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Scripta Materialia 63 (2010) 1049–1052
Scripta MATERIALIA
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Analysis of the elastic strain energy driving force for grain boundary migration using phase field simulation

Michael Tonks,[a,*] Paul Millett,[a] Wei Cai[b] and Dieter Wolf[c]

[a] *Fuel Modeling and Simulation, Idaho National Laboratory, P.O. Box 1625, Idaho Falls, ID 83415-3840, USA*
[b] *Department of Mechanical Engineering, Stanford University, Stanford, CA 94305-4040, USA*
[c] *Center for Advanced Modeling and Simulation, Idaho National Laboratory, P.O. Box 1625, Idaho Falls, ID 83415-3840, USA*

Received 10 May 2010; revised 13 July 2010; accepted 23 July 2010
Available online 29 July 2010

We investigate elastic energy-driven grain boundary migration in a strained copper bicrystal using an atomistically informed phase field model. In a bicrystal experiencing a uniform strain, the softer grain has a lower energy density and grows at the expense of the harder grain. In a bicrystal experiencing heterogeneous strain, the softer grain has a higher energy density, yet it still grows. Our findings suggest that the softer grain will grow, irrespective of the difference in the energy densities.
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Keywords: Grain boundary migration; Elastic deformation; Phase field model; Microstructure evolution

Grain boundaries (GBs) migrate due to applied driving forces. According to reaction-rate theory, the GB velocity is typically a linear function of the driving force f_d, i.e. $\mathbf{v} = m f_d \hat{\mathbf{r}}$ with m the GB mobility and $\hat{\mathbf{r}}$ a unit vector normal to the GB [1]. The driving force f_d causes GB migration to lower the Gibbs free energy of the system. The driving force can originate from various sources, including GB curvature and inhomogeneities in the elastic strain energy in elastically anisotropic materials.

In their molecular dynamics (MD) study, Schönfelder et al. [2] investigated the elastic strain energy driving force. In order to obtain values for the GB mobility m at various temperatures, they analyzed GB migration in a copper bicrystal with two flat high-angle twist GBs. Since the GBs had no curvature, a uniform deformation was applied to the simulation cell resulting in a uniform strain to drive GB migration. They measured the GB velocity for various deformation magnitudes and found that the elastic driving force was equal to

$$f_d = -\Delta E_{el} \tag{1}$$

where ΔE_{el} is the difference in the elastic energy density across the boundary.

In this work, we investigate the elastic driving force using a phase field grain growth model. We analyze a copper bicrystal under an applied strain similar to that studied by Schönfelder et al. [2]. Two cases are considered: first, the case in which the strain is uniform throughout the bicrystal, and second, the case in which the strains are heterogeneous but the average strain in the bicrystal is equal to the applied strain. The phase field model is validated by comparing to the results from Schönfelder et al. [2]. Then we investigate the elastic driving force by comparing the GB migration for the two cases.

We simulate GB migration in a two-dimensional (2-D) copper bicrystal similar to the 3-D bicrystal used in the MD simulations from Schönfelder et al. [2]. Our 2-D bicrystal has the GB normal to the x-direction and is homogeneous in the y-direction, while the 3-D bicrystal from [2] has the GB normal to the z-direction and is homogeneous in the x- and y-directions. In both bicrystals, the two grains are oriented at $\pm\theta/2$ about the [0 0 1] axis, where $\theta = 43.60^\circ$ (see the schematic in Fig. 1). Periodic boundary conditions are employed such that our 2-D bicrystal has two flat high-angle (0 0 1) symmetric tilt boundaries, in contrast to the two high-angle (0 0 1) twist boundaries in the 3-D bicrystal from [2].

The bicrystal experiences the strain used by [2],

$$\boldsymbol{\epsilon} = \frac{\epsilon}{2}(\mathbf{e}_1 \otimes \mathbf{e}_1 - \mathbf{e}_2 \otimes \mathbf{e}_2) + \epsilon(\mathbf{e}_1 \otimes \mathbf{e}_2 + \mathbf{e}_2 \otimes \mathbf{e}_1) \tag{2}$$

where ϵ defines the magnitude of the strain, and grain 1 is softer than grain 2 with respect to this applied strain.

* Corresponding author. E-mail: Michael.Tonks@inl.gov

1359-6462/$ - see front matter Published by Elsevier Ltd. on behalf of Acta Materialia Inc.
doi:10.1016/j.scriptamat.2010.07.034

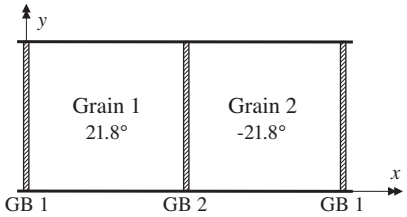

Figure 1. A schematic of our copper bicrystal with two high-angle tilt boundaries, using periodic boundary conditions.

