume are smoothly accommodated on a timescale of overdamped dynamics. Evidence of the high degree of equilibration of the GB structure achieved by this method was the absence of any residual defects when the shrinking grain completely vanished during the subsequent MD simulations.

Embedded grains with misorientation angles \( \theta_0 = 5.72°, 16.26°, 28.07°, 36.87° \) and 43.60° were created. Their evolution was studied by MD simulations performed at the temperatures of 500–1350 K. The simulation was considered complete when the embedded grain vanished and the system turned into a single crystal. The isothermal–isobaric ensemble was implemented, in which zero pressure was maintained independently in each Cartesian direction. This ensemble was chosen to ensure that as the grain shrinks, the loss of grain boundary free volume does not create significant internal stresses. Multiple copies (snapshots) of the simulation block containing the coordinates of all atoms, their energies and other relevant information were saved during the simulations and used at the post-processing stage.

An important part of this study was monitoring the size and crystallographic orientation of the embedded grain. The following procedure was developed for this purpose. The first step was to assign an orientation parameter, \( \phi \), to each individual atom in a given snapshot. This was accomplished by comparing positions \( r_n \) of \( n \) nearest neighbors of a chosen atom \( i \) with positions \( \bar{r}_{mk}(x) \) of 12 first-nearest neighbors of an atom in the perfect fcc lattice rotated by an angle \( \alpha \) around [001]:

\[
\psi_i(x) = \sum_{j=1}^{n} \sum_{m=1}^{12} \sum_{k=1}^{3} \exp \left[ - \left( \frac{r_{ijk} - \bar{r}_{mk}(x)}{a_0(x)} \right)^2 \right]
\]

(33)

Here \( a_0 \) is the equilibrium lattice constant and index \( k \) runs over three Cartesian components of a vector. The values of \( n \) around 50 were found to give the most robust computation results. The quantity \( \psi_i(x) \) was calculated for 90 equally spaced rotations \( x \) ranging from 0° to 89°. The angular range from 0 to 90° includes all distinct rotations of the grain due to the fourfold symmetry around [001].
The rotation delivering a maximum of \( \psi(x) \) and thus corresponding to the best match between the actual and ideal lattice orientations was denoted \( \phi \) and assigned to atom \( i \) as its orientation parameter. A histogram of orientation parameters of all atoms of the simulation block displayed two sharp peaks representing the two grains present in the system (Fig. 3). The difference between the average values of \( \phi \) corresponding to the peaks was taken as the grain misorientation angle \( \theta \) in the given snapshot. The atoms contributing to each peak were assigned to the corresponding grains.

The uncertainty in \( \theta \) calculated by this method is on the order of \( 1^\circ \), depending on the magnitude of \( \theta \) and temperature. Given this uncertainty, the initial misorientation angles of the grains will be henceforth referred to as \( \theta_0 = 6^\circ, 16^\circ, 28^\circ, 37^\circ \) and \( 44^\circ \), respectively.

The instantaneous area \( A \) of the embedded grain was calculated by \( A = A_0 N/N_0 \), where \( N \) is the number of atoms in the grain as determined from the orientation parameter, \( N_0 \) is the total number of atoms in the simulation block and \( A_0 \) is the cross-sectional area of the block normal to [001]. The effective radius \( R \) of the embedded grain was defined by \( R := \sqrt{A/\pi} \).

4. Grain boundary structures

For the low-angle misorientations of \( 6^\circ \) and \( 16^\circ \), the GB was found to be composed of discrete dislocations with eight Burgers vectors \( \mathbf{b} \): \( \pm[100], \pm[010], \pm1/2[110] \) and \( \pm1/2[1\overline{1}0] \). These Burgers vectors could be readily determined by drawing Burgers circuits around individual dislocation cores. Eight types of symmetrical tilt GBs could be formed out of these dislocations. Accordingly, the entire GB could, in principle, be composed of eight plane symmetrical facets, forming an octagon, as shown schematically in Fig. 4. If all facets corresponding to the same magnitude of \( \mathbf{b} \) reduce to a single dislocation, the cross-section of the grain becomes a square.

In the actual simulations, the observed grain shape was more rounded and usually close to a circle. In some cases, the grain developed “dynamic” facets, which formed and disappeared during the grain shrinkage. The faceting was
especially pronounced for the $6^\circ$ GB, which tended to have a square shape composed of $\pm 1/2[110]$ dislocations similar to Fig. 4b. Fig. 5 presents an example of octagon-type faceting of the $16^\circ$ boundary at 900 K.

At larger misorientation angles, drawing Burgers circuits around individual dislocations becomes ambiguous. However, the boundaries are typically composed of distinct structural units with the kite shape (see the examples in Fig. 4 of Ref. [28]). Because the shapes and orientations of the kites closely resemble the dislocation cores in low-angle boundaries, their rows parallel to [001] can still be interpreted as dislocation cores. Similar to the low-angle misorientations, the shapes of the shrinking high-angle grains were on average circular, but with notable distortions and occasional dynamical facets forming and disappearing during the GB motion.

