PREDICTIVE THEORY FOR THE GRAIN BOUNDARY CHARACTER DISTRIBUTION

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1. Introduction

Mesoscale experiment and simulation permit harvesting information about both geometric features and texture in material microstructures. The grain boundary character distribution (GBCD) is an empirical distribution of the relative length (in 2D) or area (in 3D) of interface with a given lattice misorientation and grain boundary normal. During the growth process, an initially random texture distribution reaches a steady state that is strongly correlated to the interfacial energy density [7]. In simulation, it is found that if the given energy depends only on lattice misorientation, then the steady state GBCD and the energy are related by a Boltzmann distribution. This is among the simplest non-random distributions, corresponding to independent trials with respect to the energy. Why does such a simple distribution arise from such a complex system?

We outline a new entropy based theory which suggests that the evolution of the GBCD satisfies a Fokker-Planck Equation. Coarsening in polycrystalline systems is a complicated process involving details of material structures, chemistry, arrangement of grains in the configuration, and environment. In this context, we consider just two global features: cell growth according to a local evolution law and space filling constraints. The growth process may be a gradient flow or curvature driven growth. We shall impose the Mullins Equation of curvature driven growth. Space filling requirements are managed by critical events, rearrangements of the network involving deletion of small contracting cells and facets. The interaction between the evolution law and the constraints is governed primarily by the Herring Condition, the boundary condition associated with the equation of curvature driven growth. It determines a dissipation relation. To assist in the derivation, a simplified model is introduced which is driven by the boundary conditions and reflects the dissipation relation of the grain growth system. It resembles an ensemble of inertia-free

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spring-mass-dashpots. For this simpler coarsening network, we learn how entropic or diffusive behavior at the large scale emerges from a dissipation relation at the scale of local evolution. We offer a simple application of the method to the solution of the Fokker-Planck Equation itself. We present evidence that the theory predicts the results of large scale 2D simulations [1]. It is consistent with experiment. We give a further illustration with the Read-Shockley Energy. Ongoing work involves analysis of 3D simulation and incorporation of normal dependence.

2. View of the problem

2.1. **Reprise of mesoscale theory.** We confine our attention to a two dimensional structure and interfacial energy densities which depend only on lattice misorientation. We describe the dissipation relation satisfied by this system. Suppose two grains are separated by an arc Γ with misorientation α, normal $n = (\cos\theta, \sin\theta)$, tangent direction b and curvature κ. Let $\psi = \psi(\alpha)$ denote the energy density on Γ, which we take to be a function of α alone. Let v_n denote the normal velocity of Γ, and asume the mobility $\mu = 1$. The Mullins Equation of evolution is

$$v_n = \psi\kappa \text{ on } \Gamma. \tag{2.1}$$

Assume that only triple junctions are stable and that the Herring Condition holds

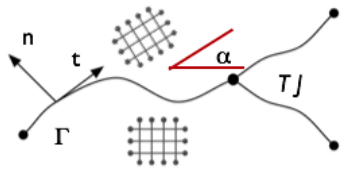

FIGURE 1. An arc Γ with normal n, tangent t, and lattice misorientation α, illustrating lattice elements.

at triple junctions. This means that whenever three curves $\{\Gamma^{(1)}, \Gamma^{(2)}, \Gamma^{(3)}\}$, meet at a point p the force balance, (2.2) below, also known as Young's Law for this case, holds:

$$\sum_{i=1,..,3} \psi b^{(i)} = 0, \tag{2.2}$$

2.2. **Dissipation relation for a network.** We turn now to a network of grains bounded by $\{\Gamma_i\}$ subject to some condition at the border of the region they occupy, like fixed end points or periodicity. The total energy of the system is given by

$$E(t) = \sum_{\{\Gamma_i\}} \int_{\Gamma_i} \psi|b|ds \tag{2.3}$$

Owing exactly to the Herring Condition (2.2), the instantaneous rate of change of the energy is

$$\frac{d}{dt}E(t) = -\sum_{\{\Gamma_i\}} \int_{\Gamma_i} v_n^2 ds + \sum_{TJ} v \cdot \sum \psi b = -\sum_{\{\Gamma_i\}} \int_{\Gamma_i} v_n^2 ds \leqq 0, \tag{2.4}$$

