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Grain rotation in 2d-hexagonal systems with competing interactions

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Abstract

We look at a grain rotation mechanism in a bidimensional pattern forming system exhibiting an hexagonal phase. This mechanism is believed to be relevant in the coarsening process of a variety of physical systems with short- and long-range competing interactions. We focus on the Cahn–Hilliard model with thermal fluctuations. The grain rotation process is uncovered from the dynamical evolution of a misoriented circular grain surrounded by an otherwise perfect crystal.

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1. Introduction

During the last two decades a great deal of effort was committed to understanding the mechanisms of pattern formation, ubiquitous in a number of diverse contexts (Rayleigh–Benard convection, ferrimagnetic repulsion in garnet films, and biological growth [1] or block copolymer thin films [2]). Among the different periodic and quasi-periodic equilibrium structures experimentally observed in two dimensional systems are stripes, hexagonally ordered disks, and quasi-crystals. The hexagonal systems pose a particularly challenging problem for analytical treatment and have been addressed in the literature, albeit less frequently than other stable phases (e.g. stripes), with varying degrees of success.

In order to understand the ways coarsening proceeds and order is achieved, several mechanisms have been proposed and studied: curvature driven grain growth [2,3], annihilation of topological defects [4] and, to a lesser extent, grain rotation [5] are examples of such mechanisms.

In particular, the mechanism of grain rotation has been addressed in the context of crystalline solids [6,7].

In this work, we consider 'soft' 2-D systems displaying hexagonal symmetry. Differently from crystalline solids or Lennard-Jones crystals, such soft materials posses the ability of relaxing the elastic free energy through the strain of local domains.

We focus on the dynamical evolution of a misoriented circular region planted in an otherwise perfect lattice exhibiting hexagonal order. For different initial misorientations of the interior region of the circle, we track its evolution at later times and compare results. The conclusions prove helpful in elucidating the various paths that such systems follow to equilibrium.

2. The model and numerical implementation

The context in which our numerical experiments were conducted is provided by the classical Cahn–Hilliard–Cook model [8], describing the dynamics for a conserved order parameter:

$$\frac{\partial\psi}{\partial t} = M\nabla^2\left(\frac{\delta F}{\delta\psi}\right) + \zeta, \tag{1}$$

where ψ is the order parameter, M is a phenomenological constant accounting for the mobility of the system, $F(\psi)$ is the free energy functional and ζ stands for a random noise

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bath that is both time and space dependent, of zero average and with constant amplitude.

The free energy functional $F(\psi)$ splits as

$$F(\psi) = F_S(\psi) + F_L(\psi), \tag{2}$$

where each term models short- and long-range interactions. The short-range term has the typical Landau form,

$$F_S(\psi) = \int d\mathbf{r}\left[H(\psi) + \frac{D}{2}(\nabla\psi)^2\right], \tag{3}$$

where $H(\psi)$ represents the mixing free energy of the two phases and the gradient term accounts for the free energy penalty generated by variations of ψ in space (energy at the interface), and D is a diffusion coefficient.

The free energy $H(\psi)$ has the form

$$H(\psi) = \alpha\psi^2/2 - \lambda\psi^3/3 - \mu\psi^4/4. \tag{4}$$

The long-range contribution to the free energy, on the other hand, is taken to be

$$F_L(\psi) = \frac{\beta}{2}\int d\mathbf{r}'\, d\mathbf{r}\, G(\mathbf{r}-\mathbf{r}')\psi(\mathbf{r})\psi(\mathbf{r}'),$$

where $G(\mathbf{r}-\mathbf{r}')$ solves $\nabla^2 G(\mathbf{r}) = -\delta(\mathbf{r})$.

The numerical experiments were performed using the cell dynamics method on a two-dimensional square lattice according to the scheme [9]:

$$\psi(\mathbf{n}, t+1) = \Gamma(\psi(\mathbf{n}, t)) - \langle\Gamma(\psi(\mathbf{n}, t)) - \psi(\mathbf{n}, t)\rangle - \beta\psi(\mathbf{n}, t) + \eta\zeta(\mathbf{n}, t), \tag{5}$$

where

$$\Gamma(\psi(\mathbf{n}, t)) = f(\psi(\mathbf{n}, t)) + D(\langle\psi(\mathbf{n}, t)\rangle - \psi(\mathbf{n}, t)), \tag{6}$$

and the map function $f(\psi(\mathbf{n}, t))$ is defined in Ref. [5]. In this context, the lattice coordinates are $\mathbf{n} = (n_x, n_y)$ and $\langle\Xi\rangle$ stands for the average of Ξ over all nearest neighbors and next nearest neighbors [9]. The Laplace operator is also in averaged form $\nabla^2(\Xi) = 3[\langle\Xi\rangle - \Xi]$.