5. Grain shrinkage and rotation

At all temperatures tested, the grains with the initial misorientations $\theta_0 = 16^\circ, 28^\circ, 37^\circ$ and $44^\circ$ shrank spontaneously until the simulation block turned to a defect-free single crystal. The grain with $\theta_0 = 6^\circ$ did not shrink significantly during reasonable simulation times, even at temperatures exceeding $T_m$. This is not surprising, given the low free energy of this small-angle boundary and thus the small capillary driving force for its shrinkage. At any given temperature, the time for the grain to disappear was longest for $\theta_0 = 16^\circ$ and much shorter and relatively independent of $\theta_0$ for larger angles (see the example in Fig. 6).

At temperatures up to approximately $0.8T_m$, the grain shrinkage was accompanied by grain rotation. Fig. 7 presents a qualitative proof of rotation using a colored vertical stripe as a marker line. Note the rotation of the segment located inside the shrinking grain relative to the outer grain. The zig-zag shape of the marker line left after the grain disappearance is a result of shear deformation of the lattice produced by coupled GB motion.

Fig. 8a illustrates the time dependencies of the misorientation angle $\theta$ at the temperature of 800 K ($0.6T_m$). Note that the grains with $\theta_0 = 16^\circ$ and $28^\circ$ rotate with increasing angle, which is in agreement with theoretical predictions [10], previous MD simulations [9] and recent PFC simulations [36]. By contrast, the grain with the initial misorientation of $37^\circ$ rotates towards smaller angles, although only by $2^\circ$ or less. Finally, the grain with $\theta_0 = 44^\circ$ shows a significant rotation with decreasing angle. Overall, it is apparent from Fig. 8a that the grains rotate towards a misorientation angle of about $36^\circ$, whether $\theta_0$ is below or above this angle.
At temperatures above $0.8T_m$, the amount of grain rotation decreases significantly. Near the melting point, the grains cease to rotate with the exception of the $\theta_0 = 16^\circ$ misorientation, which continues to display some rotation. For example, at the temperature of 1300 K ($0.98T_m$) the grain with $\theta_0 = 16^\circ$ rotates until $\theta \approx 22^\circ$ (Fig. 8b). The disappearance of grain rotation at high temperatures correlates with the disappearance of coupling found in the MD studies of stress-driven motion of plane GBs [28]. In particular, the terminal misorientation of $\sim 22^\circ$ reached by the $16^\circ$ boundary at 1300 K is in good agreement with the coupling-to-sliding transition on the temperature-angle diagram of mechanical responses shown in Fig. 8 of [28].

Simulations at even higher temperatures reveal the effects of GB melting and premelting caused by grain rotation. Previous simulation studies [46–54] showed that many GBs with relatively high misorientation angles transform to liquid layers below the bulk melting point $T_m$ (the premelting effect), whereas low-angle boundaries do not premelt and can be superheated to metastable states above $T_m$ [55]. The present simulations show that the $\theta_0 = 16^\circ$ GB belongs to the second category, in that it can be readily superheated above $T_m$. However, as the misorientation angle increases during the grain rotation and the GB free energy rises, this boundary gradually shifts towards the first category: it begins to display premelting and eventually triggers complete melting of the enclosed grain. As illustrated in Fig. 9, this GB is initially “dry” at 1350 K but gradually develops a liquid-like layer as the grain rotates towards higher angles. When the liquid layer becomes sufficiently thick, the enclosed grain essentially becomes a cylindrical crystallite embedded in a pocket of liquid (Fig. 9b). Since the temperature is above $T_m$, this crystallite melts and eventually disappears, leaving a cylindrical liquid pocket behind (Fig. 9c). The radius of the liquid pocket is apparently smaller than the critical radius $R^*$ of homogeneous liquid nucleation at this temperature. As a result, the liquid crystallizes until the entire system becomes a single crystal (Fig. 9d).

When the same simulation is repeated at the temperature of 1375 K, the boundary again develops a liquid layer and the enclosed grain melts (Fig. 10). However, the remaining liquid pocket now grows until the entire system melts. This growth indicates that the radius of the liquid pocket exceeds $R^*$ at 1375 K. These simulations allow us to roughly estimate $R^*$ as being above 100 Å (the grain radius) at 1350 K but below 100 Å at 1375 K.

6. Grain behavior when rotation is prohibited or imposed

In the simulations discussed in the previous section, the enclosed grain was free to rotate due to its isolated geometry and the periodic boundary condition along the cylindrical axis. The grains with the initial misorientations $\theta_0 = 16^\circ$ and $28^\circ$ were also tested with the center of the grain fixed in order to prohibit grain rotation. The fixed region consisted of a cylinder with $R = 25$ Å, leaving about 75 Å for GB migration before the boundary reaches the fixed region. Only the results obtained during this period of the simulation are reported here.

