Mesoscale simulation of grain growth

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Abstract. Simulation is becoming an increasingly important tool, not only in materials science in a general way, but in the study of grain growth in particular. Here we exhibit a consistent variational approach to the mesoscale simulation of large systems of grain boundaries subject to Mullins Equation of curvature driven growth. Simulations must be accurate and at a scale large enough to have statistical significance. Moreover, they must be sufficiently flexible to use very general energies and mobilities. We introduce this theory and its discretization as a dissipative system in two and three dimensions. The approach has several interesting features. It consists in solving very large systems of nonlinear evolution equations with nonlinear boundary conditions at triple points or on triple lines. Critical events, the disappearance of grains and and the disappearance or exhange of edges, must be accomodated. The data structure is curves in two dimensions and surfaces in three dimensions. We discuss some consequences and challenges, including some ideas about coarse graining the simulation.

Introduction

We discuss the mesoscale simulation of large networks of grains or interfaces in two and three dimensions. We give a brief introduction to the format and explain our algorithm. Evolution is governed by the Mullins Equations of curvature driven growth, discussed below, which consist of a system of evolution partial differential equations for each boundary curve, in two dimensions, or facet, in three dimensions. Grain boundaries typically meet at triple junctions, in two dimensions, or on triple lines, in three dimensions, where a boundary condition is required. Here we enforce the Herring Condition, a force balance. This is the natural boundary conditon for equilibrium of the

Mullins Equation, a fact that may not be well known. The resulting system is dissipative for the energy and the coarsening process may be viewed as a modified steepest descent for the total grain boundary energy. Certain critical events, such as grain disappearance and the exchange or disappearance of facets, must be accomodated. We describe our strategy for this which conserves the dissipative character of the process. A special feature of our approach is that the data structure consists only of curves, in two dimensions, and surfaces in three dimensions, which offers an opportunity to work with large systems. It offers the opportunity to employ experimentally derived energy densities and mobilities. Initial configurations may have statistically representative properties derived from experimentally characterized microstructures.

For general perspectives, references, and methods relative to the area, we refer to [1].

Since, generally, the result of such a simulation must be interepreted in some statistical terms, we are led to the companion issue of coarse graining in mesoscale simulation. By this we mean understanding what distributions are reliable properties of the computed ensemble and what equations they themselves satisfy. For reasons of space, we defer discussion of this to a future work, but we present some results in this direction. We also refer to [2], where this simulation is implemented to investigate anisotropy.

Mullins Equation and Herring Condition

The form of the Mullins Equation and the Herring Condition useful for algorithmic implementation in large scale simulation may be derived by a variational procedure. Consider a network of grains with facets which meet in triple lines. To begin, suppose given three facets represented as graphs over an $x = (x_1,x_2)$ plane in (x_1,x_2,x_3) space meeting along a triple line Γ,

$$\begin{aligned} &S^{(i)} : z = u^{(i)}(x), \quad x \in \Omega_+, i = 1,2 \\ &S^{(3)} : z = u^{(3)}(x), \quad x \in \Omega_- \\ &u^{(1)} = u^{(2)} = u^{(3)} \quad on \quad \Gamma' \end{aligned}$$

where Γ' denotes the projection of Γ onto the x-plane, $\Omega^{\pm}$ denotes the regions above and below Γ'. The energy of just a single facet,

$$S : z = u(x), x \in \Omega_+ = \Omega$$

is given by

$$\begin{aligned} &\gamma(n), \quad n = \frac{1}{W}(-p_1, -p_2, 1), \quad \textit{the normal to } S \\ &p_i = \frac{\partial u}{\partial x_i}, \quad W = \sqrt{1 + |p|^2} \end{aligned}$$

(for the moment , misorientation parameters are suppressed). The energy of S is

$$E = \int_{\Omega} \gamma(n) W \; dx_1 dx_2$$

and equilibrium is determined by

$$\delta E = 0 \;\; or \;\; div(\nabla_n \gamma(n) - \gamma \tfrac{p}{W}) = 0 \; in \; \Omega$$

This means that for a time varying family of surfaces S_t the Mullins Equation takes the form

$$S_t: \; v_n = -\mu \; div(\nabla_n \gamma(n) - \gamma \tfrac{p}{W}) \; in \; \Omega$$

The Herring Condition is the natural boundary condition connected to the equilibrium status of three surfaces meeting on the triple line Γ. It arises in a straight forward variational manner, cf. also [3], [4], by allowing unrestricted variations on the arc Γ' in the calculation of δE. To express this quantity, for a surface $S^{(i)}$, with normal $n^{(i)}$, let l denote the tangent to Γ and

$$T^{(i)} = T^{(i)}_{iso} + T^{(i)}_{an}$$
$$T^{(i)}_{iso} = \gamma(n^{(i)})n^{(i)} \times l \quad and \quad T^{(i)}_{an} = (W^{(i)})^2(\gamma_{p_1}\nu_1 + \gamma_{p_2}\nu_2)n^{(i)}$$

