

Dynamics of first-order transitions in two-dimensional systems with long-range interactions

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The dynamics of first-order phase transitions in two-dimensional Ising systems with long-range interactions has been studied by means of numerical simulations using the Metropolis Monte Carlo method. The kinetics of domain growth was examined at early times for spinodal decomposition for both conserved and nonconserved order parameters; the resultant behavior was found to be in agreement with linear theory, as predicted by Binder. The late stages of domain growth for nonconserved order parameters were also studied and the results were in agreement with the Allen-Cahn growth law.

I. INTRODUCTION

The kinetics of first-order transitions has been the subject of many recent investigations.¹ The principal theoretical model used for these investigations is the Ising model with nearest-neighbor interactions for which both nucleation and spinodal decomposition have been studied. In the studies relating to spinodal decomposition the system is usually quenched rapidly from a single phase disordered state at infinite temperature to an unstable state below the ordering temperature T_c . The system then orders kinetically via the formation of small domains and their growth to macroscopic size. Studies of spinodal decomposition have been undertaken for both conserved order parameters, in which case the kinetic ordering process is referred to as phase separation, and nonconserved order parameters for which the kinetic ordering process describes an order-disorder transition. The early-time regime leading to the formation of domains is described by a theory due to Cahn, Hilliard, and Cook.²⁻⁴ For example, for the case of a conserved order parameter, these authors predict the occurrence of a long-wavelength instability with exponentially growing modes for wave numbers less than a critical wave number, k_c . Their theory is basically a linear stability analysis of the equations of motion for spinodal decomposition about the state of the system immediately after the quench.

In this paper we examine the effect of long-range microscopic interactions on the kinetics of spinodal decomposition. We shall be especially concerned with the early stages. The experimental systems most relevant to our study include polymer blends whose properties involve long-range interactions.⁵⁻⁷ It should also be noted, however, that early-time regimes of systems with short-range interactions can also now be studied since experiments can be performed to high resolution in time.⁸ Furthermore, crossover effects between early- and late-stage dynamics can be studied experimentally.

In the initial stages of spinodal decomposition, long-range interactions inhibit the first-order transition and allow the system to remain uniform and become unstable. Nevertheless, since phase separation is driven by a long-wavelength instability, the uniform disordered state eventually breaks up as domains appear and grow. However, for sufficiently early times in a system with long-range interactions, one should be able to observe the linear instability. Binder⁹ has made this precise by noting that any ordering structure must be negligibly small for the linear regime to be valid; systems with long-range interactions can therefore exhibit a linear regime whose duration increases with the range of interaction. His predictions are discussed in more detail later. As mentioned above, long-chain molecules such as polymer blends are therefore ideal for experimental studies of early-stage spinodal decomposition. In particular, some aspects of recent light-scattering experiments on these systems have been interpreted in terms of linear theory.⁵⁻⁷ It should be mentioned that these experiments do not address the issue of whether or not a linear theory exists, at sufficiently early times, for systems with short-range interactions.

Metropolis Monte Carlo simulations on three-dimensional Ising systems with long-range interactions have previously been performed by Heermann¹⁰ for conserved order parameters and his results were in reasonable agreement with linear theory. Recently, Elder, Rogers, and Desai¹¹ solved the Cahn-Hilliard-Cook equation numerically using a discretized scheme for square lattices with conserved order parameters. Their results were also in agreement with linear theory at sufficiently early times and they showed the duration of the linear regime depends logarithmically on the range of interaction, in agreement with Binder's predictions.⁹

Both Heermann and Elder, Rogers, and Desai studied spinodal decomposition for conserved order parameters. However, linear theory should also hold at sufficiently early times for a system with long-range interactions where the order parameter is nonconserved. In addition,

the critical wave number, for a given range of interaction, should be identical in both situations, since only the dynamical aspects of the phase transitions are different, not the local quasiequilibrium aspects. We therefore studied the early stages of spinodal decomposition in systems with both conserved and nonconserved order parameters; comparing these gives us insight into the unstable dynamics. Also, by examining two-dimensional systems we generalize Heermann's three-dimensional results, and by using an Ising model with long-range interactions, we are able to study a more microscopic model than the explicitly coarse-grained Langevin model considered by Elder, Rogers, and Desai. Finally a study of the late stages of order-disorder transitions is made during the time regime when dynamics is dominated by domain growth in order to investigate the role played by the range of interaction.