Our simulations employed cyclic boundary conditions, typically on a 512×512 lattice. In Eq. (5) η is the noise amplitude and ζ are random numbers uniformly distributed in the interval $[-1, 1]$ (see Ref. [5]). The external crystal was obtained by setting the following initial condition in (1):

$$\psi(\mathbf{r}) = \frac{1}{2}\psi_0 \sum_{j=1}^{3} e^{\mathbf{k}_j\cdot\mathbf{r}} + \text{c.c.}, \tag{7}$$

where ψ_0 is the initial amplitude, $\mathbf{k}_1 = (k_0, 0)$, $\mathbf{k}_2 = (-k_0, -\sqrt{3}/2\,k_0)$ and $\mathbf{k}_3 = (-k_0, \sqrt{3}/2\,k_0)$ are the wave vectors of the hexagonal lattice. The interior of the circle was also obtained through (7) but with wave vectors rotated to the desired angle.

3. Results

A series of runs were performed for different orientational mismatches of the interior circular region, ranging from small angles (2.5°) to near the largest possible (30°). For each run, the noise amplitude was held constant ($\eta = 0.25$).

Fig. 1a shows a circular grain of radius $R \simeq 13a$, were a is the average inter-sphere distance (in our experiments $a \simeq 6.15$ lattice units) with a misorientation of 10°. Note in this figure that the disks near the interface are strongly distorted. The local field acts on these disks by changing both shape and size. This feature, distinctive of soft materials, allows for extra degrees of freedom to further relax the free energy.

Fig. 1b shows the Delaunay triangulation corresponding to Fig. 1a. The triangulation allows us to determine both local orientation of the hexagonal pattern and topological defects [5]. For all runs the only defects observed were dislocations (penta–hepta defects). Disclinations (orientational defects) did not occur. In this figure penta (dark grey/red) and hepta (light grey/ green) circles connected through a line (lighter grey/yellow) represent a dislocation. Notice that dislocations decorate the grain boundary of the grain depicted in Fig. 1a. It was observed that the spacing

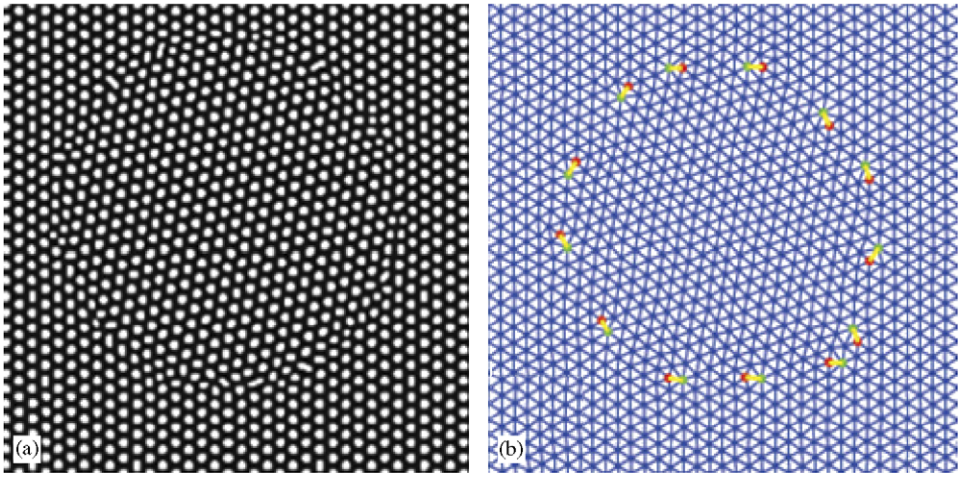

Fig. 1. Circular misoriented grain in the interior of a larger one: (a) hexagonal pattern containing a circular grain with a 10° orientational mismatch and (b) corresponding line of dislocations along the grain boundary.

between dislocations satisfies $d \sim a/\Delta\theta$, where $\Delta\theta$ is the misorientation between grains. This result is in agreement with the description of straight low angle grain boundaries as arrays of dislocations [10].

