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Mesoscale Simulation of the Evolution of the Grain Boundary Character Distribution

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Abstract. A mesoscale, variational simulation of grain growth in two-dimensions has been used to explore the effects of grain boundary properties on the grain boundary character distribution. Anisotropy in the grain boundary energy has a stronger influence on the grain boundary character distribution than anisotropy in the grain boundary mobility. As grain growth proceeds from an initially random distribution, the grain boundary character distribution reaches a steady state that depends on the grain boundary energy. If the energy depends only on the lattice misorientation, then the population and energy are related by the Boltzmann distribution. When the energy depends on both lattice misorientation and boundary orientation, the steady state grain boundary character distribution is more complex and depends on both the energy and changes in the gradient of the energy with respect to orientation.

Introduction

With the availability of reliable large scale simulations of materials behavior, a principal challenge is the development of strategies for the extraction of information. For example, relative area histograms are stable statistics of bona fide simulations, but tend to discriminate poorly among input grain boundary energy functions [1]. Here, we use our variational approach to the mesoscale simulation of grain growth in large two dimensional systems to study the evolution of the grain boundary character distribution. Like relative area histograms, histograms of the grain boundary character distribution are stable during grain growth. Unlike the relative area histograms, they are sensitive to the assumed grain boundary energy anisotropy.

In the two dimensional systems described here, the grain boundary character distribution, $f(\theta,\phi)$, is the relative length of grain boundary with respect to lattice misorientation (θ) and boundary orientation (ϕ). More precisely, $f(\theta,\phi)$ is the length of arc with lattice misorientation θ and boundary orientation ϕ divided by the total length of grain boundary in the configuration. Recent experimental studies have shown that the grain boundary character distribution is inversely correlated to the grain boundary energy [2,3]. Furthermore, the same low index and slow growing surface orientations that dominate the external forms of the materials are also the preferred grain boundary plane orientations [4-6]. Consistent with these observations, the results presented here show that during grain growth, low energy boundaries tend to populate the histogram more than higher energy boundaries. Furthermore, the simulations show that in the long term, energetic effects tend to dominate over kinetic effects.

Generally speaking, to understand a simulation requires a model or theory of it, as rigorous as possible. The feasibility of predicting relative area-type histograms by deriving master equations that accurately portray behavior of the simulation is an example of this [7]. Here we explore the challenge of extending these ideas to predict the grain boundary character distribution.

Essentials of the simulation

The thermodynamic theory of our simulation consists of the Mullins Equations, a system of evolution parabolic partial differential equations for each grain boundary curve. In two dimensions, grain boundaries meet at triple junctions, where an additional condition is required. We enforce the Herring force balance condition, which is the natural boundary condition for equilibrium, at these triple junctions. The resulting system is then dissipative for the energy [8]. The evolution of grain boundaries using the above model is thus viewed as a modified steepest descent method for the total grain boundary energy functional, where natural boundary conditions are imposed: this is the variational approach. This observation serves as the basis for our discretization method, and leads to stable semi-discrete schemes. In addition to using the gradient of the above functional to determine the grain boundary evolution we must consider certain critical events. During evolution, a grain boundary or a grain may shrink and disappear. This creates multiple junctions which are unstable. Such multiple junctions split into triple junctions in a way that is consistent with energy reduction. In summary, our implementation results in a globally dissipative system for grain growth of a polycrystalline network. Consider a collection of grain boundaries, which are curves in the plane, meeting at triple junctions. The energy per unit length of a curve Γ: $x = \Gamma(s)$, $0 \le s \le s_0$, is given by $\gamma(\Gamma,\Theta)$, where Γ denotes the angle of the normal to the boundary represented by Γ with respect to a reference axis and Θ denotes the lattice misorientation across Γ. The curve evolves according to the equation

$$v_n = \mu\left[\frac{\partial^2 \gamma}{\partial \Gamma^2} + \gamma\right]\kappa \quad \text{on} \quad \Gamma \quad \text{(Mullins Equation)} \qquad (1)$$

$$\sum \left[\frac{\partial \gamma}{\partial \Gamma} n + \gamma b\right] = 0 \quad \text{at each TJ,} \quad \text{(Herring Condition)}$$

where v_n denotes the normal velocity of Γ, μ the mobility, κ the curvature, n the normal, and b the tangent of Γ. The sum is taken over the three grain boundaries meeting at each triple junction at the endpoints of Γ. For the configuration

$$\Gamma = \{\ \Gamma_1, \ldots, \Gamma_K\ \},\ K = K(t),\ t = \text{time}$$

the total energy is given by

$$E(t) = \sum_{\Gamma} \int_{\Gamma} \gamma(\Gamma,\Theta)\,|\,b\,|\,ds \quad \text{and} \quad \frac{dE(t)}{dt} \le 0,$$

since, as we have noted above, the system is dissipative.