For the uniform strain case, the entire bicrystal is strained according to Eq. (2), resulting in the same energy densities within the grains as in the MD simulations from [2]. For the heterogeneous strain case, the average strain is equal to Eq. (2) but the two grains accommodate the applied strain differently. The *xx* and *xy* components of the strain vary between the two grains, while the *yy* component is homogeneous due to compatibility.

The grain evolution is simulated with the phase field model first presented by Fan and Chen [3], in which each grain is represented by a continuous order parameter ϕ_i equal to one within the grain and zero in all other grains. We also use expressions from Moelans et al. [4] to relate each of the model parameters to the GB surface energy σ, the GB mobility m and the diffuse GB width w_{GB}. In our simulations, we employ the GB mobility and surface energy for pure copper that were determined using MD by Schönfelder et al. [2], i.e. $\sigma = 0.708\ \mathrm{J\,m^{-2}}$ and $m = m_0 \exp\left(\frac{-Q}{k_B T}\right)$, where k_B is the Boltzmann constant, T is the absolute temperature, $m_0 = 2.5 \times 10^{-6}\ \mathrm{m^4\,J^{-1}\,s^{-1}}$ and $Q = 0.23$ eV.

To include the effect of elastic deformation on the GB migration, we add the elastic energy density E_{el} in the phase field free energy functional, i.e.

$$F = \int_V \left(f(\phi_1, \phi_2) + E_{el}\right) dV \tag{3}$$

where $f(\phi_1, \phi_2)$ is the free energy density defined by Moelans et al. [4] and $E_{el} = \frac{1}{2}(\mathscr{C}(\phi_1, \phi_2)\boldsymbol{\epsilon}) \cdot \boldsymbol{\epsilon}$. The elastic stiffness tensor $\mathscr{C}$ is a function of the order parameters defining the two grains according to

$$\mathscr{C}(\phi_1, \phi_2) = \frac{h(\phi_1)\mathscr{C}_1 + h(\phi_2)\mathscr{C}_2}{h(\phi_1) + h(\phi_2)} \tag{4}$$

where $\mathscr{C}_i$ is the elastic tensor from the corresponding grain orientation and the smooth function $h(\phi_i) = (1 + \sin(\pi(\phi_i - 1/2)))/2$. Since copper has a cubic crystal structure, the elastic tensor is fully defined by three temperature-dependent elastic constants [2]

$$\begin{aligned} C_{11} &= 181.50 - .109\,T\ \mathrm{GPa} \\ C_{12} &= 101.80 - .062\,T\ \mathrm{GPa} \\ C_{44} &= 101.80 - .057\,T\ \mathrm{GPa} \end{aligned} \tag{5}$$

For the heterogeneous strain case, the strain throughout the bicrystal is calculated with the spectral stress calculation method from Wang et al. [5]. In this method, the stress equilibrium equation is solved with the fast Fourier transform. The spectral stress calculation is loosely coupled to the phase field model, such that the heterogeneous strain is calculated at the start of a time step using the order parameter values from the previous time step. The calculated strain is then used to calculate the elastic energy density E_{el} for the phase field free energy from Eq. (3).

The order parameters defining each grain are evolved with an Allen–Cahn equation according to

$$\frac{\partial \phi_i}{\partial t} = -L\frac{\partial F}{\partial \phi_i} = -L\frac{\partial f}{\partial \phi_i} - \frac{L}{2}\left(\frac{\partial \mathscr{C}}{\partial \phi_i}\boldsymbol{\epsilon}\right) \cdot \boldsymbol{\epsilon} \tag{6}$$

where the order parameter mobility $L = \frac{4m}{3w_{GB}}$ [4]. The bicrystal is represented as a 1.28 μm × 0.64 μm rectangle. The system is solved using finite difference with a 10 nm grid spacing and backward Euler time integration. A diffuse GB width of 4.5 grid spacings is used, since it was found to be the smallest value that did not alter the order parameter evolution.

To investigate the behavior of the copper bicrystal, the GB migration is simulated at $T = 800$ K for different values of the applied strain magnitude ϵ. We first validate the model by comparing the behavior from the uniform strain case to the MD results from Schönfelder et al. [2]. The GBs migrate such that grain 1 grows linearly with time at the expense of grain 2. We determine GB velocity from the slope of plots of the *x*-position of the GB centers with time (see Fig. 2a for an example). This calculation is repeated to calculate the GB velocity for each value of ϵ. The velocities calculated with our phase field model compare well with Eq. (1) from the MD results, as shown in Figure 2b. Our results also show that grain 1 has a lower elastic energy density than grain 2 (see Fig. 3a).