The results¹² are presented in Sec. II. The early stages of spinodal decomposition are considered in Sec. II A. A linear regime for systems with sufficiently long-range interactions is observed regardless of whether or not the order parameter is conserved. The critical wave number is found to be independent of the specific dynamics to the accuracy of the calculations. Section II B contains a study of the late stages of growth. In this regime, a long-range interaction only appears to affect nonuniversal amplitudes. No study has been made of the late stages of phase separation due to limitations of computer time. Finally, in Sec. II C we conclude the paper with a summary of our results.

II. METHOD AND RESULTS

A. Early stages

We studied the two-dimensional ferromagnetic Ising model with long-range interactions on a square lattice by Monte Carlo computer simulation. The Hamiltonian is,

$$H = - \sum_{i,j} J_{ij} \sigma_i \sigma_j ,$$

where $\sigma_i = \pm 1$ are the N local spins on sites i , and J_{ij} is the coupling constant. It is taken to be a positive constant over a diamond shaped area centered on i , and equal to zero if j is outside the diamond,¹³ i.e.,

$$J_{ij} = \begin{cases} \text{const} & \text{if } j \text{ is inside the diamond,} \\ 0 & \text{otherwise.} \end{cases}$$

The number of spins interacting with the central spin in the diamond is R^2 , from which we obtain the range of interaction R . (Thus the usual nearest-neighbor-interaction Ising model corresponds to $R \equiv 2$.) The system is prepared at infinite temperature by taking a random initial configuration, and then quenched to a temperature below T_c . The critical temperature is given by¹⁴ $T_c = R^2 J$, as $R \rightarrow \infty$. We consider symmetric quenches, where the initial mean order parameter $\psi(t=0) \equiv (1/N) \langle \sum_i \sigma_i \rangle$ is equal to zero. In the case of a nonconserved order parameter, the system evolves by spin-flip dynamics, and for a conserved order parameter,

by spin-exchange dynamics. As a measure of increasing order following the quench, we calculated the structure factor, $S(\mathbf{k}, t)$, which is the Fourier transform of the real space pair-correlation function:

$$S(\mathbf{k}, t) = \left\langle \frac{1}{N} \left| \sum_{\mathbf{x}_i} \sigma(\mathbf{x}_i, t) e^{i\mathbf{k} \cdot \mathbf{x}_i} \right|^2 \right\rangle ,$$

where $\mathbf{k} = 2\pi/(\sqrt{N})(ni + m\mathbf{j})$, and $m, n = 0, 1, \dots, \sqrt{N}$. Our results are given in terms of the circularly averaged structure factor $S(k, t) = \sum' S(\mathbf{k}, t) / \sum' 1$, where \sum' denotes a sum over a circular shell defined by $n - \frac{1}{2} \leq |\mathbf{k}| \sqrt{N} / (2\pi) < n + \frac{1}{2}$.

Predictions for the early stages are in terms of the coarse-grained Langevin description. The motion of the local order parameter $\psi(\mathbf{x}, t)$ (which is the local magnetization of the previously mentioned spin system) can be described by the following Langevin equation:

$$\frac{\partial \psi}{\partial t} = -M(-\nabla^2)^a \frac{\delta F}{\delta \psi(\mathbf{x}, t)} + \eta(\mathbf{x}, t) , \quad (1)$$

where $a=1$ for phase separation where the order parameter is conserved, while $a=0$ for an order-disorder transition where there is a nonconserved order parameter. The variable, M , in Eq. (1) is the mobility, η is a Gaussian thermal noise which satisfies the fluctuation-dissipation relation,

$$\langle \eta(\mathbf{x}, t) \eta(\mathbf{x}', t') \rangle = 2TM(-\nabla^2)^a \delta(\mathbf{x} - \mathbf{x}') \delta(t - t') ,$$

and Boltzmann's constant has been set to unity. The coarse-grained free-energy functional F can be written as

$$F[\psi] = \int d^d x \left[\frac{C}{2} |\nabla \psi|^2 - \frac{|r|}{2} \psi^2 + \frac{u}{4} \psi^4 \right] , \quad (2)$$

where $f = -(|r|/2)\psi^2 + (u/4)\psi^4$ is the bulk free-energy density.

