Effect of noise on ordering of hexagonal grains in a phase-field-crystal model

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We present a quantitative analysis of grain morphology of self-organizing hexagonal patterns based on the phase-field crystal model to examine the effect of stochastic noise on grain coarsening. We show that the grain size increases with increasing noise strength, resulting in enhanced hexagonal orientation due to noise up to some critical noise level above which the system becomes disordered.

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I. INTRODUCTION

When a system is suddenly brought from its homogeneous phase to a thermodynamically unstable region of the phase diagram, the final equilibrium is not reached instantaneously. There are degenerate ground states competing to select the ordered phase, and different spatial regions of the system are in different broken symmetry states. The evolution of the pattern involves reorientation of domains of ordered phase and elimination of topological defects. As a result, a characteristic linear size of the domains, $\ell(t)$, grows with time, typically as a power law $\ell(t) \sim t^n$ at late times. A typical example of these coarsening (or phase ordering) phenomena is spinodal decomposition in binary alloys or binary mixtures of immiscible fluids. Other examples include the organization of crystalline domains (also called grains) in a solid.

A great effort has been made to explore the phase ordering dynamics in the case where the system is spatially uniform within each domain [1]. In particular, many studies focused on the growth law for $\ell(t)$, and the growth exponent n is now known both for conserved ($n = 1/3$) and nonconserved ($n = 1/2$) order parameters. In contrast, considerably less progress has been made in the study of coarsening in systems where an ordered phase is characterized by periodic patterns so that the domains themselves contain a spatial structure. For example, for the ordering of striped patterns, the growth exponents of various values are reported in the range 1/5–1/2 [2–7]. Of particular interest in this connection is the effect of noise on the coarsening of crystalline domains.

There are two types of noises, additive and multiplicative, but in the present paper we will be concerned with additive noise [8]. The term additive stands for the absence of coupling between the order parameter field and the noise, and the noise can have an internal (thermal) or external origin with respect to the system under study. (The multiplicative noise enters the system, e.g., through external perturbations that will be coupled to the order parameter.) Usually noises are expected to reduce the stability of ordered states. For stripe pattern formation, however, previous simulations [2,9,10] demonstrated that noises promote the formation of the patterns. This is attributed to the fact that noise can provide a source of depinning for topological defects, the interaction of which is known to predominantly drive the coarsening process [7]. Since there is no obvious reason that this constructive effect of noise would not occur in the ordering of hexagonal domains, the recent claim [11] to the contrary is disturbing. In this paper, we dispute this claim since we observe similar slowing of the coarsening with increasing noise strength in our grain growth simulations, but with a subsequent crossover to the speeding-up behavior at late times.

II. MODEL

We study the coarsening dynamics of hexagonal domains using the time-dependent Ginzburg-Landau equation for a conserved order parameter field ψ:

$$\frac{\partial \psi}{\partial t} = \nabla^2\big[-\epsilon\psi + \big(\nabla^2 + k_0^2\big)^2\psi + s\psi^2 + \psi^3\big] + \eta = \nabla^2 \frac{\delta F}{\delta \psi} + \eta, \tag{1}$$

where the additive noise η is assumed to be Gaussian white with zero mean:

$$\langle \eta(\boldsymbol{x},t)\rangle = 0, \quad \langle \eta(\boldsymbol{x},t)\eta(\boldsymbol{x}',t')\rangle = -2\zeta\nabla^2\delta(\boldsymbol{x}-\boldsymbol{x}')\delta(t-t').$$

In this paper, we will focus exclusively on the case where η is conserved, which is of particular interest in relation to the recent simulation result [11]. We note in passing that the previous work [2,9,10] referred to in the Introduction all focused on the nonconserved version of Eq. (1) with nonconserved noise. In the above equation, we may set $\langle\psi\rangle = 0$ without loss of generality.

The free-energy functional $F\{\psi\}$ is given by

$$F = \int d\boldsymbol{x}\left\{-\frac{1}{2}\epsilon\psi^2 + \frac{1}{2}\big[\big(\nabla^2 + k_0^2\big)\psi\big]^2 + \frac{s}{3}\psi^3 + \frac{1}{4}\psi^4\right\}. \tag{2}$$

We here consider a spatially periodic hexagonal pattern, so that the relevant order-parameter field depends only on two-dimensional coordinates, $\boldsymbol{x} = (x,y)$, and time, t. The constants ϵ, s, k_0, and ζ are phenomenological parameters. Throughout the paper we assume that $s, k_0, \zeta > 0$.