Clearly, T_{iso} is the isotropic and T_{an} the anisotropic contribution to T. We obtain the Herring Condition

$$T^{(1)} + T^{(2)} + T^{(3)} = 0$$

For example, when γ is independent of n, the familiar condition below is easily verified:

$$n^{(1)} + n^{(2)} + n^{(3)} = 0$$

The merits of this format are that the dissipative nature of the grain growth system is clear and it is readily transformed into an algorithm for simulation. To check the dissipation, suppose that a network of surfaces $\{ S^i_t \}$ is evolving according to Mullins with Herring. The total energy is

$$E(t) = \sum_{\{S\}} \int_\Omega \gamma^S(n^S) dS$$

When computing dE/dt, the integrals over the triple lines vanish thanks to Herring and we obtain

$$\frac{dE}{dt} = -\sum_{\{S\}} \int_\Omega \frac{1}{\mu^S}(v^S_n)^2 dS \leq 0$$

So the total energy is decreasing in the absence of critical events.

Description of the numerical implementation

We give a brief description of the approach. The objects involved in the three dimensional simulation are grain boundaries, triple lines, and grains (in two dimensions, we use grain boundaries and triple junctions.) Grain boundaries and triple lines are discretized and are the objects evolved. Grains themselves are not discretized. The data structure is streamlined in this way, permitting more grains in a simulation. Moreover, boundaries are actual boundaries, not very thin three dimensional arrays of elements. Our intention is that this will lead to improved accuracy. The discretization uses second order finite difference approximations, with either explict or implicit time steps. The time evolution is executed in two steps: first grain boundaries are moved by Mullins Equation and second triple lines are moved to enforce the Herring Condition. As grain boundaries and triple lines move, critical events must be considered to reflect actual changes in the physical topology.

An important feature of the critical event implementation is that it is designed so that the discrete form of the dissipation inequality above is automatically satisfied. This leads to an extremely stable simulation.

We conclude this section with a description of the critical events. Three types occur in three dimensions: loss of grains, loss of facets, and loss of triple lines. The cristical events are detected by monitoring the size and rates of changes of topological components. Components that are shrinking quickly with respect to their size trigger critical events, a scheme which follows Kuprat [5].

Loss of grain: some neighboring grain G_N will absorb a small target grain G_T. G_N is selected as the grain with the largest (contiguous) facet among all grains sharing triple lines with the smallest facet of the target grain G_T.

Loss of facet: Implementation of this event involves two processes, 1) creation of a new volume and 2) a loss of grain event. A new volume is constructed as a small cylinder about the target facet. Then a restricted loss of grain event is applied.

Loss of triple line: This is similar to loss of facet. A new volume is created by constructing a small cylinder about the target triple line.

There are challenges to implementing this strategy. Tuning the parameters for the various critical events is difficult and, although the physical configuration may vary depending on the parameters, we do not expect this to influence the statistical properties.

In two dimensions we are able to verify some important diagnostic criteria. The first of these is the so-called parabolic growth law for isotropic, γ = constant, energy densities. The second is the correct rate of growth for 'circular grains', up to quadratic error in discretization parameter. Finally, the Mullins-von Neumann n – 6 rule holds at the level of individual grains, not merely as an ensemble average, [6].

In three dimensions, we do not have many available for criteria, but we are able to write that generally grains with less than 13 facets shrink and grains with more than 14 facets grow, in line with the conclusions of [7],[8].

Discussion

In two dimensions, this algorithm gives rise to an accurate and robust scalable numerical code which we have tested for initial configurations of 25,000 to 50,000 grains. At this writing, we think that it is the only code that allows energy densities with dependence both on the normal inclination of the grain boundary n and the lattice misorientation. We remark that dependence on n poses some particular difficulties when resolving the Herring Condition. The relative area histogram is a stable long time statistic, and we report in Fig. 1a a time dependent sequence of histograms. In Fig. 1b we compare our results with an isotropic Potts Model simulation. One may easily conclude that relative area histograms tend to discriminate poorly among input grain boundary parameters. On the other hand, the relative area histogram presented now has the status of a reliable diagnostic for future algorithms.

In three dimensions, we are working toward an numerical code, that includes all five parameters of normal dependence and lattice misorientation which will have initial configurations of upwards of 15,000 grains. Elsewhere in our group, we have devised methods of generating statistically representative 3D numerical descriptions of microstructures that are statistically representative (in terms of grain shape, size distribution, grain orientation and boundary character) of a experimentally characterized material, [9]. We plan to adapt this type of data set for use here in our 3D simulations of grain network evolution. A portrait of a 3D grain is given in Fig. 2.

