

Effects of long-ranged interactions on the nucleation dynamics in a ϕ^6 model

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(Received 27 March 1996)

It is known that long-ranged interactions can play an important role in determining the dynamics of nucleation at a first-order phase transition. For example, in a recent Letter by the present authors [Phys. Rev. Lett. **74**, 3848 (1995)] it was shown that during the annealing of an amorphous thin film, long-ranged interactions generated by strains due to elastic misfits could be responsible for the system becoming trapped in a metastable secondary crystalline phase. In this paper we consider these long-ranged interactions in a more general context wherein only symmetry considerations are invoked to determine if a particular interaction potential is allowed. Their effects on a first-order phase transition, as described by a single-component scalar ϕ^6 model, are examined. These transitions are studied by means of Langevin simulations. We find, through the evaluation of the time evolution of the order parameter, the morphology of the steady state, and the time-dependent structure factor, that the precise form of the long-ranged interaction has a strong influence on the nature of nucleation dynamics, as well as on the steady-state profiles. [S1063-651X(96)09408-1]

PACS number(s): 64.60.Cn, 64.70.Dv

I. INTRODUCTION

When a system initially in thermal equilibrium is quenched into a metastable state, by a change in some external control parameter such as the temperature, it equilibrates via the nucleation of the new stable phase. There is a competition between the lowering of the free energy with the formation of the stable phase and the energy expense due to the interfaces that necessarily arise between regions of the metastable phase and the newly formed stable phase. Because of this energy balance, a nucleus generated by thermal fluctuations must exceed a certain critical size in order to grow. In many instances this process is well described by classical nucleation theory [1].

In an interesting series of papers Littlewood and Chandra [2,3] showed that the dynamics of nucleation can be strongly affected by the presence of long-ranged interactions, as arising, for example, from strain misfits of inhomogeneous phases. Their considerations were motivated by the experiments of McWhan *et al.* [4] on barium titanate (BaTiO_3), whereby a pulsed electric field was used to cross the paraelectric (PE)–ferroelectric (FE) phase boundary. McWhan *et al.* observed a time delay in the rise of the spontaneous polarization after the onset of the electric field, indicating a delay in the nucleation of the ferroelectric phase after field quenching. This behavior is not described by classical nucleation theory, which predicts a smooth transformation to the FE phase without delay. In the actual experiments, the delay in nucleation was six orders of magnitude larger than the characteristic transformation time according to classical theory [3]. Littlewood and Chandra suggested that this phenomenon may be associated with long-ranged strain interactions. Since the PE phase of BaTiO_3 has a cubic crystal structure, while the FE phase is tetragonal, spontaneous polarization implies a spontaneous strain. The strong coupling between the strain and polarization order parameters acts to suppress the effect of the external electric field. Initially, therefore, the critical size R_c required for the growth of a nucleus of the stable phase is very large. Nucleation is de-

layed as the system needs to wait for thermal fluctuations of the length scale R_c to occur. However, once a critical nucleus is formed and begins to grow, the effective bias field is increased (see below), reducing R_c and aiding further nucleation and growth. This feedback mechanism leads to a sharp transition to the stable phase.

Because of the long-ranged nature of strain interactions, the energy penalty for elastic misfits is proportional to the volume of the transformed phase. Littlewood and Chandra thus included these interactions in their model free energy in a mean-field manner, which corresponds to the infinite-ranged limit.

A similar interaction of this type has also been considered by Fisher [5] in the different context of a charge-density wave (CDW) pinned by impurities. In this case, the coupling between the local CDW phases $\{\phi_i\}$ represents the elasticity of the charge-density wave that favors a uniform phase. Fisher studied a time-dependent mean-field theory of the problem, in which each local phase ϕ_i is coupled to ϕ , the average phase over the whole system. In other words, the interactions between the phases are again approximated by an infinite-range model.

More recently, the present authors [6] showed that long-ranged interactions, as generated by elastic misfits of inhomogeneous crystalline phases, are crucial in understanding the effects of rapid thermal annealing on the crystallization of lead zirconate titanate (PZT) thin films. For PZT, apart from the ferroelectric perovskite phase, which is in fact the true ground state, there exists also a secondary metastable defect-pyrochlore phase. The competition between the two crystalline phases is enhanced by long-ranged elastic strains, with the consequence that the steady state reached after a heat treatment of the amorphous PZT film depends strongly on the initial heating rate [7]. In particular, for slow heating, as in conventional furnace annealing, long-ranged forces are responsible for the system being trapped in the metastable defect-pyrochlore phase. Similar behaviors have also been observed in lead iron niobate thin films [8].