rendering the network dissipative for the energy in any instant absent of critical events. Indeed, in an interval $(t_0, t_0+\tau)$ where there are no critical events, we may integrate (2.4) to obtain a local dissipation equation

$$\sum_{\{\Gamma_i\}} \int_{t_0}^{t_0+\tau} \int_{\Gamma_i} v_n^2 ds dt + E(t_0+\tau) = E(t_0) \tag{2.5}$$

which bears a strong resemblance to the simple dissipation relation for an ensemble of inertia free springs with friction. It is complicated, however, because the first term in (2.5) is not the sum of velocities of elementary 'elements'.

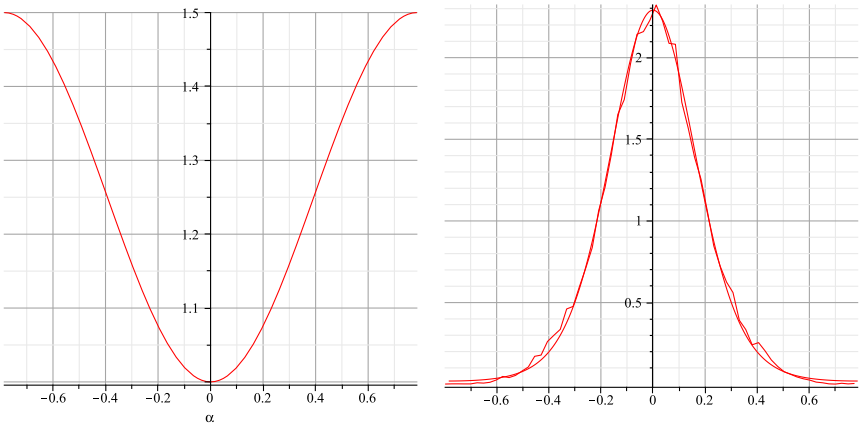

FIGURE 2. (left) The energy density $\psi(\alpha) = 1 + \epsilon \sin^2 2\alpha$, $\epsilon = \frac{1}{2}$, used for the examples in this note. (right) GBCD and Boltzmann distribution for the potential ψ with parameter $\sigma \approx 0.1$ as predicted by our theory. This GBCD is averaged over 5 trials.

3. Entropy and rearrangement events

3.1. **A role for rearrangement events.** To begin we define the grain boundary character distribution, GBCD,

$$\begin{aligned} &\rho(\alpha, t) = \text{relative length of arc of misorientation } \ \alpha \ \text{ at time } t, \\ &\text{normalized so that } \int_\Omega \rho d\alpha = 1. \end{aligned} \tag{3.1}$$

The GBCD is a derived statistic, that is, it is not among the variables being resolved directly in simulation. These are the positions, curvatures, and velocities of the curves which constitute the configuration. The GBCD is harvested from this data. Thus it is a consequence of upscaling a finer level process. In addition to this, the coarsening process itself is irreversible. Once the system has rearranged following the deletion of a cell or a facet, its history is lost and cannot evolve backwards. For these reasons, it is natural to think that entropy may play a role in the description of GBCD evolution. Rearrangement events themselves play an important role in this process. Consider, for example, cells with 5 facets. A given cell with 5 facets has decreasing area according to well known von Neumann-Mullins $n-6$-rule, [10], [8]. However, for example, there is both experimental and computational evidence

that the relative area of 5-sided cells is increasing. We deduce that the population of 5-sided cells at a given time consists of cells which had $6, 7, 8 \dots$ facets at earlier times. Thus in the network setting, the rearrangement events play a major role. Although we may be reasonably confident that small cells with small numbers of facets will be deleted, their effect on the configuration is essentially random. A significant difficulty in developing a theory of the GBCD, and understanding texture development in general, lies in the lack of understanding of the relationship between these stochastic or critical or rearrangement events and the configuration. This leads us to study a technically simpler coarsening model.