We found that at temperatures when the free grain shrank with rotation, it continued to shrink when rotation was prohibited. However, the shrinkage occurred significantly more slowly than for the free grain. This shrinkage retardation effect when grain rotation is blocked will be quantified in Section 7.

The same fixed region was used to apply a torque to the enclosed grain, using the $\theta_0 = 16^\circ$ grain as a model. To this end, the fixed region was rotated around the axis passing through its center of mass by $\pm 0.05^\circ$ every 10 ps during an MD simulation performed at 300 K. This slow rotation created an elastic strain field, which propagated towards the GB and exerted a shear stress. The direction of the imposed rotation was reversed after a set period of time, resulting in a sawtooth loading pattern. Note that, since the rotation rate was fixed, selecting a different period changed the angular amplitude of the rotation.

The results for the loading periods of 1, 2, 3 and 4 ns are reported in Fig. 11. These periods correspond to the imposed angular amplitudes of $2.5^\circ$, $5^\circ$, $7.5^\circ$ and $10^\circ$. 

![Fig. 8. Time dependence of the lattice misorientation angle $\theta$ for different initial misorientations of an enclosed grain at two temperatures: (a) 800 K and (b) 1300 K.](image-url)
respectively. It should be noted that the angles plotted in this figure were averaged over the entire cylindrical grain. Due to the elastic strains and statistical errors, the computed angular amplitudes are somewhat different from the imposed. In all four cases, we find that the imposed torque causes oscillatory GB displacements, indicating the existence of coupling. The grain shrinks when rotated to larger angles and grows when rotated to smaller angles. For the amplitudes of 2.5°, 5° and 7.5°, the GB oscillations are nearly reversible, suggesting almost perfect coupling and thus conservation of most of the dislocation content during the boundary motion. For the largest amplitude (10°), the oscillations become strongly biased towards net shrinkage of the grain, signifying a substantial reduction in the number of GB dislocations from one cycle to the next. Most of the dislocations are eliminated during the growth phase, when the angle decreases by about 10° while the increase in the grain radius is relatively small.

We emphasize that none of the free grains studied in this work shrank by capillary forces alone at the temperature of 300 K. Nevertheless, we find that an applied cyclic torque of sufficient amplitude activates grain shrinkage even at this low temperature.

7. Grain boundary mobility

GB migration experiments and simulations are traditionally interpreted in terms of the GB mobility $M$, defined as the linear coefficient between the normal GB velocity $v_n$ and the driving force $P$, i.e. $v_n = MP$. As was discussed in Section 2, in the presence of coupling, the relation between $v_n = -dR/dt$ of a circular GB and the capillary driving force $P = \gamma/R$ can be nonlinear. Furthermore, $\gamma$ is not known exactly in most experiments and simulations. Despite these uncertainties, we found it useful to extract from our simulations the so-called reduced mobility $A$ defined by

$$A := \frac{1}{2} \frac{dR^2}{dt}$$

(34)
This quantity represents the product of $\gamma$ times an effective (linearized) mobility $M$. Both $\gamma$ and $M$ depend on the misorientation angle and can therefore vary during the simulation. To reduce this uncertainty, $A$ was computed by a linear fit of only the initial portion of the $R^2$ vs. $t$ plot, in which $\theta$ varied by less than $1^\circ$.

The results of the reduced mobility calculations are presented in Fig. 12 as an Arrhenius diagram for the temperature range 700–1300 K. The non-linearity of the plots reflects the fact that the mechanisms responsible for the GB motion, as well as other factors such as the presence or absence of grain rotation and the amounts of coupling and sliding, change with temperature. An interesting feature of this plot is the convergence of all curves to the same “universal” mobility at $T \rightarrow T_m$, which suggests the formation of a liquid-like GB structure near $T_m$. Furthermore, for all initial misorientations except for $\theta_0 = 16^\circ$, the Arrhenius plot turns over near the melting point, producing a slight decrease in $A$ when approaching $T_m$. This can be explained by a reduction in $\gamma$ when the GB turns to a liquid layer.

Fig. 12 also shows that, at a given temperature, the reduced mobility is smallest for $\theta_0 = 16^\circ$ and higher and nearly independent of misorientation at larger angles. For clarity, the misorientation dependence of $A$ is plotted separately in Fig. 13. This plot shows that, at 700 K, $A$ increases by an order of magnitude between $\theta_0 = 16^\circ$ and $28^\circ$. By contrast, at high temperatures the reduced mobility depends on misorientation only slightly and approaches a “universal” value of about $10^{-7} \text{ m}^2 \text{ s}^{-1}$.

