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Mathematical model of the evolution of polycrystalline structures – Manifold-valued equation with singular diffusivity –

Ryo Kobayashi[1], James A. Warren[2]

[1]Department of Mathematical and Life Sciences, Hiroshima University,
Higashi-Hiroshima, 739-8526 Japan
[2]Center for Theoretical and Computational Materials Science and Metallurgy Division
National Institute of Standards and Technology, Gaithersburg, MD 20899, USA

Abstract

In this letter we describe a phase field model of polycrystalline solidification and grain structure evolution. Our model is constructed by introducing a orientation variable to the conventional phase field model. The orientation is expressed by the element of the group $SO(2)$ in 2D case, and by the element of $SO(3)$ in 3D. The singular diffusivity theory was developed to derive the evolution equation of the orientation variable.

Introduction and Preliminaries

Phase field model was originally designed as a model of solidification of pure materials. It could reproduce complicated patterns such as dendrites although it is written in the very simple form. This model had become popular in the society of material science since it is extendable to the alloys, multi-component systems, multi-phase systems and so on. These approaches can express inter-phase boundaries and its motion successfully, while they can not handle grain boundaries since they have no information of crystalline orientation.

The study of grain boundary formation and dynamics is central to materials science. All but the most perfect single crystals have grain boundaries, and the behavior of these interfaces can have an enormous influence on the many of the materials properties of interest to the materials engineer. Thus, over the years, there have been a number of approaches to the modeling of grain boundaries, all of which have limitations and advantages. Of particular interest are phase field models, which have gained popularity as their ability to compute realistic microstructures has been demonstrated. For an overview of this approach, the reader is recommended some of the review articles on this topic. [3]

A basic model of grain boundaries in 2D (see [4, 5]) can be derived from the total free energy

$$\begin{aligned}\mathcal{F} &= \int dV \left[f(\phi, T) + \frac{\alpha^2}{2} |\nabla \phi|^2 \right. \\ &\left. + s g(\phi) |\nabla \theta| + \frac{\epsilon^2}{2} h(\phi) |\nabla \theta|^2 \right], \end{aligned} \tag{1}$$

where $f(\phi, T) + \frac{\alpha^2}{2}|\nabla\phi|^2$ are the terms found in classical phase field models of solidification, namely the bulk free energy density, which depends on the phase field ϕ and the temperature T, with minima in the liquid and solid phases $\phi = 0, 1$ plus a gradient penalty for interfaces. For this discussion we have omitted terms accounting for interface energy anisotropy, although such effects are both important and can be accounted for with well known extensions to this theory. [4]

The final terms in the free energy are functions of the gradient in the orientation, θ; introduced to allow for grain boundary energy misorientation penalties. These terms inclusion also provides a realistic description of related phenomena such as polycrystalline growth and nucleation.[6],[7] The couplings $g(\phi)$ and $h(\phi)$ are chosen so there are no energy penalties in the liquid (i.e. $g(0) = h(0) = 0$.) The dynamics of the system are found by imposing the thermodynamic requirement that ϕ and θ evolve so as to minimize the free energy $\mathcal{F}$.

In mathematical sense, the evolution equation of the orientation variable θ is not an usual partial differential equation but an extended one. We had developed the singular diffusivity theory for the convex and non-differentiable energy.[1] The evolution equation derived from such an energy includes a non-local interaction through the infinitely large diffusivity, which keep the value of orientation variable constant in each grain. Such an unusual situation is justified by the singular diffusivity theory.

In numerics, there are difficulties to solve the equation, not only becuase it includes very strong singularity but also because it takes values in the manifold $SO(2)$.[2] We developed the numerical code by resolving all these difficulties, which we will demonstrate in the last figure for $SO(3)$-valued simulation.

Extension to 3D: Formulation and Solution

We note that a single angle cannot represent an orientation in 3D, and thus this concept must be replaced with a more robust mathematical description of orientation. Specifically, θ must be replaced with an object that captures the three rotational degrees of freedom available in 3D. Additionally, we must also define the norm of this object. With these two mathematical

concepts the transition to three dimensions is fully posed.

Formulation

There are many ways to represent orientations in 3D, most quite familiar to crystallographers: Euler angles, rotation vectors, Rodrigues vectors, quaternions, etc. All of these representations are mathematically equivalent representations of the group $SO(3)$ (special orthogonal group 3D), but retain advantages and disadvantages, depending on the application. If we call a member of this group P, then P is a 3×3 orthogonal matrix ($P^T P = I$,where I is the identity matrix), and P has a positive determinant $\det P = 1$. Thus, we say $SO(3)$ is naturally embedded in $\boldsymbol{R}^9$, as it can be represented as a nine-dimensional object (with 6 constraints originating from the orthogonality condition).