In the range of parameters considered in this work the equilibrium state the system reaches is an hexagonal lattice with long range order, and its free energy is roughly invariant under rigid rotations. The paths towards this equilibrium consists mostly of the shrinking of the circular domain possibly accompanied by grain rotation. This shrinking process induces configurations in which the dislocations spanning the grain boundary are progressively closer to their neighbors, favoring recombination and annihilation events. Reducing the average distance between neighboring dislocations implies the rotation of the interior circular domain increasing its misorientation. As the misorientation is bounded above, the grain rotation mechanism saturates and shuts off. Shrinking, then, stands as the only resource to achieve order.

This picture does not describe accurately what goes on at small angle misorientations if the initial setup is not rescaled. Evidently, the description of grain boundaries as an array of dislocations fails for configurations in which the average distance between dislocations becomes of the order of the grain size.

Fig. 2 shows a snapshot of the evolution of the misorientation angle as a function of the initial misorientation. Notice that the mechanism the system chooses to adjust to local equilibrium depends strongly on the initial conditions: at the rightmost endpoint of the misorientation range, the configuration stays put as it cannot increase its misorientation any further. In this case, the mean distance between dislocations becomes comparable to a lattice constant favoring the recombination of dislocations.

For initial misorientations of the order 10° and 22° the system, not unexpectedly, increases the orientational mismatch ($\Delta\theta/\Delta\theta_0 > 1$) when kicked by the thermal bath, trying to fall into a local minimum of the potential. Although according to the Read–Shockley formula [10] the line tension is expected to increase with misorientation, the surface energy decreases as a consequence of the circle shrinking. Throughout this process, the number of dislocations remains constant. As time proceeds, the simultaneous effects of shrinking and rotation induce the recombination of dislocations, rearranging the boundary of the grain.

Much like in crystalline solids the line tension is expected to be orientationally dependent. Fig. 3 clearly shows anisotropic effects for a configuration with initial misorientation of 15°. In this configuration the grain adopts a quasi-hexagonal form in the sense that dislocations no longer span a circular boundary but accommodate to the orientational anisotropy forming an hexagon-like figure (obviously, in a purely isotropic case the surface minimizing the free energy is a circle).

In Fig. 2, we observe that the local misorientation is decreased for low angles (less than 7.5°), contrary to what occurred for previously discussed cases. Although $\Delta\theta/\Delta\theta_0 \to 0$ at the center of the circle, there are important distortions in the orientational field in the neighborhood of the core of the dislocations (see Fig. 4b). Since the typical distance between dislocation is of the order of R no screening effects of the long range potential occur as the ones reported for straight grain boundaries [11]. Then, the reorganization of the pattern at the center of the initial circle is a consequence of the interaction between the long range fields of the individual dislocations.

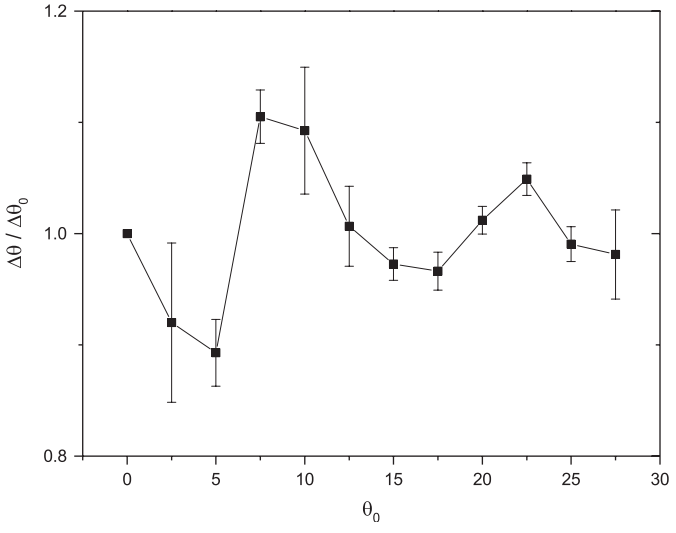

Fig. 2. Snapshot of the time evolution of $\Delta\theta/\Delta\theta_0$ as a function of the initial misorientation after 10^6 time-steps.