Equation (1) is difficult to solve due to the quartic non-linearity in ψ . This dominates the dynamics in the late stages of growth and gives rise to a complicated domain morphology. In the very early stages of growth, it is expected that one can neglect this term (at least for the coarse-grained Langevin model) and linearize around the initial value of the order parameter, namely $\psi(t=0)=0$. This gives rise to the linear Langevin equation,

$$\frac{\partial \psi}{\partial t} = M(-\nabla^2)^a [|r| + \nabla^2] \psi + \eta(\mathbf{x}, t) .$$

Fourier transforming this, one readily obtains the structure factor

$$S(\mathbf{k}, t) = \int d^d x e^{i\mathbf{k} \cdot \mathbf{x}} \langle \psi(\mathbf{x}, t) \psi(0, t) \rangle ,$$

which is given by the form due to Cahn, Hilliard, and Cook,

$$S(k, t) = [S(k, 0) - S'(k)] e^{A(k)t} + S'(k) , \quad (3)$$

where $S(k, 0)$ is the initial structure factor. $S'(k)$ is given by

$$S'(k) = \frac{T}{C(k^2 - k_c^2)} , \quad (4)$$

and k_c is a critical wave number given by

$$k_c = \sqrt{|r|/C} = \left[-\frac{1}{C} \left[\frac{\partial^2 f}{\partial \psi^2} \right] \right]^{1/2}. \quad (5)$$

The function, $A(k)$, in Eq. (3) is the amplification factor and is given by

$$A(k) = 2MCk^{2a}(k_c^2 - k^2). \quad (6)$$

To our knowledge, this result has not been observed experimentally in systems with short-range interactions because nonlinear effects become important very quickly. Indeed, neglect of the quartic term in the linear theory gives

$$S(t) \sim e^t [1 + O(\psi^2)]$$

for the structure factor. But the higher-order term itself behaves like e^t , resulting in a singular perturbation theory.¹⁵ Binder⁹ has shown, however, that systems with long-range interactions behave linearly during the early stages of coarsening after the quench. Note that the constant $C \sim R^2$ gives the range over which spatial inhomogeneities can persist. This implies that the wave number $k_c \propto 1/R$. Then, if one lets $\tilde{k} = kR$, and $\tilde{t} = t/R^{2a}$, we have the scaled expression for linear theory:

$$S(\tilde{k}, \tilde{t}) = \left[S(\tilde{k}, 0) - \frac{T}{\tilde{C}(\tilde{k}^2 - \tilde{k}_c^2)} \right] e^{2M\tilde{C}\tilde{k}^{2a}(\tilde{k}_c^2 - \tilde{k}^2)\tilde{t}} + \frac{T}{\tilde{C}(\tilde{k}^2 - \tilde{k}_c^2)}, \quad (7)$$

where $\tilde{C} \equiv C/R^2$, and $\tilde{k}_c \equiv Rk_c$. We use this to estimate the next term in the expansion.¹⁵ That term must grow to be of order a correlation volume, R^d , to be appreciable. This implies a time, $e^t \sim R^d$, and we thus recover Binder's result that linear theory holds for the time regime $t < O(R^{2a} \ln R)$.

We studied the early stages of the order-disorder transition (nonconserved ψ) by means of simulations on systems of size 256×256 , with periodic boundary conditions, choosing $R^2 = 220, 312, 420, 544, 684, 760, 840$, and 1200. Systems were quenched to $T = 0.4T_c$ with 40 independent runs performed for each interaction range. Simulations were terminated at 5 Monte Carlo steps per spin (MCS), since the dynamics was very fast. The first MCS was split into 64 substeps, while the remaining 4 MCS were split into 16 substeps. This was done to improve resolution during the early-time dynamics. In Fig. 1, typical configurations for $R^2 = 840$ are shown. Initially, up to about 0.5 MCS, the system is diffuse. As time increases, diffuse domains, without well-defined domain walls, begin to form. During these times the dynamics involves domains becoming more compact, as they increase in magnetization (i.e., local value of order parameters) without a significant increase in size. Note that a domain morphology, which is indicative of late-stage growth, is apparent by $t = 2.5$ MCS. Thus one should only expect a linear regime for times less than this, while fluctuations are small. By studying the system for several values of R , we found that changing the range of interac-