To proceed, we must find the 3D analog to the fundamentally 2D quantity $|\nabla\theta|$. A gradient is simply a difference over an infinitesimal distance, thus we need to compute the *norm of the difference, in some sense, between two 3D orientations.* We consider two possible choices for this measure, employing a function of two $SO(3)$ matrices $\rho(P,Q)$:

$$\begin{aligned} \text{Type I} \quad &: \quad \rho(P,Q) = |P-Q| = |PQ^{-1} - I| \\ \text{Type II} \quad &: \quad \rho(P,Q) = \sqrt{2}\cos^{-1}\frac{\mathrm{tr}PQ^{-1}-1}{2} \end{aligned}$$

Note that, PQ^{-1} is the *misorientation* between two crystals. The meaning of the Type I measure is trivial, as it measures the distance between two matrices in $\boldsymbol{R}^9$. The Type II measure, on the other hand, measures the length of the geodesic in $SO(3)$ connecting two matrices. These two measures coincide when P and Q are infinitesimally close, but they will yield different values when there is a discontinuity between P, Q *as is often the case for discrete computations on a lattice*

With these definitions we can now write down our model, by simply substituting $|\nabla\theta| \rightarrow |\nabla P|$ in Eqn. 1:

$$\begin{aligned} \mathcal{F} \quad &= \int dV \big[f(\phi,T) + \frac{\alpha^2}{2}|\nabla\phi|^2 \\ &\quad + sg(\phi)|\nabla P| + \frac{\epsilon^2}{2}h(\phi)|\nabla P|^2 \big], \end{aligned}$$

A more explicit form can be obtained using $|\nabla P| = \sqrt{|\nabla P|^2} = \sqrt{\sum_{i,j=1}^{3} |\nabla p_{i,j}|^2}$, where $p_{i,j} = [P]_{i,j}$.

Solution

Having posed the above free energy, we must now perform a minimization, to derive equations of motion for both the phase field ϕ and the orientation. We must proceed with care to ensure that the equations of motions keep the variables in $SO(3)$. There are several ways to proceed: (1) Derive equations for the constrained free energy, which has only 3 degrees of freedom or (2) derive equations on $\boldsymbol{R}^9$, and project the results back into $SO(3)$. For both methods we need to derive the variational derivative of the free energy with respect to orientation, which is simply

$$\frac{\delta \mathcal{F}}{\delta P} = -\nabla \cdot \left(sg(\phi) \frac{\nabla P}{|\nabla P|} + \epsilon^2 h(\phi) \nabla P \right).$$

In deriving the equations of motion for the constrained free energy, an element of $SO(3)$ is written in the form $P = P(u, v, w)$ where the triplet (u, v, w) is some local coordinate, for example, the Rodrigues vector. The equations of motion for these variables should be

$$\tau_P \frac{\partial u}{\partial t} = \left\langle -\frac{\delta \mathcal{F}}{\delta P}, \frac{\partial P}{\partial u} \right\rangle \tag{2}$$

with identical equations for $u \to v, w$. Note that the quantity $\langle \cdot, \cdot \rangle$ is the usual inner product in $\boldsymbol{R}^9$ (a fully contracted matrix product), and τ_P is an inverse mobility.

Alternatively, to use a projective formulation, we develop 9 equations of motion in $\boldsymbol{R}^9$ keeping the solution within $SO(3)$ by taking a projection of driving force onto the tangential plane of $SO(3)$. It is given in the form $\tau_P \partial P/\partial t = \pi_P \left(-\delta \mathcal{F}/\delta P\right)$ where π_P is the projection operator. This approach allows for substantially improved numerical efficiency. However, we reserve discussion of this technique for a later publication.

Following the preceding arguments we can derive the evolution equation for ϕ and P, implement the equations in computer code and solve. Our first calculations were for a thin film where the grains are nearly 2D objects, but *their orientation is 3D, and the dynamics will be governed by the evolution of all of these angles.* There are numerous experimental systems analogous to this calculation (see Fig.1.) Our results showing growth, impingement and coarsening are given in Fig. 2. Also fully 3 dimensional simulation is shown in Fig.3.