In this paper we investigate, via a Landau theory and

utilizing Langevin simulations, the dynamics of crystal nucleation and growth when long-ranged interactions are present. Here, such interactions are, as in the aforementioned studies, included in a mean-field manner. However, more general forms of the interactions, which fulfill system-specific symmetry requirements, will be considered. We demonstrate how the specific form of the long-ranged interactions can influence the steady state of a model system for which there exists a doubly degenerate ground state, as is found in ϕ^6 models.

Our paper is organized as follows. In Sec. II a description of the Landau free energy that we have studied will be given, together with a discussion of the physical meaning of the order parameter and the symmetry requirements of our model system. The principal numerical results will be presented in Sec. III, followed by a summary of the physically relevant conclusions in Sec. IV.

II. MODEL

In constructing a phenomenological Landau free energy that describes any particular phase transition, one is constrained in that it must be consistent with the symmetries of the system of concern. In a Landau theory of solidification and crystal nucleation [9–11], the free energy can be written as an expansion in the coefficients of the Fourier components of the solid phase density $\phi_{\mathbf{q}}$:

$$\mathcal{F} = \sum_{\mathbf{q}} a_{\mathbf{q}} \phi_{\mathbf{q}} \phi_{-\mathbf{q}} + \sum_{\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3} a_{\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3} \phi_{\mathbf{q}_1} \phi_{\mathbf{q}_2} \phi_{\mathbf{q}_3} \times \delta(\mathbf{q}_1 + \mathbf{q}_2 + \mathbf{q}_3) + \dots, \quad (2.1)$$

where the \mathbf{q}_i 's are reciprocal lattice vectors of the solid. Because of rotational symmetry, the coefficient a 's depend only on the magnitudes $|\mathbf{q}_i|$. As explained by Alexander and McTague [9] and Shih *et al.* [11], near a phase transition the coefficients $a_{\mathbf{q}}$, $a_{\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3}$, \dots will have minima at some wave vector $|\mathbf{Q}|$ so that the free energy \mathcal{F} may be truncated to include only a single set of nonzero reciprocal lattice vectors of equal length. The dominant set is generally the smallest nonzero set [11]. Furthermore, in order to obtain a solid with the correct point-group symmetry, all the magnitudes of the Fourier components belonging to the dominant set must be identical, i.e., $\phi_{\mathbf{Q}_i} = \phi_{\mathbf{Q}}$. Hence the free energy can be expanded in a single order parameter $\phi_{\mathbf{Q}}$. The powers of $\phi_{\mathbf{Q}}$ that appear in the expansion can be deduced by considering geometrically whether it is possible to construct, with the wave vectors \mathbf{Q}_i , a closed polyangle (which may be nonplanar) with the corresponding number of sides. For the bcc crystal, for example, one must consider [9–11] a free-energy expansion consisting of quadratic, cubic, and fourth-order terms.

In the cases of the simple cubic and fcc crystals, however, both cubic and fifth-order terms are absent in such an expansion. Only even powers are allowed. Physically, this is because the conjugates of these two lattices, which correspond to a change in the sign of $\phi_{\mathbf{Q}}$, are identical to the lattices themselves, differing only by a simple translation. Symmetry of the free energy thus excludes odd powers of the order parameter. The bcc crystal, however, is distinct from its con-

jugate, permitting a cubic term in the free-energy expansion. For simple cubic and fcc crystals, to obtain a free energy that describes a first order transition [12], one must therefore include terms up to sixth order. In the present investigation, we focus on a “2-4-6” local potential, appropriate for these two crystal structures. More specifically, we have studied a coarse-grained free energy (already scaled) given by

$$\mathcal{F}[\phi] = \sum_i \left\{ \frac{1}{2} a \phi_i^2 - \frac{1}{4} \phi_i^4 + \frac{1}{6} \phi_i^6 + \frac{K}{2} |\nabla \phi_i|^2 \right\} + V_{\text{int}}, \quad (2.2)$$

where the index i denotes discrete sites on a $L \times L$ lattice. This is applicable to the crystallization of a thin film from the amorphous phase.

Another motivation for studying this potential is as follows. Our local Landau free energy employs a single-component, scalar order parameter. In the case of $\mathcal{F}[\{\phi_i\}] = \mathcal{F}[-\phi_i]$, this is the simplest phenomenological theory by which we can study symmetry-breaking first-order transitions.