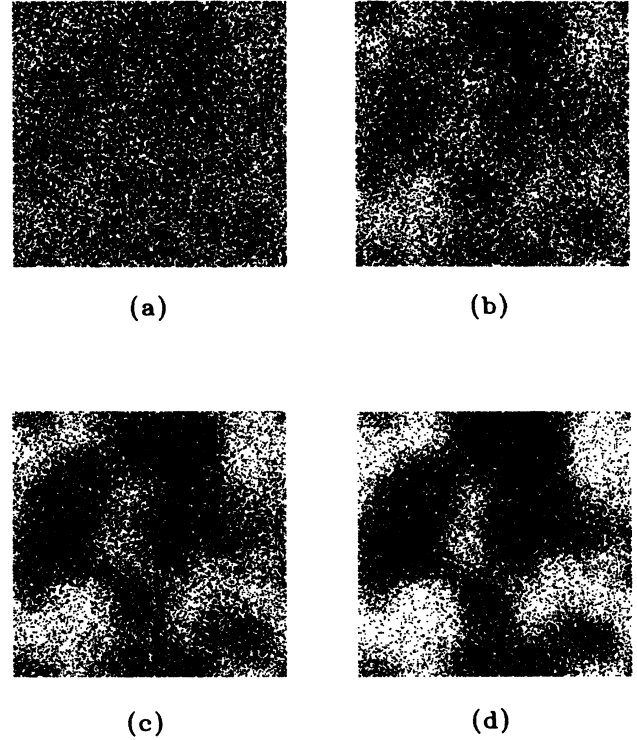


FIG. 1. Configurations for $R^2 = 840$. Times are $t = 0.5, 1.25, 1.875$, and 2.5 MCS. (Nonconserved order parameter.)

tion affects the size of domains rather than the growth rate, as expected. As domains become compact, i.e., their mean order parameter becomes equal to the equilibrium value, dynamics involves domain growth, which corresponds to the late-time scaling regime. This will be discussed in the next subsection. We also found that when the temperature of the quench increases towards T_c , domain walls become more diffuse.

A typical example of early-time evolution of the structure factor is shown in Fig. 2 for $R^2 = 420$. The structure factor has been fitted for each wave number k separately, up to 0.4375 MCS, for all the systems investigated. From this fit we obtained $A(k)$ as a function of k^2 , as shown for $R^2 = 544$ and 760 in Fig. 3. Note that the behavior is in agreement with linear theory [Eq. (6) with $a = 0$]. We extracted the critical wave number k_c from the form of $A(k)$ just mentioned. In Fig. 4 we show that $k_c \propto 1/R$, as predicted by Eq. (5), while in Fig. 5, we see that $-1/S'(k)$ is proportional to k^2 , in agreement with Eq. (4). Another indication of the linear regime is the scaling $S(k, t, R) = S(kR, t)$. As is shown in Fig. 6, this scaling works for early times, as anticipated, but breaks down at later times.

We next investigated the early-time behavior of phase separation by means of simulations on systems of size 256×256 , with $R^2 = 84, 144, 220$, and 312 . Again, we considered quenches to $T = 0.4T_c$, and 100 independent runs were performed for each interaction range. In contrast to the order-disorder transition, the dynamics here is much slower, due to the presence of the conservation of

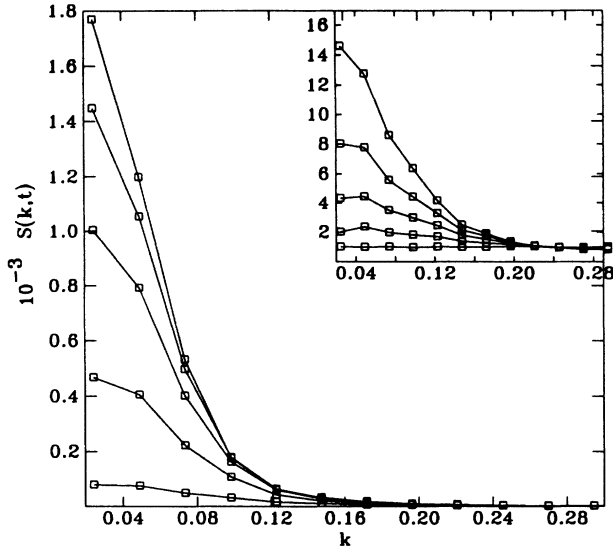


FIG. 2. Structure factor $S(k,t)$ for $R^2=420$. Times are $t=1, 2, 3, 4$, and 5 MCS. Inset shows the very-early-time evolution from 0 up to 0.5 MCS, with a period of 0.125 MCS. (Nonconserved order parameter.)