The free energy (2) has been originally derived for the special case of $s = 0$ by Swift and Hohenberg in the description of patterns in Rayleigh-Bénard convection [12]. In this case the competition between the surface energy contribution given by the $(\nabla\psi)^2$ term and the curvature energy term [$\propto (\nabla^2\psi)^2$] gives rise to spatially modulated structures (stripes, or interchangeably called rolls and lamellae) with period $\sim 2\pi/k_0$ for $\epsilon > 0$.

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When $s \neq 0$, the symmetry with respect to the reflection $\psi \to -\psi$ is broken, and the transcritical bifurcation to hexagonal patterns becomes possible. In the crystal growth literature, Eq. (1) is called the phase-field crystal (PFC) model [13], and has been intensively used during recent years in the study of a wide range of problems such as solidification and elastic and plastic deformations in nonequilibrium microstructure formation [14].

III. SIMULATION RESULTS AND DISCUSSIONS

We have simulated Eq. (1) on a square lattice system of size 1024×1024 with periodic boundary conditions. The code is pseudospectral using fast Fourier transforms and uses the exponential time-differencing method [15] combined with a fourth-order predictor-corrector method [16] for time advancement. The spatial mesh size Δx and the time step Δt used in the numerical integration are set to $\Delta x = 1.0$ and $\Delta t = 0.05$. The parameters used are $\epsilon = 0.1$, $s = 0.4$, and $k_0 = 1$ at variable values of ζ. The choice of parameters ϵ and s has been made so that those values lead to the stable hexagonal phase. [The range of ϵ and s where the hexagonal and striped phases are favored can be obtained for the PFC model (1) by the standard calculation (see, for example, Ref. [13]) with use of a common tangent construction.] The initial conditions are a uniformly random distribution of the ψ's in the range $[-0.1,0.1]$. All the quantitative results represent the average over five samples.

In order to quantify the degree of ordering in the system, we have calculated the orientational field correlation function. It is defined by

$$C_6(|\,\mathbf{r}-\mathbf{r}'\,|,t) = \langle e^{i6[\theta(\mathbf{r},t)-\theta(\mathbf{r}',t)]}\rangle, \tag{3}$$

averaging over the spatial coordinates $\mathbf{r}$ and $\mathbf{r}'$ for fixed $|\,\mathbf{r}-\mathbf{r}'\,|$ for each time t. The local orientation $\theta(\mathbf{r},t)$ of the hexagonal grains was computed with a slight modification of the wavelet transform method due to Cross *et al.* [15]. Since more details about the method can be found elsewhere [6], we only note here that for this data processing the precise determination of the main peak of the scattering function is necessary. To that end we computed the circularly averaged scattering function $S(q,t)$ defined by $S(q,t) = \langle \psi(\mathbf{q},t)\psi^*(\mathbf{q},t)\rangle$, with $\psi(\mathbf{q},t)$ being the Fourier transform of the order parameter, and the orientation of the wave vector $\mathbf{q}$ was averaged over. We fitted the primary peak of $S(q,t)$ to a squared Lorentzian form [3]

$$S(q,t) = a^2/[(q^2-b)^2+c^2]^2. \tag{4}$$

The value of the peak position ($q=\sqrt{b}$) thus extracted was then used in the subsequent wavelet transform. In the definition (3), a factor of 6 is required by a sixfold symmetry of hexagonal patterns.

As exhibited in Fig. 1, the correlations $C_6(r,t)$ decay with increasing separation r, and from this decay we can extract the orientational correlation length, $\xi_6(t)$, as a measure of the domain size at time t. We choose as $\xi_6(t)$ the value of r at which $C_6(r,t)$ reaches the value of 0.4 [note that $C_6(0,t)=1$]; with the other values chosen we found that it entailed little change in the growth exponent. The length scale $\xi_6(t)$ measured in this way is displayed in Fig. 2. In addition, we considered another measure of the domain growth. The width of the fundamental peak in $S(q,t)$ was determined as mentioned above. Then the full width at half maximum, δq, contains information about a typical size of domains by $\xi(t) = 2\pi/\delta q$. The time evolution of $\xi(t)$ is also shown in Fig. 2.