In the free energy Eq. (2.2), a squared gradient term, with K being constant, is included to account for the energy cost of a spatially inhomogeneous order parameter. The last term in Eq. (2.2) represents a long-ranged interaction and is given by the general form

$$V_{\text{int}} = \frac{l}{2} \sum_{i,j} \beta_{ij} \phi_i^m \phi_j^n. \quad (2.3)$$

In this equation, l is a coupling constant, while m and n are integers. The symmetry $\mathcal{F}[\{\phi_i\}] = \mathcal{F}[-\phi_i]$ leads to the constraint that $m+n$ must be even.

For the coupling parameter β_{ij} in Eq. (2.3) we have considered

$$\beta_{ij} = \delta_{ij} - 1/L^2, \quad (2.4)$$

where δ_{ij} is the Kronecker delta. The second (constant) term in this definition signifies an infinite-ranged interaction and its magnitude ensures that the free energy \mathcal{F} is properly extensive. Since V_{int} represents some generalized strain energy due to elastic misfits, its value should be zero in the homogeneous system, i.e., when $\{\phi_i\} = \phi$ for all i . The δ_{ij} term is included in Eq. (2.4) to this effect. That this is so can be made apparent by substituting Eq. (2.4) in Eq. (2.3) to obtain

$$V_{\text{int}} = \frac{l}{2} L^2 [\overline{\phi^{m+n}} - (\overline{\phi^m})(\overline{\phi^n})], \quad (2.5)$$

where $\overline{\phi^m} = (1/L^2) \sum_i \phi_i^m$ is the spatial average of m th power of the order parameter. In the homogeneous case, Eq. (2.5) is identically zero.

In the present study we limit ourselves to the study of $m=n$. In this case Eq. (2.3) can be rewritten in the form

$$V_{\text{int}} = \frac{l}{2} \sum_i (\phi_i^n - \overline{\phi^n})^2, \quad (2.6)$$

which for $n=1$ represents exactly the same mean-field long-ranged interactions considered by previous authors [2,3,5,6]. Of course, the local Landau potentials used in these other

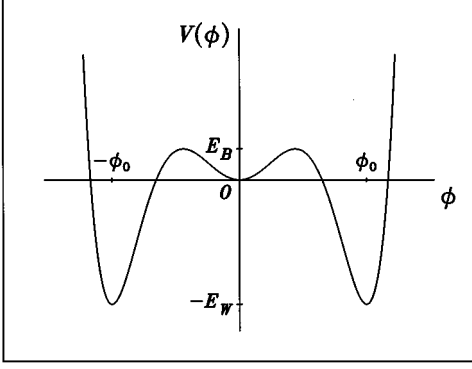


FIG. 1. Local potential in the Landau free energy Eq. (2.2). In terms of the parameter a , $\phi_0 = \sqrt{(1 + \sqrt{1 - 4a})/2}$ and $E_B/E_W = \{-1 + 6a + (1 - 4a)^{3/2}\} / \{1 - 6a + (1 - 4a)^{3/2}\}$. The potential shown corresponds to $a = 0.135788$, for which $E_B/E_W = 1/4$.

studies are different from that being considered here. In this paper we will demonstrate that an $n = 1$ interaction leads to a very different nucleation and growth behavior than for $n = 0$ (i.e., absence of long-ranged interaction) or $n = 2$; more generally, an odd n leads to a very different behavior than an even n .

We focus on the case where the parameter a in Eq. (2.2) lies in the range $0 < a < 3/16$, for which the local on-site potential has a metastable local minimum at $\phi = 0$, and two degenerate global minima located at $\phi = \pm \phi_0$ (Fig. 1). Physically, the central minimum corresponds to the amorphous phase, while the global minima at nonzero ϕ represent the crystalline solid. Because of the energy barrier E_B , in the absence of thermal fluctuations a system initially localized in the metastable state will remain trapped in this state. Thermal fluctuations, therefore, play an important role in the dynamical evolution towards steady state. Such dynamics can be generated according to a Langevin equation

$$\frac{\delta \phi_i}{\delta t} = -\Gamma \frac{\delta \mathcal{F}}{\delta \phi_i} + \eta_i, \quad (2.7)$$

where Γ is a kinetic coefficient and η_i is a Gaussian thermal noise with zero mean satisfying the ensemble average

$$\langle \eta_i(t) \eta_j(t') \rangle_{\text{ens}} = 2\Gamma k_B T \delta_{ij} \delta(t - t'). \quad (2.8)$$

Simulations are performed by averaging over a number of runs, each of which constitutes a different sequence of the random thermal noise. In Eq. (2.8) the ensemble average $\langle \rangle_{\text{ens}}$ signifies a combination of averaging over separate runs and over the entire lattice. In the following averages over runs will be represented by $\langle \rangle$, while averages over the lattice will be denoted by an overbar $\overline{}$.