3.2. **A simplified model.** The simplified model is a one dimensional coarsening process, a gradient flow, with a dissipation inequality amenable to upscaling to the misorientation ensemble α, [3],[2] . In passing to the higher level ensemble, an entropic contribution

$$+\int_\Omega \rho \log \rho d\alpha, \tag{3.2}$$

appears. It is is minus the usual physical entropy. Minimizing (3.2) favors the uniform state, which would be our situation were $\psi(\alpha)$ =constant. The new extended dissipation relation gives rise to an iterative implicit scheme involving the Monge-Kantorovich-Wasserstein metric. The solution of this scheme has been established, [6], to be the solution of the Fokker-Planck Equation

$$\mu \frac{\partial \rho}{\partial t} = \frac{\partial}{\partial \alpha}(\lambda \frac{\partial \rho}{\partial \alpha} + \psi' \rho) \text{ in } \Omega,\ 0 < t < \infty, \tag{3.3}$$

where $\mu > 0$ is a constant, with, in this situation, periodic boundary conditions. There is presently an extensive literature about MKW-implicit schemes, eg. [9]. We do not know if our statistic $\rho(\alpha, t)$ is a solution of (3.3) but we may ask if characterizations of solutions have desirable properties which are also shared by ρ.

3.3. **Relative entropy and validation of the scheme.** The procedure which leads to the implicit scheme is based on a dissipation inequality, like (2.5), which holds for the entire configuration but does not identify individual intermediate level 'spring-mass-dashpots'. The consequence is that we cannot set the temperature-like parameter σ, but in some way must decide if one exists.

To investigate this, let us introduce the Boltzmann distribution and relative entropy

$$\rho_\lambda(\alpha) = \frac{1}{Z_\lambda} e^{-\frac{\psi(\alpha)}{\lambda}},\ \text{where } Z_\lambda = \int_\Omega e^{-\frac{\psi(\alpha)}{\lambda}} d\alpha,\ \text{and } \Phi_\lambda = \int_\Omega \rho \log \frac{\rho}{\rho_\lambda} d\alpha \tag{3.4}$$

By Jensen's Inequality, $\Phi_\lambda \geqq 0$. If ρ is a solution of (3.3), then

$$\Phi_\lambda \to 0 \text{ as } t \to \infty,\ \text{exponentially fast.} \tag{3.5}$$

We therefore seek to identify the particular $\lambda = \sigma$ for which Φ_σ defined by the GBCD statistic ρ tends monotonely to the minimum of all the $\{\Phi_\lambda\}$ as t becomes large. We then ask if the terminal, or equilibrium, empirical distribution ρ is equal to ρ_σ. For our purposes, we simply decide the question of equality by inspection.

To understand how this is implemented, we offer an illustration using the solution of the (3.3) itself, $u(\alpha, t)$ of with the choice $\lambda = \sigma = 0.1$, and a collection of relative entropy plots $\{\Phi_\lambda\}$ where values of λ are close to σ, cf. Figure 3(left). The plot of Φ_σ vs. time t is noted in red and it is decreasing and tends to 0. A glance at the

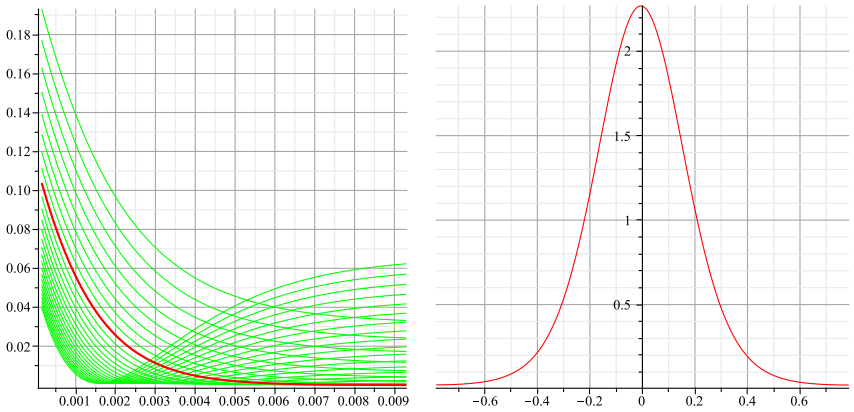

FIGURE 3. (left) The relative entropy of the solution $u(\alpha, t)$ of the Fokker-Planck Equation (3.3) with the choice $\lambda = \sigma = 0.1$, computed by a routine numerical method, compared with a sequence of Φ_λ with the curve for $\sigma = 0.1$ noted in red. (right) The computed equlibrium solution, which is indistinguishable from ρ_σ, the Boltzmann distribution of (3.4).

resulting equilibrium u, Figure 3(right), identifies it as the Boltzmann distribution ρ_σ, as constructed.