Energy depending only on lattice misorientation

First consider a situation where we assume that $\gamma = \gamma(\theta)$, that is, the energy per unit length depends on lattice misorientation alone. Our objective is to discuss the histogram of the grain boundary character distribution, which we define here as the relative arc length corresponding to a given θ, versus θ. Here we may write a very simple formula for $E(t)$. Let $f(\theta,t)$ denote the relative length of arc with misorientation θ at time t. Then

$$E(t) = \sum_a \gamma(\theta) f(\theta,t) \cdot l(t), \qquad l(t) = \text{total length of boundary at time } t,$$

or assuming for convenience that θ varies continuously, we write simply, assuming cubic symmetry,

$$E(t) = \int_I \gamma(\theta) f(\theta,t) d\theta \cdot l(t), \qquad I = [-\pi/4,\pi/4].$$

Our observation is that after some fairly short time, $f(\theta,t)$ assumes a self-similar form, say $f(\theta,t) = f(\theta)$. Also, as a consequence of the 'parabolic growth law' for average grain size, which is obeyed by the simulation, $l(t)$ varies inversely as the square root of t. Hence,

$$E(t) = \int_I \gamma(\theta) f(\theta) d\theta \cdot \frac{c}{\sqrt{a+bt}} \tag{2}$$

In this situation, we find that

$$f(\theta) = \frac{1}{Z} e^{\beta\gamma(\theta)/\gamma}, \quad \theta \in I,$$

a Boltzmann distribution, as illustrated in Fig. 1 for

$$\gamma(\theta) = 1 + \varepsilon(\sin 2\theta)^2, \quad \varepsilon = 0.125 \tag{3}$$

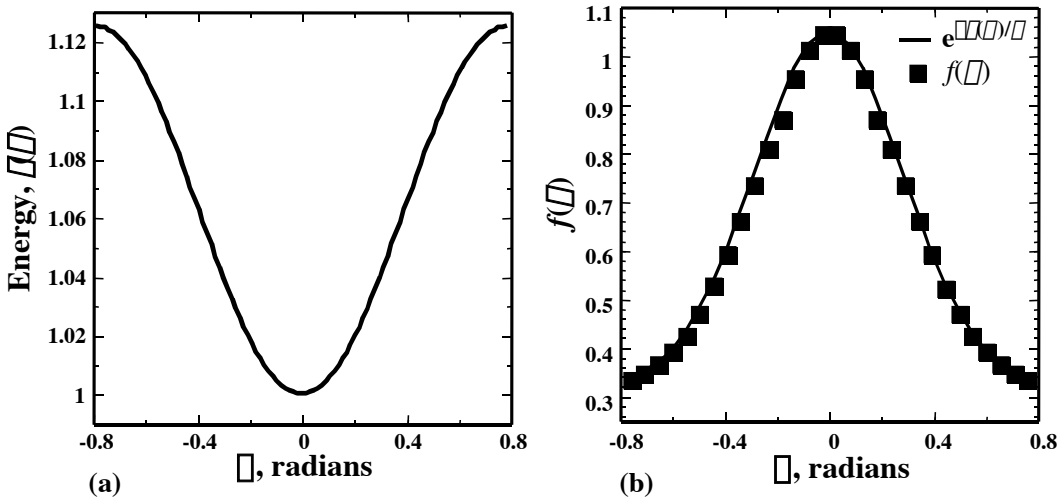

Figure 1. (a) Energy per unit length, $\gamma(\theta)$, Eq. (3) and (b) the grain boundary character distribution histogram and the fitted Boltzmann distribution.

We have chosen this particular form of $\gamma(\theta)$ for clarity, but the result holds for all assumed energy anisotropies that we simulated. We are able give an interpretation by regarding the collection

{□(t)} of configurations at all times in the simulation as an ensemble and (2) as a stable statistic, or its internal energy.