Equation (2.7) may be solved numerically by the Euler method [13,14]. In this algorithm time is discretized into steps of the size δt . We have used $\delta t = 0.1$ and found that this time step gives numerically converged results. Substituting the free energy $\mathcal{F}[\phi]$, as given by Eq. (2.2), into the Langevin equation Eq. (2.7) and rescaling the variables appropriately, we obtain

$$\phi_i(t + \delta t) = \phi_i(t) + \delta t \left\{ a \phi_i - \phi_i^3 + \phi_i^5 + K \sum_{\mathbf{a}} (\phi_{i+\mathbf{a}} - \phi_i) - \ln \phi_i^{n-1} (\phi_i^n - \overline{\phi^n}) \right\} + \sqrt{2\delta t} \xi_i(t), \quad (2.9)$$

where \mathbf{a} is a nearest-neighbor lattice vector and $\xi_i(t)$ satisfies

$$\langle \xi_i(t) \xi_j(t') \rangle_{\text{ens}} = \epsilon \delta_{ij} \delta_{tt'}. \quad (2.10)$$

Here ϵ is the scaled dimensionless temperature.

III. SUMMARY OF RESULTS AND ANALYSIS

In this section we present a summary of our extensive numerical results obtained from Langevin simulations. Representative data are found for simulations performed on a 128×128 lattice with periodic boundary conditions. For the free energy Eq. (2.2) we have chosen the parameter $a = 0.135788$, for which the ratio of the energy barrier to the energy minimum of the local potential is $E_B/E_W = 1/4$ (Fig. 1). In what follows, all references to the energy barrier E_B correspond to this particular value of a .

Since we wish to primarily address the issue of crystallization from an amorphous phase we have focused on initial conditions in which the whole system is localized in the central energy minimum, i.e., $\{\phi_i\} = 0$ for all i . In a previous study of a similar model, Valls and Mazenko [13] have considered initial conditions given by a random distribution of the order parameters $\{\phi_i\}$. We have also applied these initial conditions in our present system, even though they are not physically relevant in the present context. For either of these conditions, the system is allowed to evolve according to Langevin dynamics, as given by Eqs. (2.9) and (2.10), with the temperature being fixed at a constant value ϵ . Here we note that the random initial conditions lead to similar intermediate and late-time dynamics as the first type of initial conditions.

Starting with the initial state of $\{\phi_i\} = 0$ and averaging over 20 runs we obtained the dynamical evolutions shown in Figs. 2(a) and 2(b). For these simulations the coupling constant of the squared gradient term is chosen to be $K = 6E_B$ and a temperature of $\epsilon = 0.5E_B$ is used, with E_B being the energy barrier in the absence of long-ranged forces. Figure 2(a) indicates a rapid initial rise in the root mean square order parameter $\sqrt{\langle \phi^2 \rangle} / \phi_0$ from the starting value of zero, both in the absence and presence of long-ranged forces. This transient behavior corresponds to the initial thermalization about $\phi = 0$, which does not involve overcoming any energy barrier. The early stages that follow immediately are governed by the activation over the energy barrier separating the metastable state and the global minima. The figure demonstrates a delay in nucleation of the stable phase when long-ranged interactions are present, as reported in previous studies [2,3,6]. Figure 2(b) shows a different average $\mathcal{M} = \langle |\overline{\phi}| \rangle / \phi_0$. This quantity is computed by first averaging the order parameter $\{\phi_i\}$ over the lattice for each run, then evaluating its absolute value, followed by an average over separate runs. The behavior for zero and $n = 2$ long-ranged forces is very similar, in that the long-time averages are small. In fact, from these results, together with simulations