the order parameter in phase separation. From observation of coarsening in the configurations, we expect a linear regime (when the fluctuations in the local order parameter are small) for times less than about 60 MCS for $R^2=312$, and about 30 MCS for $R^2=220$. Figure 7

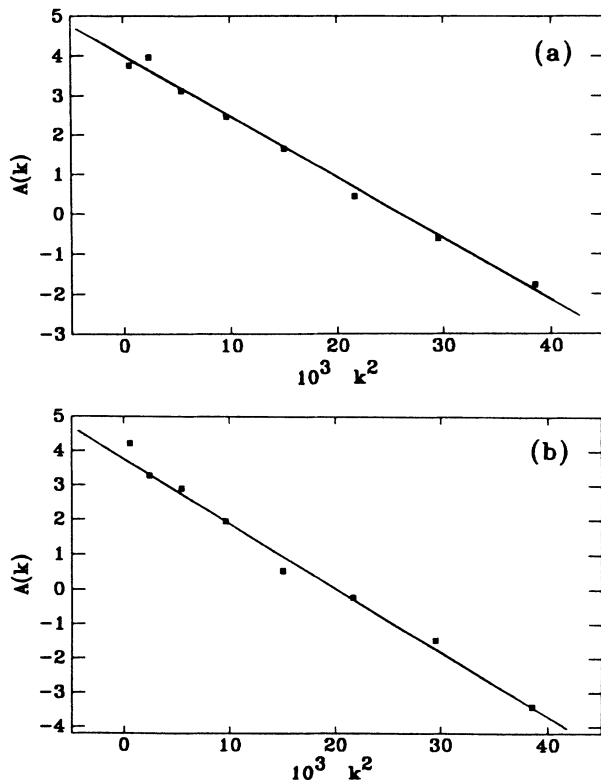


FIG. 3. Amplification factor $A(k)$ vs k^2 , for (a) $R^2=544$ and (b) $R^2=760$. (Nonconserved order parameter.)

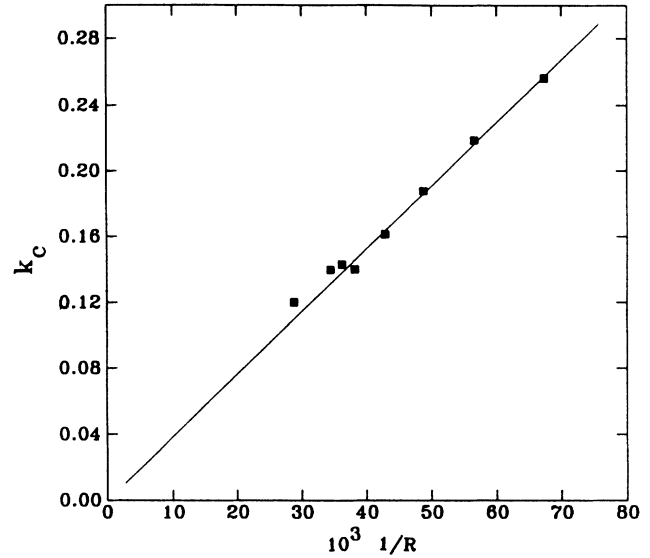


FIG. 4. Critical wave number k_c vs $1/R$. (Nonconserved order parameter.)

shows the time evolution of the structure factor for $R^2=220$ up to 70 MCS. We note that $S(0,t)$ is fixed during the phase separation due to the conservation law. Note also that the maximum of $S(k,t)$ is approximately fixed for early times. This implies that domains form and become more compact, but their sizes remain approximately constant. As in the order-disorder transition, we fitted the structure factor for $R^2=220$ and 312 to linear theory [Eq. (3)]. The resulting amplification factors are shown in Fig. 8, from which it is evident that,

$$A(k)/k^2 \propto (k_c^2 - k^2),$$

which is in agreement with Eq. (6) for phase separation. The critical wave number, k_c , was then extracted from the fit in Fig. 8, and the critical wave numbers for both the order-disorder transition and phase separation were

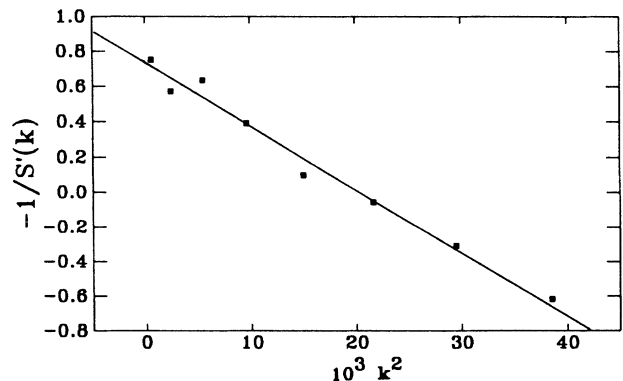


FIG. 5. Fitted results for $-1/S'(k)$ vs k^2 for $R^2=760$. (Nonconserved order parameter.)