Energy depending on lattice misorientation and normal

A much different situation prevails when the energy □(□,□) varies both with lattice misorientation and normal inclination. To formulate this, we assume the interfacial energy to be the sum of two orientation "surface" energies, □(□), where □ is a parameter measured with respect the axes of each grain. This form was developed in analogy to the observation that to a first order approximation, the grain boundary energy is proportional to the sum of the energies of the surfaces adjacent to the boundary [9]. Given an arc □ separating two grains with orientations □ and □*, and a free surface energy □, let

□(□,□) = □(□□□□) + □(□□□□*) = □(□□□□) + □(□□□□□□).

Two hypothetical energy functions are illustrated in Figures 2a and 3a.

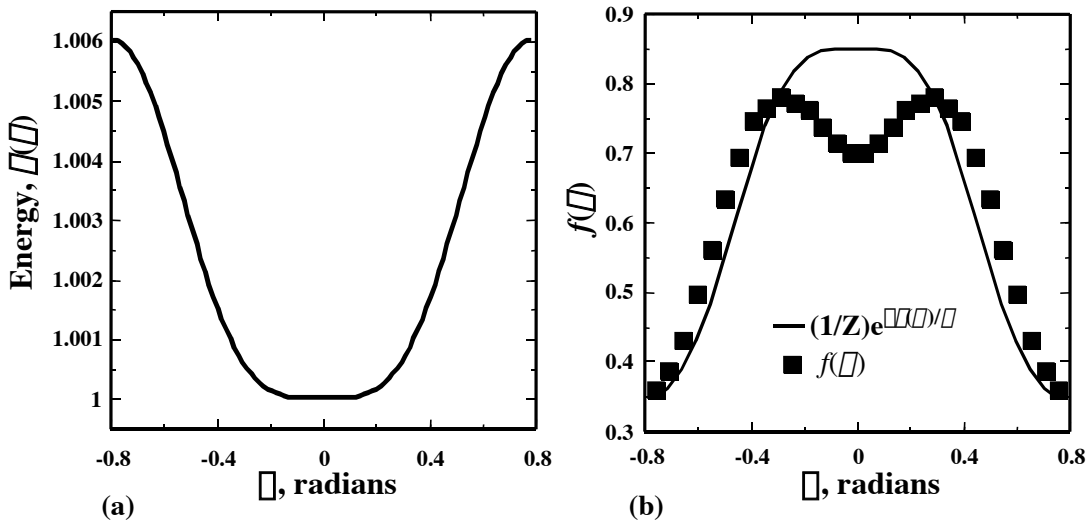

Figure 2. (a) Energy per unit length, □(□) = 1 + □(*sin 2*□)^4, □= 0.006 and (b) and the corresponding grain boundary character distribution (squares), and Boltzmann distribution (line).

We form a histogram of the arc-length corresponding to the collection of numbers □ = □□□, □ − □*, where the orientations □, □* are assigned randomly at the beginning of the simulation. Two illustrations are provided in Figs. 2b and 3b, which show the grain boundary character distribution that develops. Figs. 2 and 3 also show that second derivatives of the energy are involved in the relative arc-length distributions. We do not yet have a complete theory for this.

Discussion and summary

The mechanisms by which a grain boundary character distribution develops from an initially random boundary population are not yet fully understood. One possible mechanism might be connected to the kinetic properties of the boundaries. High mobility boundaries have, on average, greater velocities and will be preferentially annihilated during neighbor switching and grain collapse events. Additional simulations, carried out with anisotropic grain boundary mobilities,

showed that the mobility has a subordinate influence on the development of the grain boundary character distribution. Only when the mobilities varied by more than an order of magnitude was it possible to detect an influence on the distribution. In other words, much smaller changes in the energy have a much larger influence on the distribution.