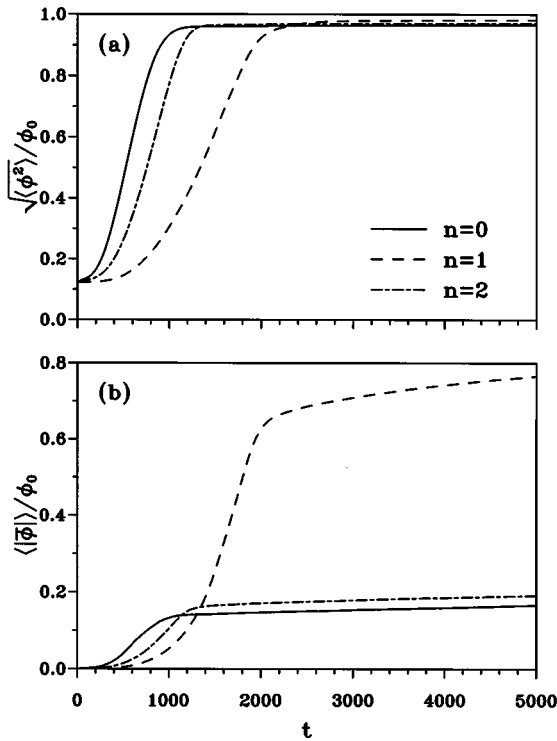


FIG. 2. Time evolution of averages (a) $\sqrt{\langle \phi^2 \rangle} / \phi_0$ and (b) $\langle |\bar{\phi}| \rangle / \phi_0$ obtained for $n=0$ (—), $n=1$ (---), and $n=2$ (-·-) long-ranged interactions. These data correspond to $a=0.135788$, $K=6E_B$, and $\epsilon=0.5E_B$. For both $n=1$ and $n=2$, the strength of the long-ranged interaction is $l=2E_B$.

on 32×32 and 64×64 lattices, we find that in the limit of an infinite lattice size, the long-time average \mathcal{M} approaches zero for these two types of interactions. However, the behavior for $n=1$ interactions is qualitatively different. In this case a long-time average of \mathcal{M} approaching 1 is obtained. Combined with the root mean square averages, these results indicate that for $n=1$ a single domain consisting entirely of either the positive or negative phase is obtained at long times, while for the other two cases the steady state is a mixed positive and negative phase.

Figure 3 shows the evolution of the order parameter configuration $\{\phi_i\}$ obtained for a typical run for the $n=1$ interaction. In the figure, the occupation of the three potential wells centred on $\phi = -\phi_0$, 0 , and $+\phi_0$ are indicated by black, gray, and white pixels, respectively. Initially, nuclei of both the positive and negative phases are formed. At long times, however, the system evolves into a single phase. The long-time configurations obtained for the zero and $n=2$ long-ranged interactions are shown in Figs. 4(a) and 4(b) (we find no finite-size effects in this result, at least for lattices as large as 256×256 , the largest we have studied). The figures show that the steady-state morphology for $n=2$ interactions is indeed very similar to that for the absence of long-ranged interactions ($n=0$), both corresponding to a mixed state with coexisting domains of the two degenerate low-energy phases.

These late time results may be understood by examining how the long-ranged contribution modifies the local potential in the Landau free energy Eq. (2.2). For $n=1$, expanding

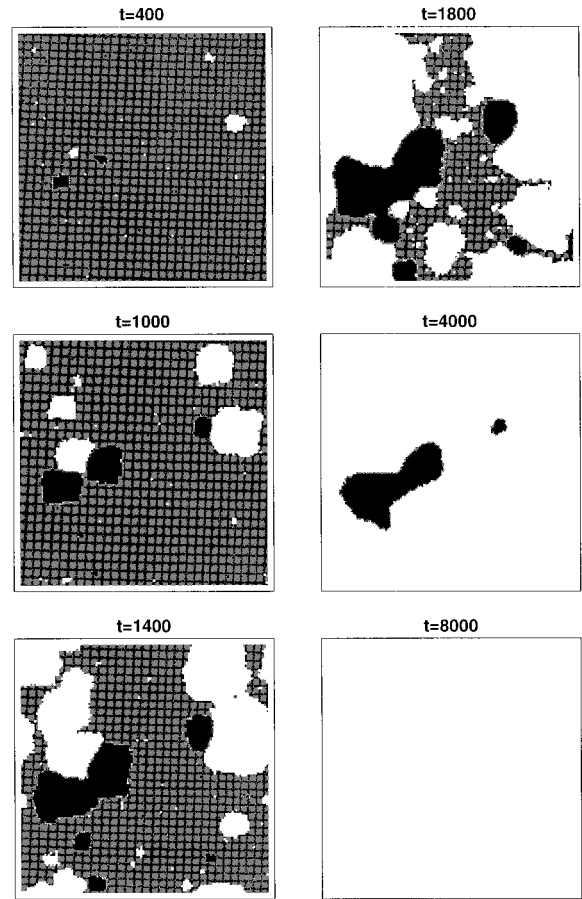


FIG. 3. Evolution of the $\{\phi_i\}$ configuration obtained for the $n=1$ long-ranged interaction, with the same parameters as used in Fig. 2. The occupation of the three local minima is presented by the shading of the pixels as follows: black ($\phi = -\phi_0$), gray ($\phi = 0$), and white ($\phi = +\phi_0$).