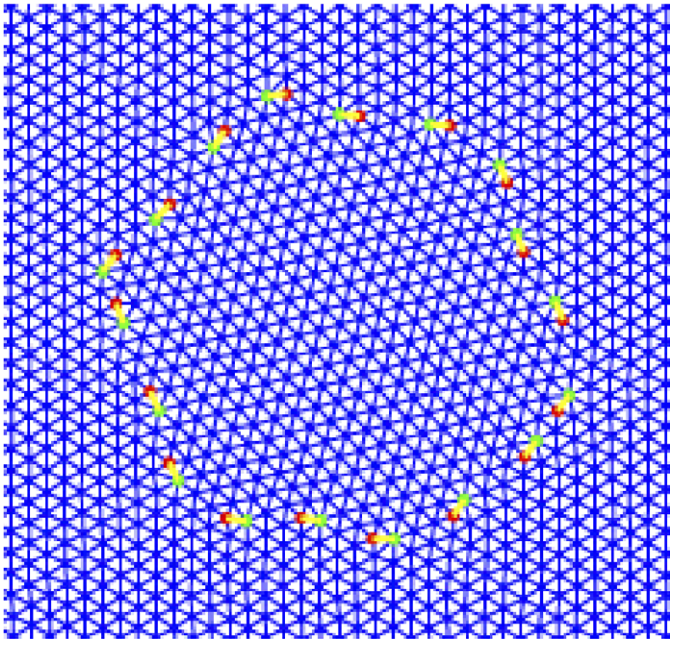

Fig. 3. Grain configuration at intermediate times for an initial condition with 15°. Note the appearance of facets as consequence of line tension anisotropy.

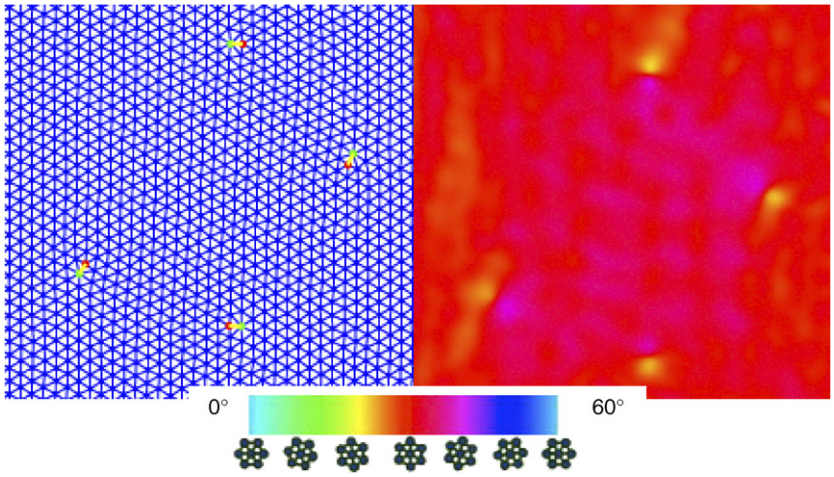

Fig. 4. For small values of $\Delta\theta_0$, the orientational energy is not longer uniformly distributed: (a) dislocation array at intermediate times corresponding to $\Delta\theta_0 = 2.5^\circ$ and (b) associated orientational map.

Regarding the noise, it was observed that the relaxation processes were sensitive to its amplitude (for more details see Ref. [5]).

4. Conclusions

A grain rotation mechanism was reported for 2D systems displaying a stable hexagonal phase. Through numerical simulations of the Cahn–Hilliard–Cook model, a configuration consisting in a misoriented circular grain was set up and its evolution tracked. The notion of grain rotation together with the mechanism of grain shrinking was addressed through orientational maps and topological defects. For suitable choices of the grain size and noise amplitude, the systems ability to reach an ordered state was shown to correspond to the formation of well defined grain boundaries by an even distribution of dislocations. This was achieved mostly by grain shrinking (effectively bringing dislocations together) or by an *a priori* counter-intuitive rotation towards a more disoriented configuration. A more complete picture of mechanisms leading to order and analytical treatments will be the subject of further research.

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