The relatively small reduced mobility of low-angle GBs and its increase with misorientation angle are consistent with experimental measurements [56] and previous atomistic simulations [57] using a half-loop geometry. In this geometry, the GB is also driven by curvature, although the GB plane spans the angular interval of $180^\circ$, as opposed to $360^\circ$ in our work. An increase in GB mobility with misorientation angle was also reported in the PFC.
Note, however, that the misorientation dependence of $A$ found in this work (Fig. 13) disagrees with the approximation $M_{db} \approx M_{pf}/\theta$ discussed in Section 2. This approximation, combined with the Read–Shockley formula (Eq. (20)) for $c$, predicts a decrease in $A = \gamma M_{db}$ with $\theta$, not an increase. This prediction relies on the assumption of conservation of dislocations during the GB motion. This disagreement presents another demonstration that the dislocation content of the shrinking GBs studied in this work was not conserved. Nor was it conserved in the previous half-loop simulations and experiments.

The reduced mobility $A$ was also computed from the simulations in which the grain rotation was blocked. The results for $h_0 = 16^\circ$ and $28^\circ$ are shown in Fig. 14. This plot reveals that at low temperatures, when coupling is strong and the free grain rotates as it shrinks, prohibiting the grain rotation results in a significant reduction in $A$. As coupling weakens and the rotation gradually disappears at high temperatures, the free- and fixed-grain mobilities converge, as expected. For $\theta_0 = 16^\circ$ (Fig. 14a), some difference between the free- and fixed-grain mobilities still remains even near the melting point, which is consistent with the existence of rotation of this grain near $T_m$ (cf. Fig. 8a). For the grain with $\theta_0 = 28^\circ$, the two mobilities completely merge above 1100 K, which is in agreement with the absence of rotation and premelting behavior of this boundary. These results demonstrate that the ability or inability of grains to rotate can have a significant impact on GB dynamics.

8. Discussion

8.1. Comparison of simulations with the Cahn–Taylor model

Free grain at high temperatures. In agreement with previous work [28], our simulations confirm the significant decrease ($\theta_0 = 16^\circ$) or complete disappearance (all other $\theta_0$) of coupling at high temperatures. Under such
Grain rotation towards local minima of $\gamma(\theta)$ was observed in recent MD simulations of cylindrical grains with $30^\circ < \theta_0 < 48^\circ$ in a two-dimensional Lennard–Jones system [8]. In that system, however, the boundaries exhibited deep cusps of energy at several special misorientations and apparently had a lower sliding resistance. On the other hand, the recent PFC simulations [36] have shown that grain rotation ceases to occur when $\theta$ exceeds $15^\circ$. Furthermore, the shrinkage kinetics were found to be accurately parabolic, in agreement with both the Cahn–Taylor model [10] and our simulations. The PFC method represents material’s behavior on diffusive timescales and approximately corresponds to MD simulations near the meting point. It is, therefore, reassuring that our high-temperature results are in qualitative agreement with the PFC simulations [36]. A quantitative comparison cannot be made for several reasons, including the different crystallography and dimensionality of the two simulations.

**Free grain at low temperatures.** At low temperatures the GBs are coupled and the grains are expected to rotate [10] (Case A2). The GB shrinkage kinetics do not have to be parabolic because the coupling factor, GB free energy and GB mobility can vary with $\theta$. The $R^2$ vs. $t$ plots observed in our simulations do exhibit a distinct curvature at low temperatures (Fig. 6). The Cahn–Taylor model predicts that at small $\theta$ the parabolic law should continue to work, see Eq. (22). The $R^2$ vs. $t$ plot for $\theta_0 = 16^\circ$ is indeed initially linear, but develops significant curvature during later stages of the simulation.

We find that the grains with $\theta_0 = 16^\circ$ and $28^\circ$ rotate to larger angles (Fig. 8a), which validates the model predictions and is in agreement with previous MD [9] and PFC [36] simulations. The grain with $\theta_0 = 37^\circ$ shows little rotation, whereas the grain with $\theta_0 = 44^\circ$ rotates with decreasing angle. A possible explanation for the reversal of grain rotation around $36^\circ$ will be discussed in Section 8.3.

In the absence of coupling, the grain can rotate by GB sliding in a direction that would lead to a reduction in $\gamma$ (Section 2.2, Case B). Small-angle grains can rotate to even smaller angles, large-angle grains to a nearby minimum of $\gamma(\theta)$. Our simulations did not detect such rotations. We find that at high temperatures, when coupling disappears, the grains simply cease to rotate (Fig. 8b). (The observed slight rotation of the $\theta_0 = 16^\circ$ grain is towards larger angles and is due to partial coupling of this boundary.) We can conclude that the GBs studied in this work have a large sliding resistance preventing grain rotation in the absence of coupling. In addition, our boundaries have only shallow minima of $\gamma(\theta)$ (except for $\theta = 0$) even at 0 K [28]. These minima are likely to become even more shallow at finite temperatures.