V_{int} and substituting in Eq. (2.2), we obtain

$$\mathcal{F} = \sum_i V_i + \frac{K}{2} |\nabla \phi_i|^2, \quad (3.1)$$

in which the on-site potential is given by

$$V_i = -l\bar{\phi}\phi_i + \frac{1}{2}(a+l)\phi_i^2 - \frac{1}{4}\phi_i^4 + \frac{1}{6}\phi_i^6, \quad (3.2)$$

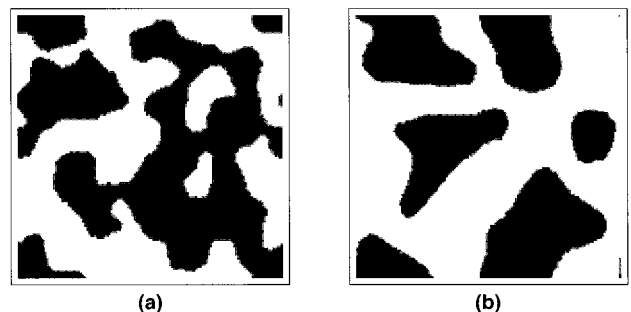


FIG. 4. Typical late-time $\{\phi_i\}$ configurations obtained for $E_B/E_W = \frac{1}{4}$, $K=6E_B$, and (a) $n=0$ (at $t=2000$) and (b) $n=2$, $l=2E_B$ (at $t=6000$).

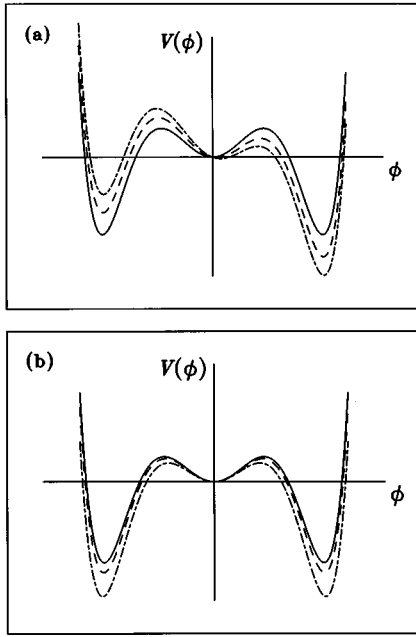


FIG. 5. Local potential modified in the presence of (a) $n=1$ and (b) $n=2$ long-ranged interactions. In (a), the curves correspond to $\bar{\phi}=0$ (—), 0.5 (---), and ϕ_0 (- - -), while in (b), $\bar{\phi}^2=0$ (—), 0.25 (---), and ϕ_0^2 (- - -).

where we have dropped a term that pertains to a global shift in the free energy. The long-ranged contribution thus renormalizes the second-order term of the on-site potential. In addition, it generates a first-order term that is equivalent to an effective bias field whose magnitude is proportional to the spatial average of the order parameter. Figure 5(a) depicts the modified on-site potential, for the values of the parameters that we have used ($a=0.135788$ and $l=2E_B$). In this case, if the mean order parameter $\bar{\phi}$ is zero, then the on-site potential has an increased energy barrier. This accounts for the delayed formation of critical nuclei of the stable phase. In the event that the random thermal fluctuations drive the average $\bar{\phi}$ to a nonzero value, the resulting effective bias field will tilt the on-site potential so that the double degeneracy of the global minima is lifted, as shown in the figure. Moreover, the energy barrier is lowered in the direction of the tilt, thus aiding further transitions to the low-energy phase. This effect can be seen in the $\{\phi_i\}$ configurations shown in Fig. 3. In this particular example, fluctuations first produce an excess of the positive stable phase over the negative one. It is apparent from the figure that nucleation and growth of the negative phase is suppressed, due to the effective bias. On the other hand, nucleation of the positive phase is enhanced. Eventually, the domains of the negative phase are swallowed up completely by the dominant positive phase. One might question whether this steady-state morphology is a finite-size effect, incurred because fluctuations do not produce exactly equal amounts of the positive and negative phases in a finite system. However, the nature of the effective bias field in Eq. (3.2) is that even for an infinitesimal excess of one phase over the other, the degeneracy of the potential is lifted, leading to broken symmetry. In our simulations of up to 256×256 lattices for the $n=1$ interaction, a single phase is always obtained at long times.

For $n=2$ the on-site potential becomes

$$V_i = \frac{1}{2} (a - 2l\bar{\phi}^2) \phi_i^2 - \frac{1}{4} (1 - 2l) \phi_i^4 + \frac{1}{6} \phi_i^6. \quad (3.3)$$

In this case the long-ranged term renormalizes the coefficients of the second and fourth powers in V_i . Unlike the $n=1$ case, there is no additional first-order term. Because the structure of the on-site potential is unchanged from that when long-ranged interactions are absent [Fig. 5(b)], in that the two global minima remain degenerate, the steady state is again a mixed phase, the same as for $n=0$.