There is one point to be made in passing before discussing these results. In studies of the domain coarsening, particularly in macrophase separation as the spinodal decomposition, the scattering function $S(q,t)$ and hence $\xi(t)$ are most commonly used as a measure of domain growth. However, in the spatially periodic patterns as observed in the present study, the length scale extracted from $S(q,t)$ does not capture their inherent

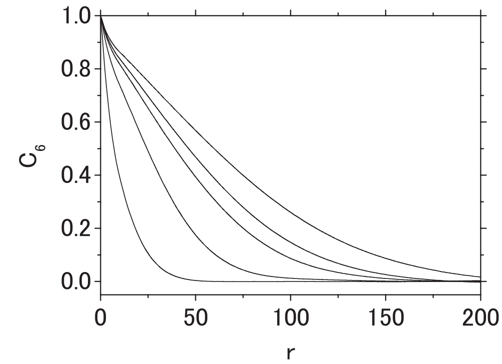

FIG. 1. Time evolution of the orientational correlation function $C_6(r,t)$ for $\zeta = 0.006$. The times are $t = 99.8$, 997.65, 5000, 9976.35, and 25059.4 increasing from left to right.

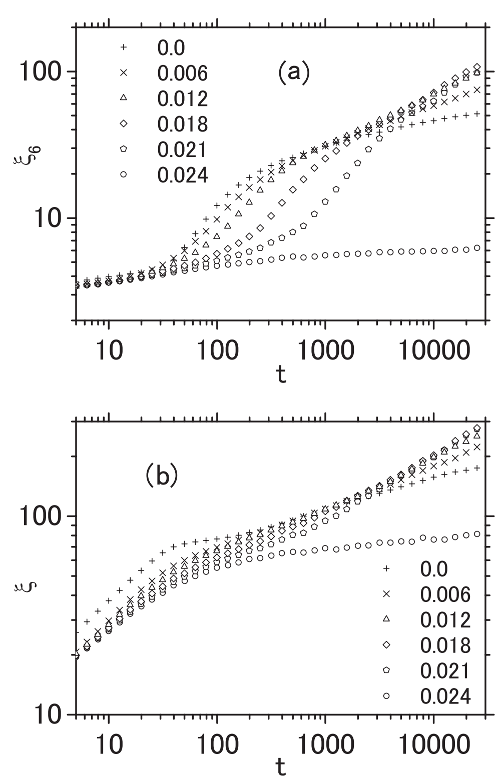

FIG. 2. Time evolution of the orientational correlation length $\xi_6(t)$ (a) and the correlation length $\xi(t)$ determined from scattering function (b) for different noise strengths; the legend shows the values of ζ for symbols used in the plot.