**Reduced Mobility A (m^2/s) vs. 1/Temperature (1/K)**

![Fig. 14. Arrhenius plot comparing the reduced grain boundary mobility computed under two different conditions: when the grain was free to rotate and when the center of the grain was fixed to prohibit rotation. (a) Initial misorientation $\theta_0 = 16^\circ$, (b) initial misorientation $\theta_0 = 28^\circ$.](image)

**Reduced Mobility A (m^2/s) vs. 1/Temperature (1/K)**

![Fig. 15. Grain radius squared as a function of time for capillary driven shrinkage of an isolated grain with different initial misorientation angles $\theta_0$ at the temperature of 1300 K. The linearity of the plots confirms the parabolic kinetics.](image)
The simulations offer the opportunity to test the dislocation conservation condition Eq. (25), which states that the parameter

\[ C := \frac{R \sin \theta}{R_0 \sin \theta_0} \tag{35} \]

must remain constant and equal to 1. This parameter was recorded in our simulations and is plotted in Fig. 16 as a function of \( \theta \) for all \( \theta_0 \) and temperatures tested. Notice that all curves, except for \( \theta_0 = 16^\circ \), become nearly vertical at high temperatures, reflecting the disappearance of grain rotation and a sharp decline of \( C \). Even in the cases of largest grain rotation, however, such as \( \theta_0 = 16^\circ \) and \( 28^\circ \) at 700 K, \( C \) is not constant. In fact, by the moment these grains disappear, \( C \) declines to around half its initial value.

Part of the observed variations in \( C \) could be attributed to inaccuracy of Eq. (24) underlying the predicted constancy of \( C \). Indeed, Eq. (24) was originally derived \[27,28\] for plane symmetrical tilt GBs. It is not clear how averaging over all inclination angles modifies this equation. However, the large magnitude of this discrepancy leads us to believe that it is dominated by another factor. Namely, the dislocation content of the GBs is actually not conserved due to the presence of some amount of sliding, simultaneously with coupling, in all our simulations. As pointed out in Section 2, sliding along a curved GB must be accompanied by creation or destruction of some of its dislocation content. The drastic decrease in \( C \) observed in Fig. 16 indicates that a substantial part of the dislocation content was eliminated in all cases.

When \( \theta \) was relatively small and temperature not too high, it was possible to confirm the elimination of dislocations by directly counting their numbers in MD snapshots. As an example, Fig. 17 compares two snapshots taken during an MD run with \( \theta_0 = 16^\circ \) at 700 K. During the time period when the angle increases from 18.5° to 26.8°, the total number of dislocations decreases from 60 to 44. Possible mechanisms of the dislocation elimination process will be discussed in Section 8.2. It is expected that, if MD simulations could be performed at significantly smaller angles, the moving dislocations would remain clear of each other and would be conserved. This mode of GB motion was indeed observed in the PFC simulations \[36\] for angles between 5° and 10°. Implementation of this mode of GB motion in MD simulations is problematic due to the timescale limitation of the method. As mentioned above, the GB with \( \theta_0 = 6^\circ \) did not migrate on the MD timescale, even above the melting point.

**Grain rotation prohibited.** Simulations with prohibited grain rotation allow us to test the parabolic relation Eq. (31) predicted by the model. Such simulations were performed for grains with \( \theta_0 = 16^\circ \) and \( 28^\circ \). As expected from.

Fig. 16. The dislocation conservation criterion \( C \) defined by Eq. (35) as a function of misorientation angle for various temperatures and initial misorientation angles.

Fig. 17. Snapshots of MD simulations of the shrinking grain with the initial misorientation of 16° at the temperature of 700 K. The dislocation cores are outlined. (a) After 5 ns; the grain boundary contains 60 dislocations. (b) After 17.5 ns; the grain boundary contains 44 dislocations.
Eq. (31), the plots of $R^2$ vs. $t$ were found to be fairly linear at low temperatures when the GBs are strongly coupled (see the examples in Fig. 18). These plots are also linear at high temperatures when coupling disappears, which is also in agreement with the Cahn–Taylor model [10]. At intermediate temperatures the plots develop a slight curvature at late stages of the simulation.

As demonstrated in Fig. 14, when the grain is fixed the GBs migrate significantly more slowly in comparison with a free grain at the same temperature and $\theta_0$. This retardation effect is consistent with our conclusion that the GBs studied here have a large sliding resistance. Indeed, as discussed in Section 2.3, the constraint imposed to prevent the grain rotation causes additional GB sliding in order to cancel the coupling velocity $\beta_{\text{vn}}$. When sliding is difficult, the shrinkage process is controlled by the sliding mobility coefficient $M_0$ (see Eq. (32)) and the process is slow. On the other hand, when coupling disappears at high temperatures, so does the need for additional sliding. Accordingly, the shrinkage kinetics of the free and fixed grains must converge. This is confirmed by the reduced mobility plots in Fig. 14.