In general, for an odd value of n in Eq. (2.6), the long-ranged interaction generates the odd-power terms $l\bar{\phi}^n \phi_i^n$ in the Landau free energy. In the event of thermal fluctuations causing $\bar{\phi}^n$, the n th moment of the order parameter, to deviate from zero, such additional terms lift the degeneracy of the local potential. In this case, it is expected that the system will evolve towards a single phase, as observed for $n=1$. However, for any even n , all additional terms comprise of even powers of the order parameter. Therefore, the symmetry of the local potential is always preserved and a mixed phase is expected.

Another quantity of interest [15], one that provides quantitative information of characteristic domain sizes, is the time-dependent structure factor $S(\mathbf{k}, t)$. This quantity is defined as

$$S(\mathbf{k}, t) = \left\langle \left| \frac{1}{L^2} \sum_i e^{i\mathbf{k} \cdot \mathbf{r}_i} [\phi_i(t) - \bar{\phi}(t)] \right|^2 \right\rangle / \phi_0^2 = \langle \phi_{\mathbf{k}} \phi_{\mathbf{k}}^* \rangle, \quad (3.4)$$

where

$$\phi_{\mathbf{k}} = \frac{1}{L^2} \sum_i \exp(i\mathbf{k} \cdot \mathbf{r}_i) [\phi_i(t) - \bar{\phi}(t)] / \phi_0 \quad (3.5)$$

is the Fourier transform of the deviation of ϕ_i from the average over the whole lattice. We compute the circularly averaged structure factor, which is given by

$$S(k, t) = \frac{\sum_{\mathbf{k}}' S(\mathbf{k}, t)}{\sum_{\mathbf{k}}' 1}, \quad (3.6)$$

where the summations are over \mathbf{k} such that $k - \pi/L < (k_x^2 + k_y^2)^{1/2} \leq k + \pi/L$. The time evolution of this quantity is plotted in Fig. 6, for $n=0, 1$, and 2 , as obtained from the same sets of simulation that give the average order parameters in Fig. 2. From $S(\mathbf{k}, t)$, the correlation function $C(\mathbf{r}, t)$ can be calculated by taking its inverse Fourier transform. A further circular average leads to the radial correlation function $C(r)$ shown in Fig. 7.

In Fig. 7, for the cases of $n=0$ and 2 , the plots show that there is a monotonic increase in the spatial correlation $C(r)$ with time. For $n=1$ interaction, however, a different behavior is observed. It appears from the figure that although the amplitude of $C(r)$ increases with time initially, the trend is reversed at long times. Quite simply, for this case, since the system has fully evolved into a single phase and the

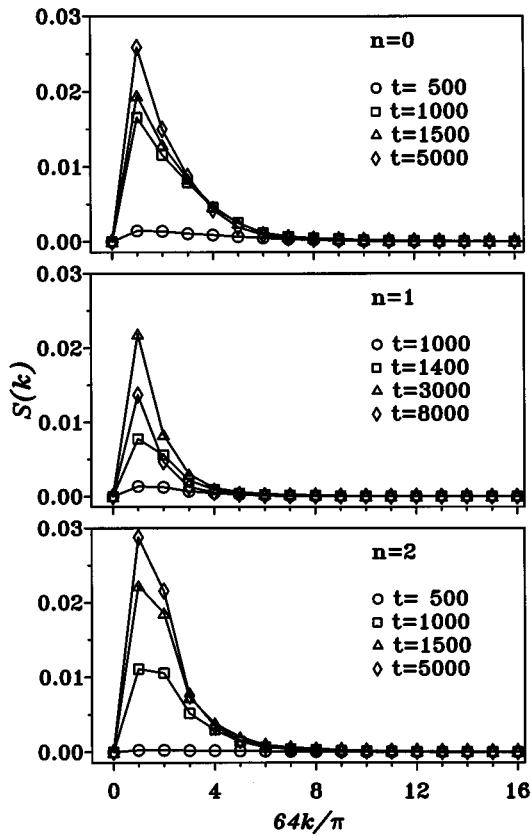


FIG. 6. Time evolution of the circularly averaged structure factor $S(k)$, obtained for $n=0$, $n=1$ ($l=2E_B$), and $n=2$ ($l=2E_B$) interactions.

energy barrier to escape the global minimum is so much larger than the temperature, we find a $C(r)$ and $S(k)$ indicative of a harmonic system with strong intersite coupling. It should be noted that it is the amplitude of $C(r)$, the correlation of the fluctuations of the order parameter from the average, that exhibits a decrease with time and not its spatial extent.