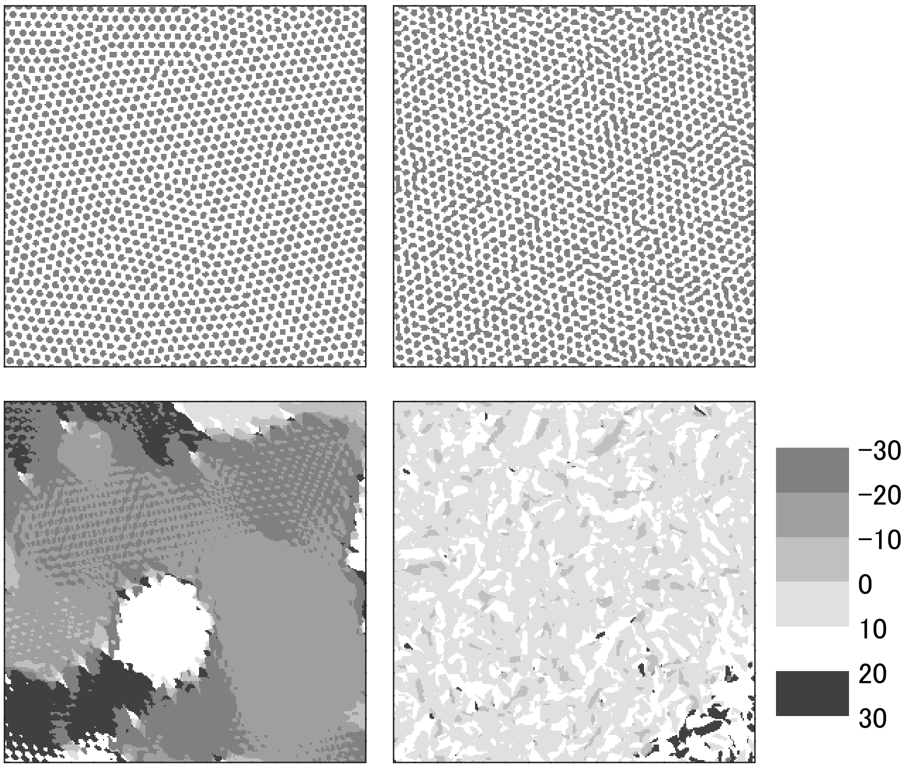

FIG. 3. Top: hexagonal patterns achieved at $t = 25\,059.4$ for $\zeta = 0.0$ (left) and 0.018 (right) with the same initial condition. The bright regions denote positive values of the order parameter ψ while the dark ones denote negative ψ. Each figure exhibits a 256^2 portion of the 1024^2-lattice system. Bottom: orientational maps corresponding to the figures on top, with the gray scales shown at the right indicating the orientational angle θ over the range $[-30^\circ, 30^\circ]$ as appropriate for a sixfold symmetric pattern.

feature that the orientation of grains is a continuous variable. Consequently, although it does capture the variation in the local wave number, it does not have the same immediate geometrical interpretation of the degree of alignment as the length scale extracted from the orientational correlation function [5].

There are several striking features to be noticed in Fig. 2. First, at a time around $t = 2000$, there is a qualitative change in the evolution of the system. During the earlier times the grain size decreases with increasing noise strength. It seems likely that this is a time range of observation reported in Ref. [11]. At the later times, by contrast, the coarsening dynamics have sped up, and we have in fact found that a higher value of ζ results in a configuration with a larger average domain size. Figure 3 compares the order-parameter configurations for $\zeta = 0$ and $\zeta = 0.018$. The high degree of translational and orientational order in the presence of noise is evident, though ordered pores are roughened due to the noise. Finally, for the stronger noise a steady state is rapidly reached, which is characterized by the small range of spatial correlation. Eventually, if the noise strength is large enough, the system becomes completely disordered. In the stripe pattern coarsening, the one-point distribution function, $\rho(\psi) \equiv \int d\boldsymbol{r}\,\delta[\psi - \psi(\boldsymbol{r})]$, has also been monitored [2,9]. In those numerical simulations, the transition from a low to high noise-strength state was marked by a crossover from a bimodal to unimodal distribution in ρ and a sharp decrease in its peak position. In contrast, in the process of hexagonal pattern formation such a pronounced difference is not apparent in $\rho(\psi)$, as shown in Fig. 4(a).

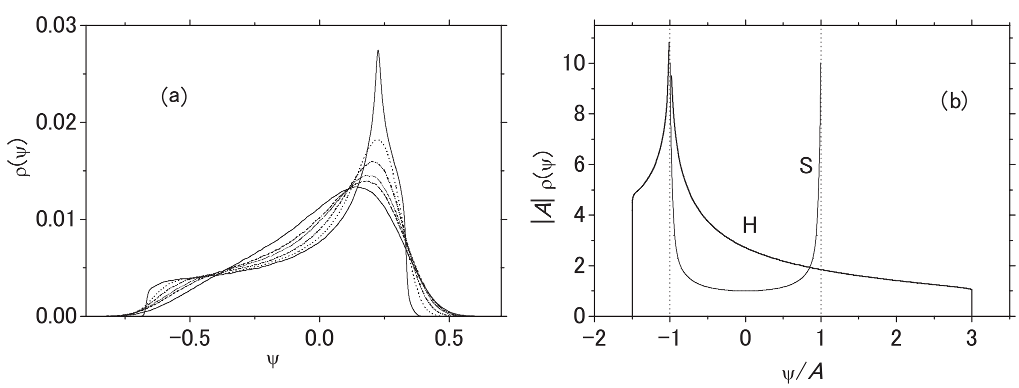

FIG. 4. (a) The one-point distribution function $\rho(\psi)$ at $t = 25\,059.4$ for noise intensities $\zeta = 0$, 0.006, 0.012, 0.018, 0.021, and 0.024 with decreasing peak values. (b) The scaled one-point distribution function $|A|\rho(\psi)$ vs the scaled ψ, ψ/A, for an ideal striped pattern (S): $\psi = A\cos x$, and an ideal hexagonal pattern (H): $\psi = A[\cos x + \cos(\frac{1}{2}x + \frac{\sqrt{3}}{2}y) + \cos(\frac{1}{2}x - \frac{\sqrt{3}}{2}y)]$. One sees that the data given in (a) are to be associated with the curve H with a choice of $A < 0$.