8.2. Mechanisms of grain boundary motion and dislocation annihilation

In this section we discuss possible mechanisms of GB motion during the enclosed grain evolution. For simplicity we will consider low-angle boundaries composed of discrete dislocations. However, most of this discussion is transferable to high-angle boundaries in which the role of dislocations is played by rows of structural units.

Two questions arise regarding the dislocation mechanisms. The first one is related to the motion of dislocations during coupled GB migration. For symmetrical tilt GBs composed of identical dislocations, the migration is by collective dislocation glide along parallel slip planes [9,10,26–28]. However, a curved boundary contains at least two types of dislocations with intersecting slip planes. How do these dislocations glide without blocking each other?

The second question is related to dislocation annihilation, a process which is required for GB sliding when it leads to a decrease in the misorientation angle. Srinivasan and Cahn [9] suggested that the only possible dislocation reaction would be the annihilation of dislocation pairs with opposite Burgers vectors located on opposite ends of the cylinder diameter. Because dislocation glide across the grain diameter is unlikely, especially for large $R$, it was concluded that the dislocations must be conserved. Yet the simulations [9], as well as the present work, indicate that the dislocation conservation criterion is violated. What is the mechanism of dislocation annihilation?

While we cannot provide complete answers to these questions, the following considerations may hold a clue. Observations of dislocation movements during the MD simulations revealed very extensive dislocation reactions and their fast propagation along the boundary. The most elementary reactions that we frequently observed were the disassociation of a (100) dislocation in two $1/2\langle 110 \rangle$ dislocations and the recombination of two $1/2\langle 110 \rangle$s into one (100). Comparison of multiple MD snapshots similar to Fig. 5 revealed what at first sight appeared like a rapid migration of dislocations along the GBs. However, such a process is prohibited by the geometry of our system: since the Burgers vectors of all dislocations have a significant $n$-component, they could not simply glide along the boundary. Furthermore, this process was found in simulations at low temperatures and on the nanosecond timescale, ruling out any possibility of dislocation climb.

A closer inspection of the atomic configurations revealed that dislocations do not move over large distances; it is their dissociated/associated state that propagates rapidly by a chain of dislocation reactions. The process is illustrated in Fig. 19 for a single $1/2\langle 110 \rangle$ dislocation propagating through an array of (100) dislocations. At each step, a (100) dislocation from the array dissociates into two $1/2\langle 110 \rangle$s, one of which glides over a short distance (comparable to the dislocation spacing) and recombines with the 1/2(110) on its left to form a new (100). The second product dislocation of the 1/2(110) type then continues to propagate to the right by the same mechanism. This process looks as if a 1/2(110) dislocation has migrated along the boundary (compare the initial a and final b states in Fig. 19). In fact, however, it is only the 1/2(110) Burgers vector that propagates over many steps, whereas each individual dislocation glides only over a short distance. We indicate this process by a dashed arrow as in Fig. 19i.

The example in Fig. 19 demonstrates only one possible case of dislocation propagation. Fig. 20 summarizes other possible cases: (b) propagation of a (100) dislocation through an array of $1/2\langle 110 \rangle$s, (c) a (100) through (100)s and (d) a $1/2\langle 110 \rangle$ through $1/2\langle 110 \rangle$s. Note that, as a result of the propagation process, the initial dislocation array moves slightly up or down, depending on the directions of local glides of the dislocations. Multiple
passes of dislocations can produce greater GB displacements. Such chains of dislocation reactions may constitute the migration mechanism of GBs composed of different dislocation types without climb or formation of dislocation locks.

The proposed mechanism can also be responsible for dislocation annihilation. Instead of crossing the grain through its interior, a dislocation \( \mathbf{b} \) can propagate along the GB until it reaches the opposite side of the grain and recombines with its counterpart \(-\mathbf{b}\). The process is illustrated in Fig. 21. For simplicity, this figure shows an octagonal grain with facets composed of a single type of dislocation. In reality, dislocations with different Burgers vectors can be intermixed (see e.g. Fig. 5) and the grain shape can be different from an octagon and can vary in time. It is believed, however, that the dislocations can still move and annihilate by mechanisms involving combinations of local (short-range) glides and dissociation/recombination reactions.