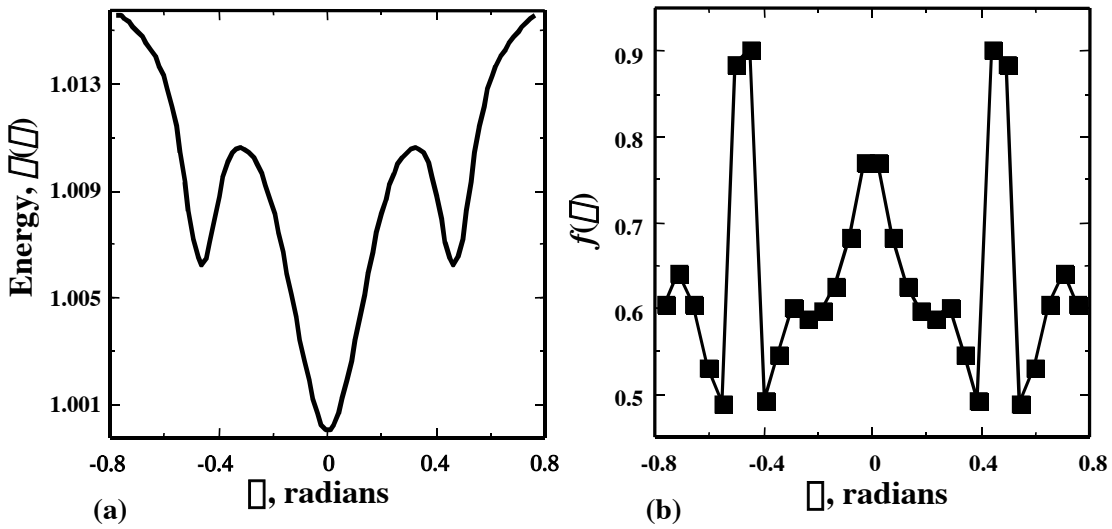

Figure 3. (a) Plot of the energy per unit length $\gamma(\theta)$, a cubic spline, with 1.5% anisotropy and (b) the corresponding grain boundary character distribution.

Earlier simulations using phase field and Monte Carlo techniques have also shown that the grain boundary character distribution is influenced more by grain boundary energies than by mobilities [10, 11]. The assumed energy anisotropies used in the prior studies depended only on the lattice misorientation and showed that boundaries with relatively low energies accumulate during the course of grain growth. The results presented here are consistent with the earlier results and we have further shown that the steady state population can be related to the energy through the Boltzmann distribution. However, this is true only in the case where the energy depends on the lattice misorientation and is not thought to be representative of the real situation, where the energy varies with respect to the orientation of the boundary plane. In fact, in the one case where the five parameter grain boundary character distribution was compared to the measured grain boundary energy in a real material, a Boltzmann distribution was not observed [9].

The simulations demonstrate that when the energy also depends on the grain boundary plane, the grain boundary character distribution is not a Boltzmann distribution. Two distinct processes can alter the distribution as grain growth occurs. The first is the constant boundary annihilation and creation that occurs as a result of neighbor switching and grain collapse. This process alters both the distribution of lattice misorientations and the distribution of grain boundary orientations. The second process has to do with changes in boundary orientation. Such rotations affect only the distribution of orientations while preserving the distribution of lattice misorientations. If there in anisotropy in the space of grain boundary planes, then a boundary will tend to reorient itself in a low energy configuration, subject to the constraint of equilibrium at the triple junction. Note that if the boundary energy depends only on the lattice misorientation, these rotations will not occur. In other words, the differences in the distribution that develop when grain boundary orientation anisotropy are added to the simulation are the result of this second process.

The reorientation events that change the boundary character without changing the lattice misorientation are most likely to occur at the triple junctions, where boundaries are free to rotate to achieve the equilibrium specified by the Herring condition. We can use our simulation to test this idea. One of the advantages of our simulation method is that it is easy to change the boundary conditions and equations of motion. When a simulation was run with anisotropic grain boundary energies, but also with the boundary condition that all triple junction dihedral angles be 2π/3 (the condition for isotropy), the grain boundary character distribution remained isotropic. From this result, we conclude that it is local adjustments at the triple junctions that lead to the anisotropy in the steady state grain boundary character distribution.

Finally, it is worth noting that the dimensionality of the system may be influential in determining the steady state grain boundary character distribution. Here, boundaries are characterized by only two parameters: lattice misorientation and boundary orientation. In three dimensional systems, there are five independent parameters and the average number of neighbors each grain has increases by more than a factor of two. Therefore, we are currently implementing a three-dimensional version of this simulation to study the evolution of the grain boundary character distribution with a more realistic model. If these studies also show that an initially random distribution evolves to a steady state that depends on the grain boundary energy anisotropy, then it may be possible to determine grain boundary energies from observations of the grain boundary character distribution.

The results presented here show that during two dimensional grain growth, the grain boundary character distribution that develops from an initially random distribution depend on the energy. The steady state distribution that develops when the energy varies with lattice misorientation and boundary plane orientation is distinct from that which develops when the energy depends only on lattice misorientation. Grain boundary mobility plays a subordinate role in the evolution of the grain boundary character distribution.

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