For the heterogeneous strain case, grain 1 has a higher elastic energy density (Fig. 3b). This occurs because grain 2 is stiffer and therefore accommodates less of the strain, resulting in a lower energy density. The strain distribution is uniform in both grains and changes smoothly but rapidly over the GB width. In spite of having a higher energy density, grain 1 still grows at the expense of grain 2 for the heterogeneous strain case. As grain 1 grows, the energy density in both grains decreases, as does the energy density difference over the boundary. The GB *x*-positions vary linearly with time and, again, we use the slope to determine the GB velocity (see Fig. 4a for an example). The GB velocity varies

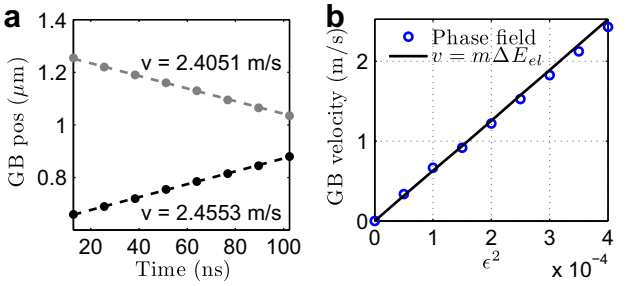

Figure 2. GB migration in a copper bicrystal for the uniform strain case, with (a) a plot of the GB *x*-position at various times for $\epsilon = 0.02$, where the slope is the GB velocity, and (b) the GB velocity from the phase field simulations for various values of ϵ^2, showing excellent agreement with the results from Schönfelder et al. [2].

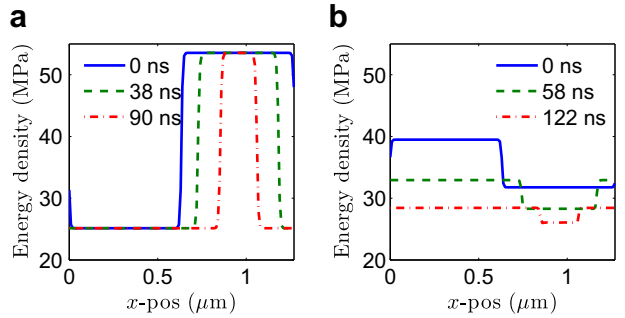

Figure 3. The elastic energy density along the bicrystal with $\epsilon = 0.02$ at several times during the GB migration for (a) the uniform strain case and (b) the heterogeneous strain case. The energy density is higher in grain 2 for the uniform strain case but lower for the heterogeneous strain case.

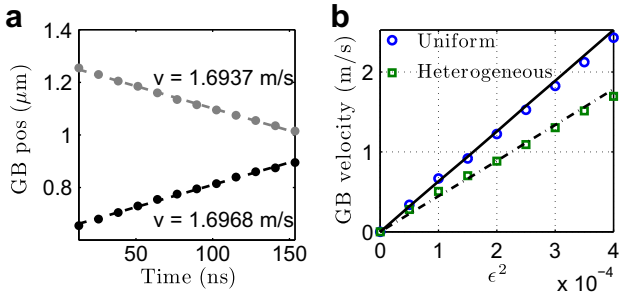

Figure 4. GB migration for the heterogeneous strain case, with (a) a plot of the GB x-position vs time for $\epsilon = 0.02$ and (b) the GB velocity for various values of ϵ, where the uniform strain case is shown for reference. The dotted line is a linear relationship between GB velocity and ϵ^2 with the slope a factor of $\sqrt{2}$ lower than for the uniform strain case.

linearly with ϵ^2, as in the uniform strain case; however, the linear slope is a factor of $\sqrt{2}$ lower (Fig. 4b).

The total free energy in the bicrystal decreases with time for both strain cases, reaching the same steady-state value once only grain 1 remains (see Fig. 5 for the total free energy with time for $\epsilon = 0.02$). The total energy decreases in the uniform strain case because the grain with the lower free energy is growing; it decreases in the heterogeneous strain case because the energy density in both grains decreases with time.