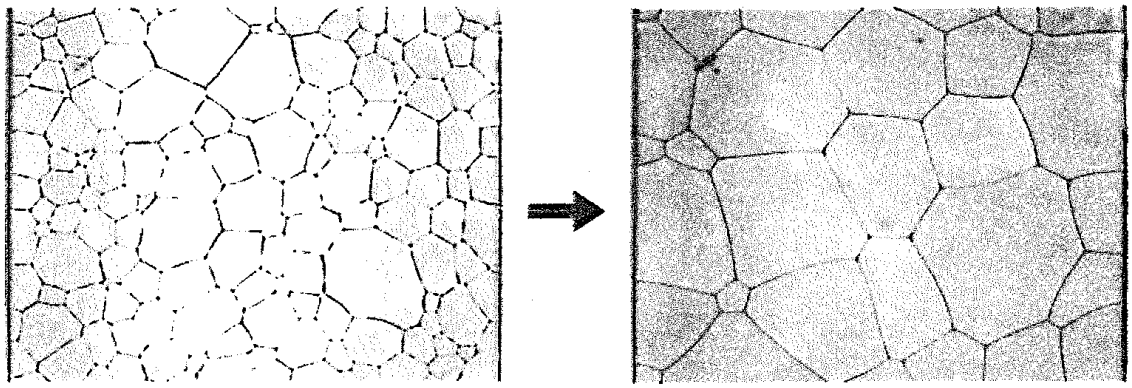

Figure 1: Coarsening process of grain structure of succinonitrile. The grain structure is almost 2D, while the orientation of each grain is necessarily 3D. (courtesy of Drs. Lee and Losert, U. Maryland)

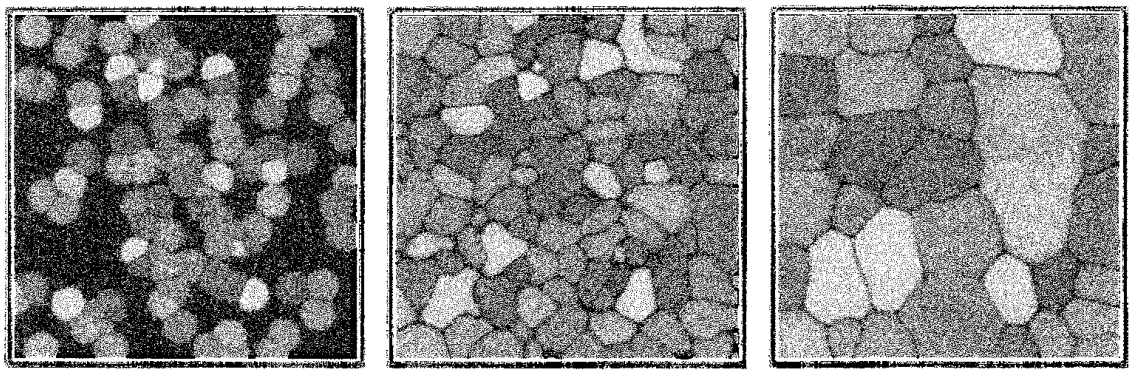

Figure 2: Simulation of solidification and coarsening process (the color indicates one of the three Euler angles (all of which were solved for).

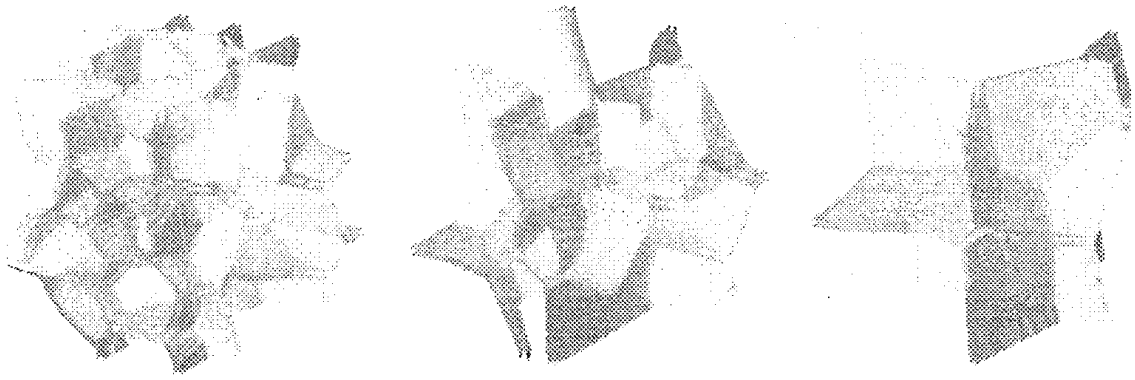

Figure 3: Simulation of fully 3 dimensional coarsening process

Herein, we have extended our previous work in 2D to 3D. For a complete formulation to be obtained, we must also include the *important* consequences of anisotropy. In other words we have examined the consequences of misorientation, but not the consequences of inclination on the statics and dynamics of grain boundaries. Additionally, we have yet to account for the underlying crystal symmetries These effects can be included, and will be discussed in future work.

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