In the recent PFC study of a two-dimensional circular grain, Wu and Voorhees [36] also observed the motion of dislocations along the GB during the grain shrinkage. Some of the dislocations were observed to pass through each other, encounter other dislocations and eventually combine with them to form a new dislocation. These processes appear to be similar to the dislocation reactions observed in our work. It should be noted, however, that the two-dimensional hexagonal lattice studied in Ref. [36] produces a different dislocation structure of GBs. In particular, the GB dislocations can have six possible Burgers vectors of the same magnitude. In addition, due to the diffusive timescales implemented in the PFC simulations, those dislocations move by a combination of glide and

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**Fig. 19.** Schematic illustration of the dislocation propagation process along a grain boundary by a chain of dislocation reactions. The dislocation notation is the same as in Figs. 4 and 5. The dislocation pairs undergoing dissociation and recombination reactions are encircled. (a) Initial state. (b, c and f) Dissociation-recombination processes. (d and g) Intermediate states. (h) Final state. (i) Notation of the whole process.

**Fig. 20.** Summary of dislocation propagation reactions in grain boundaries composed of two types of dislocations. See Figs. 4 and 5 for dislocation notations.

**Fig. 21.** Illustration of dislocation annihilation by propagation of chains of reactions along the GB. The process involves steps (a–c) corresponding to the propagation reactions in Fig. 20. The annihilation reaction is encircled.
climb. By contrast, the GBs studied in our work are composed of two types of dislocations with different magnitudes of the Burgers vector. This enables the dissociation–recombination reactions described above, which lead to the propagation of the Burgers vector by a chain of dislocations reactions, to occur. In addition, the dislocation processes discussed in this work do not involve dislocation climb and occur at both high and low temperatures.

8.3. What is so special about 36°?

Both previous simulations [9] and this work show that, at low enough temperatures, low-angle grains rotate towards larger angles until the misorientation reaches approximately 36°. For grains with initial misorientations close to 36°, rotation nearly disappears. Furthermore, we find that a grain with $\theta_0 = 44°$ rotates backwards, to smaller angles, and approaches around 37° before disappearing. Thus, it appears that a narrow angular range around 36° plays the role of an “attractor” for grain rotation.

Srinivasan and Cahn [9] suggested that the grains rotate towards the 36.9° angle, corresponding to the Σ5 coincident site lattice (CSL) (Σ being the reciprocal density of CSL sites). They explained the absence of rotation of the Σ5 grain in their simulations by a particular structure of the Σ5 CSL unit cell. However, our simulations show that the Σ5 grain does rotate backwards, albeit by only a small angle (see Fig. 8a and 16). It is also well established that plane Σ5 GBs are coupled over a wide range of temperatures [28].

We propose the following alternate explanation of the 36° angle. It is known that, due to crystal symmetry, the coupling factor $\beta$ is a multi-valued function of crystallographic parameters of the GB [27,28]. For the particular case of [001] symmetrical tilt boundaries, two branches of $\beta(\theta)$ were identified and studied by MD simulations [27,28]: the so-called (100) branch, which starts at $\theta = 0$ and is described by Eq. (24), and a (110) branch, which starts at $\theta = 90°$ and has the coupling factor

$$\beta = -2 \tan \left( \frac{\pi - \theta}{4} \right)$$

(36)

The (100) branch corresponds to positive values of $\beta$ while the (110) branch corresponds to negative values. Thus, for the same direction of coupled GB motion, the (100) and (110) branches predict relative translations of the grains in opposite directions.

Previous MD simulations [27,28] have shown that symmetrical GBs with $\theta$ up to 32° are predominantly coupled in the (100) mode, while GBs with $\theta$ above 37° are predominantly coupled in the (110) mode. There is a jump between the two branches at an angle that was not determined precisely but is somewhere between 32° and 37°. This angular interval corresponds to the so-called “dual” behavior, in which the boundary can start moving in one coupling mode but later switch to another [27,28].

For a cylindrical grain, misorientations corresponding to initial angles $\leq 32°$ and $\geq 37°$ correspond to different signs of $\beta$ and should produce grain rotation in opposite directions. Namely, the grains with $\theta_0 < 32°$ have $\beta > 0$ and should rotate with increasing $\theta$, whereas grains with $\theta_0 > 37°$ have $\beta < 0$ and should rotate with decreasing $\theta$. This is exactly what we found in the MD simulations (Fig. 8a).

In the angular range of “dual” behavior, centered at $\theta_0 \approx 36°$, the grain is in a “frustrated” state of rotation. If the GB initially moves in the (100) mode, the misorientation angle increases and eventually reaches a transition to the (110) mode. Once there, the angle starts to decrease, producing grain rotation in the opposite direction. This eventually leads to a transition back to the (100) mode, and the process repeats. This can lead to small fluctuations of the angle around 36°, but the net rotation of the grain trapped in this “frustration” angular interval should cease to occur.

9. Conclusions and outlook

We have studied the evolution of an isolated cylindrical grain in copper with a set of initial misorientation angles $\theta_0$ around the [001] axis. Although this highly idealistic grain geometry ignores the numerous constraints present in real polycrystalline materials, it is perfect for studying the dynamics and mechanisms of GB motion and grain rotation per se.