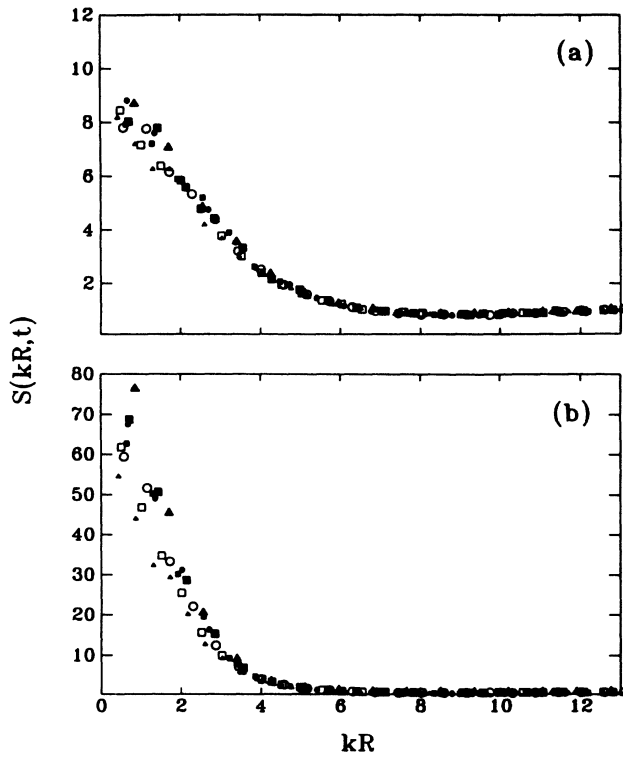


FIG. 6. Scaling of early-time results $S(kR, t)$. Times are (a) $t=0.375$ MCS and (b) $t=0.75$ MCS. (Nonconserved order parameter.)

found to be equal within approximately 12% for $R^2=220$ and 7% for $R^2=312$. This shows that k_c does not depend on the dynamics, but only the local quasiequilibrium aspects of spinodal decomposition. Also, as in the order-disorder transition, we verified the scaling of $S(k, t)$ with R , i.e., $S(kR, t/R^2)$, which is displayed in Fig. 9 for early times.

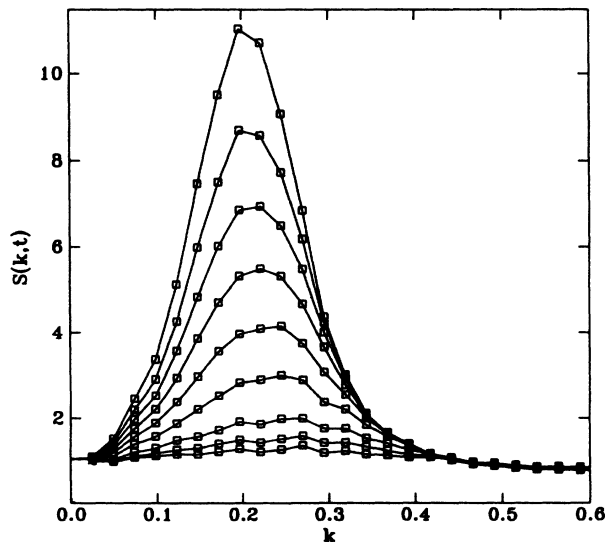


FIG. 7. Structure factor $S(k, t)$ for $R^2=220$. (Conserved order parameter.)