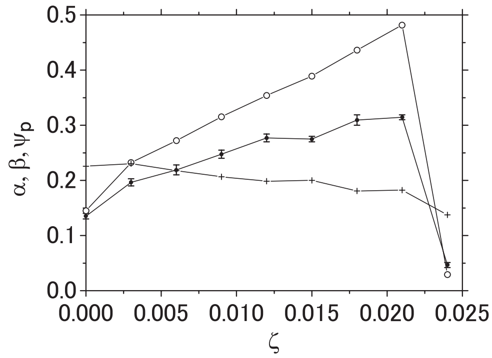

FIG. 5. Growth exponents versus noise strength ζ: (• with error bars) α for ξ_6; (○) β for ξ, the numerical uncertainty being within the size of the symbol. Also shown by daggers is the position ψ_p of the peak in $\rho(\psi)$ at $t = 25\,059.4$.

For all noise strengths, $\rho(\psi)$ is asymmetric and single peaked [cf. Fig. 4(b) [17]], and the peak position gradually moves toward $\psi = 0$ with the increase of noise strength. In Fig. 5, the peak position in ρ is shown as a function of ζ. It should be noted that a peak remains at finite ψ even for the strong noise strength $\zeta = 0.024$. Needless to say, such asymmetries are due to the presence of a symmetry-breaking term ($\propto s$) in the PFC equation (1).

At each noise strength, both characteristic length scales are well fitted by a power law at late times [18], but we find different exponents in the two cases: $\xi_6(t) \sim t^\alpha$ and $\xi(t) \sim t^\beta$ with $\alpha > \beta$. The higher exponent for $\xi_6(t)$ than for $\xi(t)$ is in accord with the recent result of Vega *et al.* [20] which was obtained by numerical simulations of a model equation [21] for two-dimensional hexagon-forming block copolymers. This result implies a breakdown of the dynamic scaling hypothesis that a single characteristic length scale dominates the asymptotic pattern formation process. The growth exponents α and β are obtained by fits to the power law to ξ and ξ_6 shown in Fig. 2. They are plotted as a function of the noise strength ζ in Fig. 5. It is clear that there is a dependence on the value of ζ. The growth exponent first becomes progressively larger for higher values of ζ, and it drops rather abruptly as ζ increases further. Recently, by using scanning electron microscopy on a block copolymer thin film, Harrison *et al.* [22] found the exponent $\alpha = 1/4$ for ξ_6. Numerical results from their group [20] also showed that $\alpha = 1/4$ and $\beta = 1/5$. If the noise strength ζ is treated as an adjustable parameter [23], these results are consistent with our present result at $\zeta \approx 0.005$.

IV. CONCLUSIONS

In summary, we have examined the noise effects on coarsening of hexagonal patterns using numerical simulations of the PFC equation. The results we have obtained are closely analogous to the previous results for ordering dynamics of striped patterns [2,9,10] in two important respects: (1) The additive noise increases the rate of growth of domains. (2) Systems are best able to coarsen at the edge of a disordered state. This suggests the generality of the role played by the noise in ordering dynamics of a variety of spatially periodic structures.

In closing, the final caveat is in order. In light of the complex transient shown in Fig. 2, one might argue that the asymptotic region where we have fitted the growth exponents is not sufficiently large. Then it might well be that the sensitivity to noise strength is a transient effect. Due to computational limitations, we are unable to refute this possibility. It is certainly worth investigating in the future.

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for $-3/2 < x \leqslant -1$, $F(x) = \frac{4}{\sqrt{3}} \frac{1}{\sqrt{2f(x)}} K(\sqrt{\frac{g(x)}{2f(x)}})$ for $-1 \leqslant x < 3$, where $f(x) = 2\sqrt{2x+3}$, $g(x) = 3 - x^2 + 2\sqrt{2x+3}$, and $K(m)$ is the complete elliptic integral of the first kind.

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