In our phase field simulations, the softer grain (grain 1) grows for both strain cases, in spite of its higher energy density in the heterogeneous strain case. This

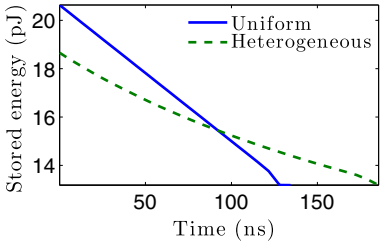

Figure 5. The total stored elastic energy with time in a bicrystal strained with $\epsilon = 0.02$. Values for the stored energy are shown until only grain 1 remains. The energy decreases with time for both cases and both reach the same final energy.

behavior is not consistent with the driving force described by Eq. (1).

To verify our finding that grain 1 grows in the heterogeneous strain case, though it has a higher energy density, we consider a simplified model of the bicrystal GB migration. We consider two linear elastic rods connected in series that are deformed some distance x. The two rods have lengths l_1 and l_2, which are initially equal to $l/2$, where l is the total length. Both rods have an equal cross-sectional area A and Young's moduli $E_2 > E_1$. From the expression for the elastic rod stiffness $k_i = E_i A/l_i$, we know that $k_2 > k_1$. When deformed, the stiffer rod (rod 2) deforms less than rod 1 and therefore stores less energy, i.e. $U_1/U_2 = k_2/k_1$.

We assume that one rod grows at the expense of the other ($l_1 = l - l_2$) to minimize the total enthalpy in the system, such that $\dot{l}_1 = -\dot{l}_2 = -M\,\partial H/\partial l_1$, where the total enthalpy

$$H = \frac{1}{2}k_1 x_1^2 + \frac{1}{2}k_1 x_1^2 \tag{7}$$

$$= \frac{1}{2}\frac{AE_1E_2x^2}{E_1(l-l_1)+E_2l_1} \tag{8}$$

By substituting Eq. (8) into the l_1 evolution equation, we obtain

$$\dot{l}_1 = \frac{M}{2}\frac{AE_1^2E_2(E_2-E_1)x^2}{(E_2l_1+E_1l_2)^2} \tag{9}$$

Eq. (9) indicates that rod 1 will grow, though it stores more energy. However, since the softer rod grows, the effective stiffness of the two rods will decrease, causing the total stored energy to decrease. This finding is consistent with the results from our phase field simulation.

To better understand the direction in which the elastic energy drives the GB to migrate, we analyze the elastic energy term from Eq. (6). For grain 1, $\partial E_{el}/\partial\phi_1 = 1/2\ (\partial\mathscr{C}/\partial\phi_1\boldsymbol{\epsilon})\cdot\boldsymbol{\epsilon}$, where

$$\frac{\partial\mathscr{C}}{\partial\phi_1} = \frac{\partial h(\phi_1)}{\partial\phi_1}\frac{h(\phi_2)}{(h(\phi_1)+h(\phi_2))^2}(\mathscr{C}_1-\mathscr{C}_2)$$

$$= \tilde{f}(\phi_1,\phi_2)(\mathscr{C}_1-\mathscr{C}_2) \tag{10}$$

Therefore, whether ϕ_1 will increase or decrease is determined by the expression

$$\frac{\partial\phi_1}{\partial t} \sim -((\mathscr{C}_1-\mathscr{C}_2)\boldsymbol{\epsilon})\cdot\boldsymbol{\epsilon} \tag{11}$$

From this expression, it appears that the direction in which the elastic energy drives the GB migration is determined by the difference in the elastic constants rather than the difference in the energy densities. This migration direction reduces the total free energy in the system because, due to the coupled nature of the strain experienced by the two grains, the transfer of atoms across the boundary decreases the energy density in the harder grain more than it increases the energy density in the softer grain. For the specific case in which the strain is uniform throughout the body, the softer grain also has a lower energy density, and therefore the migration follows Eq. (1).

We simulated GB migration in a copper bicrystal with a phase field model coupled to a spectral stress

calculation method. Using material parameters measured from MD simulations by Schönfelder et al. [2], we found that when the bicrystal experiences a uniform strain, the softer grain has a lower energy density and grows at the expense of the harder grain. However, when the grains have heterogeneous strains, the softer grain has a higher energy density, yet it still grows. This finding is consistent with a simplified analysis of two elastic rods connected in series. Therefore, it appears that the softer grain with respect to the applied strain will grow irrespective of the difference in the energy densities.

The authors wish to thank Anter El-Azab of Florida State University for useful discussions on how to include the effects of stress in phase field models. This work was supported by the US Department of Energy, Office of Nuclear Energy, Advanced Modeling and Simulation program. This manuscript has been authored by Battelle Energy Alliance, LLC under Contract No. DE-AC07-05ID14517 with the US Department of Energy (INL/JOU-10-18575).

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