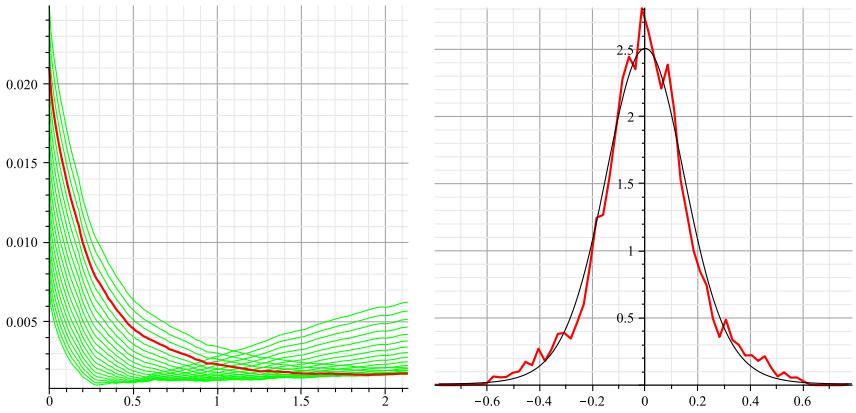

FIGURE 4. (left) The relative entropy of the grain growth simulation for a sequence of λ with the optimal choice $\sigma \approx 0.1$ noted in black. (right) Comparison of the empirical distribution at time $t = 2$, when 0.8 of the cells have been deleted, with ρ_σ, the Boltzmann distribution of (3.4).

We now engage in the identical procedure for the GBCD derived from the simulation data. We identify σ as the value of λ for which Φ_λ is decreasing and tends to a minimum, as shown in Figure 4(left). We then compare the empirical distribution at time $t = 2$ with the Boltzmann distribution ρ_σ, as shown in Figure 4(right). There is good agreement. Averaging over several trials, five in this case, there is excellent agreement, Figure 2(right). This constitutes the validation of our theory.

4. A Read-Shockley example

Executing a simulation with a Read-Shockley energy is a test of the effectiveness of the algorithm for energy densities which are not continuously differentiable. We

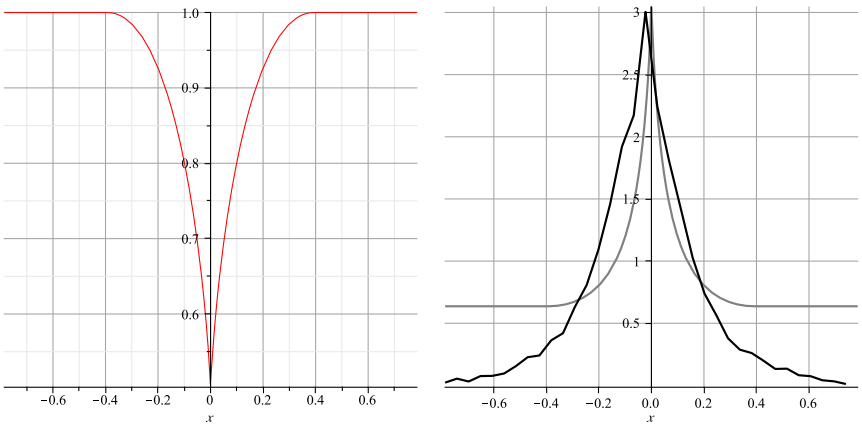

FIGURE 5. (left) A version of a Read-Shockley energy density. (right) Computed near stationary distribution (black) compared with a Boltzmann distribution (gray) of the energy density.

report the results of a first series of simulations in Figure 5. The two-dimensional simulation shows the result of a completely energy driven GBCD evolution, whereas the MacKenzie results in [4],[5] are better interpreted as a perturbation of the random distribution in favor of the energy. We shall continue to explore this situation.

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