A further investigation of the dynamics of the nucleation and growth process is provided by a calculation of the characteristic length scale or domain size of the system. We define this as the inverse of the first moment of the circularly averaged structure factor [16], which is given by

$$k_1(t) = \frac{\sum_k k S(k, t)}{\sum_k S(k, t)}, \quad (3.7)$$

with the summation being truncated at $k = \pi$. The characteristic length scale is then $R(t) = 1/k_1(t)$. A plot of $\ln(R)$ vs $\ln(t)$ is shown in Fig. 8 for intermediate and late times. Very similar behavior is found for $n=0, 1$, and 2 , irrespective of whether the final state is a single or mixed phase. In all three cases there is a regime of rapid increase of R with time, representing the nucleation and growth of the stable phases. This is followed by a very weak power-law behavior, $R \sim t^n$, corresponding to the smoothing of domain boundaries and a swallowing up of any subcritical nuclei. However, we

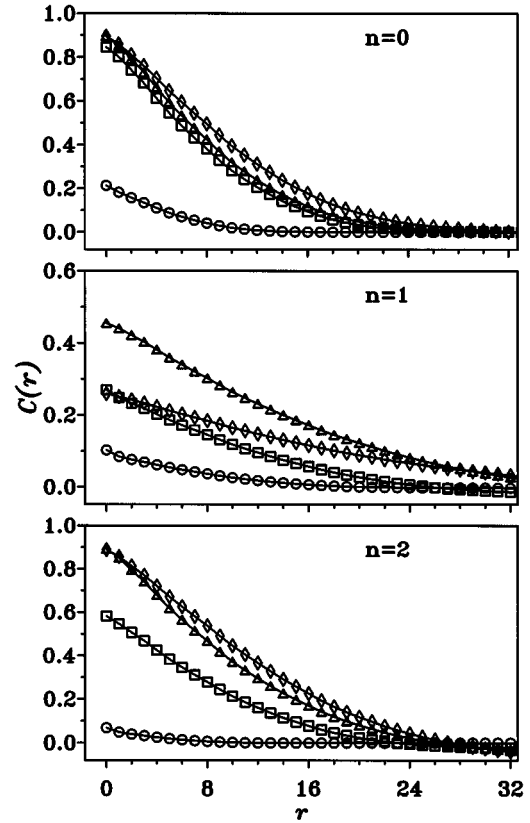


FIG. 7. Evolution of the radial correlation function $C(r)$ for $n=0, 1$, and 2 interactions. The symbols in this figure correspond exactly to those of Fig. 6.

do not observe any regime characterized by the coarsening of domains, for which a growth exponent $n = \frac{1}{2}$ is expected [17,18]. Clearly, these results deserve further analytical and numerical study.

IV. CONCLUSION

Utilizing Langevin simulations we have studied the dynamics of nucleation and growth as described by a single-component scalar ϕ^6 model with the addition of long-ranged interactions. Physically, the 2-4-6 local potential that we

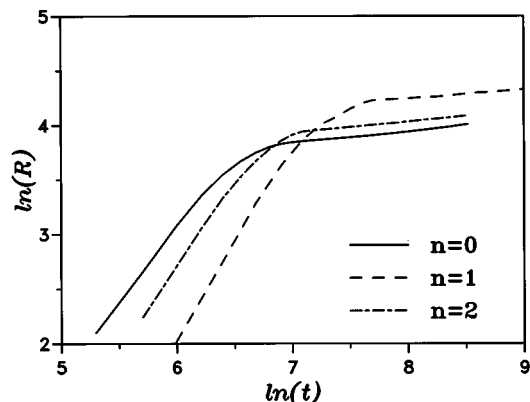


FIG. 8. $\ln R$ vs $\ln t$, where R is the characteristic length scale, for $n=0, 1$, and 2 .

have examined is appropriate as a description of the crystallization of simple cubic or fcc thin films from the amorphous phase. A general form for the long-ranged interaction, consistent with the symmetry of these two crystal structures, is investigated. Such interactions may be generated by strains due to elastic misfits in the crystallized film. We find that the precise form of the long-ranged interactions has a strong influence on the steady-state profiles, as in whether a pure

phase or a mixed phase consisting of two degenerate ordered domains of the crystalline phase is obtained.

ACKNOWLEDGMENTS

We would like to thank Ken Elder for helpful discussions. This work was supported by the Ontario Centre for Materials Research, an Ontario Centre for Excellence.

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