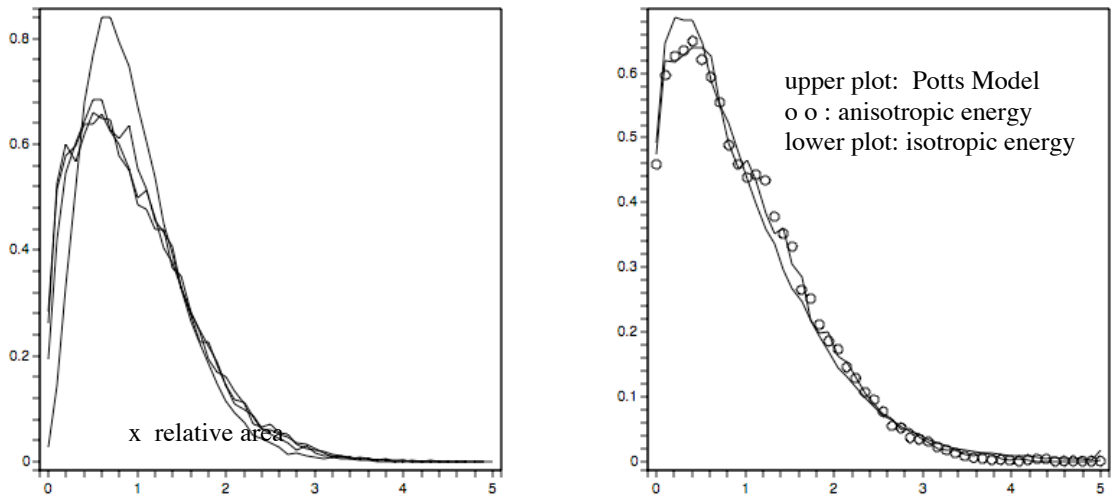

Figure 1. (a) Time dependent sequence of relative area histograms, x = relative area, y = relative count, at times t = 1, 5, 10, and 15, showing trend to self similarity. This computation is for isotropic energy. (b) Comparison of relative area histograms for isotropic (lower curve), typical anisotropic (dotted curve), and Potts-type simulation (upper curve).

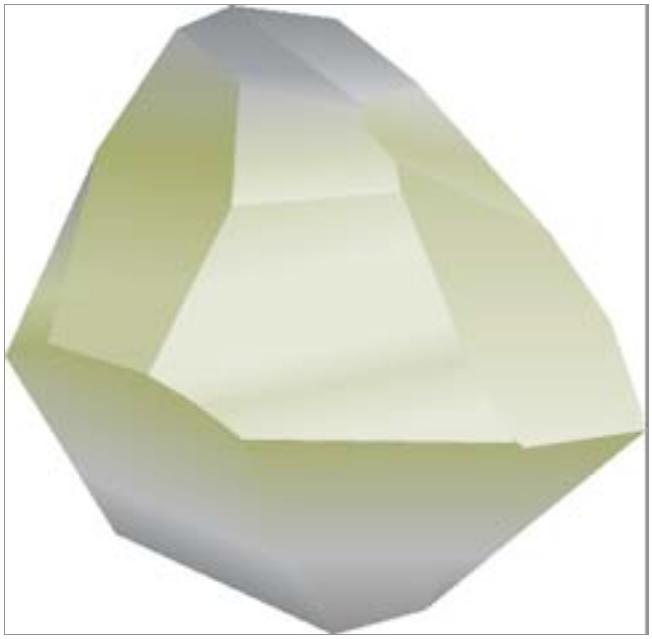

Figure 2. Depiction of a 3 D grain from an ensemble of about 500 grains. The facets are curved but are rendered as polygons for simplicity.

We are also beginning an analysis and comparison of our simulation with Al thin film experiments in collaboration with K. Barmak, *cf.* [10].

Summary

Grain growth is a complex dynamic process. Even the challenge of its simulation according to established thermodynamic principles is difficult owing to the requirements of accuracy, statistical significance of scale, and the involved data structure for this large nonlinea system. We exhibit

here what we consider to be significant progess toward implementing a flexible and reliable algorithm based on a variational approach which views the ensemble as a dissipative system, both in theory and in its implementation.

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References

[1] D. Raabe, F. Roters, F. Barlat, and L.-Q. Chen: *Continuum scale simulations of engineering materials: fundamentals-microstructures-process applications*, Wiley-VCH Verlag (2004)
[2] D. Kinderlehrer, I. Livshits, G.S. Rohrer, S. Ta'asan and P. Yu: these proceedings
[3] D. Kinderlehrer and C. Liu: Math. Models and Meth. Appl. Math. Vol. 11 (2001) 713
[4] D. Kinderlehrer, J. Lee, I. Livshits, and S. Ta'asan: in ref. [1]
[5] A. Kuprat: SIAM J. Sci. Comp. Vol. 22 (2000) 535
[6] D. Kinderlehrer, I. Livshits, and S. Ta'asan: submitted
[7] S.A. Hilgenfeldt, A. Kraynik, S. Koehler and H. Stone: PRL Vol. 86 (2001) 2685
[8] M. Glicksman: to appear
[9] D.M. Saylor, J. Fridy, B.S. El-Dasher, K.-Y. Jung, and A.D. Rollett: Metallurgical & Materials Transactions,in press (2004)
[10] K. Barmak *et a*l., Proc MRS (2004), to appear