We find that at temperatures above $\sim 0.5T_m$ the grain shrinks and eventually disappears. At low temperatures ($\leq 0.8T_m$) the GB motion produces grain rotation with an increasing or decreasing misorientation angle $\theta$, depending on whether $\theta_0$ is below or above $\theta_0 \approx 36°$ (Fig. 8a). Together with previous work [9,36], these observations provide convincing evidence that coupled GB motion can induce grain rotation, including rotation from low-angle to high-angle misorientations.

This coupling-induced grain rotation is observed at temperatures at which diffusion processes are effectively “frozen out”, at least on the timescale of the MD simulations. For example, Fig. 7 gives no indication of diffusion scattering of the labeled atoms out of the marker stripe at 900 K (0.68$T_m$). Thus, the grain rotation studied here is physically different from the rotation controlled by diffusion fluxes along GBs [3,11,12] or by mechanisms involving dislocation climb [3,5,13,14,36]. The reversal of the rotation direction at $\theta_0 \approx 36°$ can be explained by the existence of a discontinuous transition between two coupling modes with opposite signs of the coupling factor. A grain with this angle is “frustrated” by the two conflicting coupling modes and is unable to rotate. This critical angle does not depend on temperature with the statistical errors of our simulations. Future studies will show if and how it may depend on the crystal structure of the material.

The simulations provide significant evidence that the coupling of a shrinking GB is never perfect and is always

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accompanied by some amount of GB sliding. As a result, the dislocation content of the GB is not conserved during its motion. Generally, sliding along a curved GB must alter its dislocation content in order to accommodate the change in $\theta$. Thus the sliding process requires either nucleation or annihilation of dislocations, depending on whether the misorientation angle increases or decreases. It is only for a plane boundary that sliding can occur without nucleation or annihilation of dislocations since the misorientation remains fixed.

That some of the dislocation content was irreversibly lost during the GB motion in all our simulations is evidenced by the rapid decline of the dislocation conservation parameter $C$ (Fig. 16). Dislocation elimination was also directly observed by examining MD snapshots (Fig. 17). Processes that would require nucleation of dislocations were never observed. In other words, the GB sliding was always towards decreasing angles. The mechanism of dislocation annihilation suggested by the analysis of MD configurations is propagation of dislocation content along the GB by a chain of reactions until dislocations with opposite Burgers vectors recombine (Fig. 21). A similar reaction-based mechanism is likely to be responsible for the motion of a curved GB without dislocation climb or formation of locked configurations. Further simulation studies are needed for validation of the proposed mechanism or discovery of alternate mechanisms.

The simulations have shown that, when grain rotation is blocked by imposed constraints, the GB motion slows down. In the absence of grain rotation, the GB motion is accompanied by additional GB sliding acting to cancel the rotation caused by coupling. The retardation of GB migration is an indication of high sliding resistance of the GBs studied in this work. This retardation effect was observed in grains with $\theta_0 = 16^\circ$ and $28^\circ$ which, when free, rotate to larger misorientation angles. When the rotation is blocked, the sliding rotates these grains back to smaller angles, which requires annihilation of GB dislocations.

When coupling disappears at high temperatures, the rotation of free grains stops. Note that the grains with $\theta_0 = 16^\circ$ and $28^\circ$ could reduce their $\gamma$ by rotation to smaller angles by GB sliding. The absence of this rotation in our simulations confirms the large sliding resistance of these boundaries.

Overall, our simulations suggest that the rate-controlling step in the motion of curved GBs is the annihilation of GB dislocations rather than their motion.

The simulations at temperatures approaching the melting point of the material offer the opportunity to probe the effect of GB premelting on grain evolution. While previous studies of GB premelting tested one misorientation at a time, here we have a continuous change of the misorientation angle as the grain rotates. When the initial misorientation angle is small, the GB structure is well ordered and the boundary is coupled, inducing grain rotation to higher angles at and even above $T_m$. As $\theta$ increases, the GB eventually undergoes a premelting transition. As a result of this transition, the coupling disappears and the rotation stops. In the future, this approach could be applied for a systematic study of premelting transitions at GBs in the temperature–angle coordinates.

Many results of our simulations are in agreement with predictions of the Cahn and Taylor model [10]. In particular, in several cases when the model predicts parabolic kinetics, they were indeed found in the simulations (see e.g. Figs. 15 and 18). However, the present simulations have probed only a limited domain in the parameter space of the model. Many interesting predictions of this model await testing by further simulations.

Finally, the present simulations were focused on the initially circular cross-section of the grain. Taylor and Cahn have recently developed a more general model which describes evolution of non-circular cylindrical grains based on an expanded variational approach [58]. MD simulations are in progress to test predictions of this model with respect to the shrinkage kinetics, rotation of the crystal lattice, rotation of the shape of the cross-section, and the relative roles of coupling, sliding and GB diffusion.

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