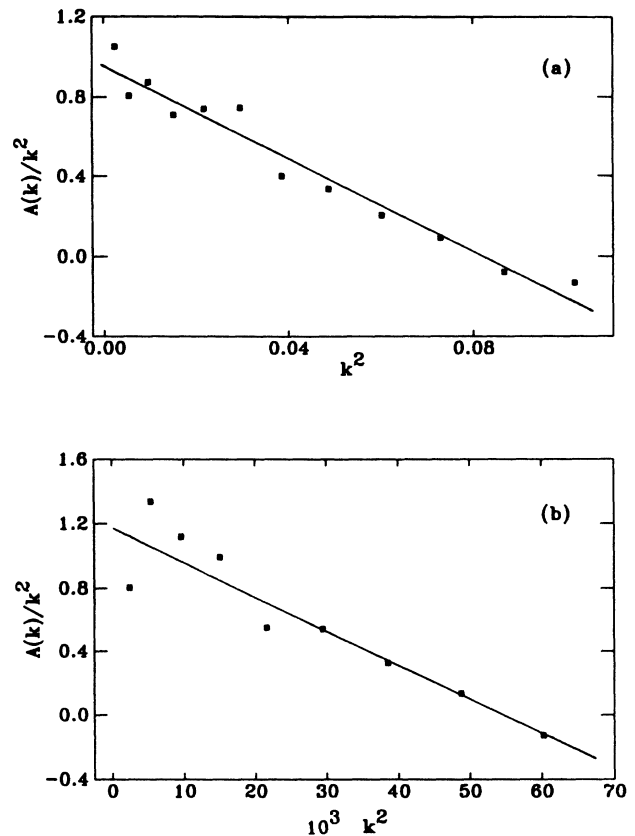


FIG. 8. Amplification factor $A(k)/k^2$ vs k^2 , for (a) $R^2=220$ and (b) $R^2=312$. (Conserved order parameter.)

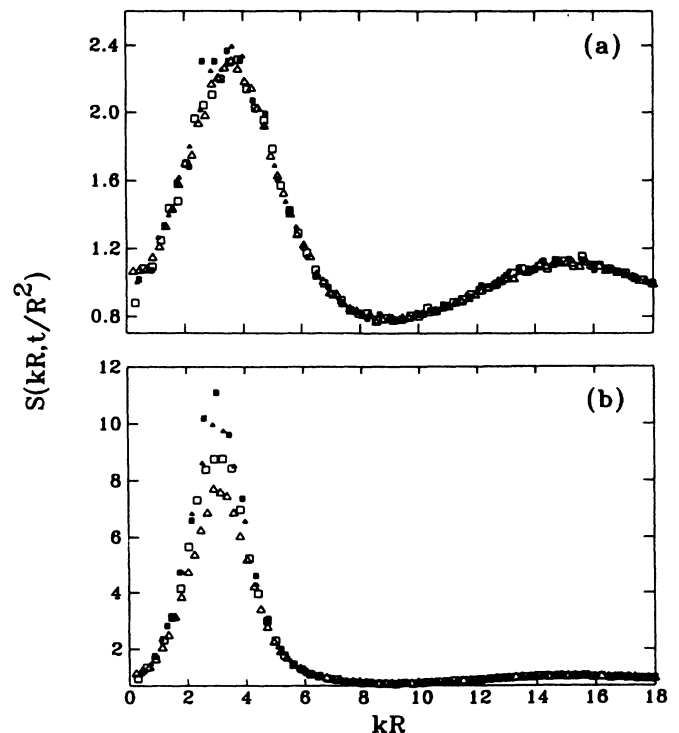


FIG. 9. Scaling of early-time results $S(kR, t/R^2)$. (a) $t/R^2=0.06$ and (b) $t/R^2=0.31$. (Conserved order parameter.)

B. Late stages

During the late stages of spinodal decomposition, the system is composed of domains of ordered phase which are separated by thin well-defined interfaces. The dynamics during these stages is such that the total surface free energy of these interfaces is minimized.^{16,17} The mean domain size grows in time via $L(t) \sim t^n$, while the structure factor scales with $L(t)$ by

$$S(k, t) = L^d(t) F(kL(t)). \quad (8)$$

The growth exponent n and the scaling function F are properties of the dynamical universality class. We have only considered the order-disorder transition for this section of the paper. In this case the Allen-Cahn¹⁶ growth law holds and therefore $n = \frac{1}{2}$. For our purposes, a convenient definition of length scale is given by¹⁸

$$L(t) = S(0, t)^{1/2}. \quad (9)$$

The scaling function has been calculated by Ohta, Jasnow, and Kawasaki.¹⁷ For $d=2$, they found

$$F(x) = \int dy \frac{y^2}{(e^{y^2} - 1)} J_1(xy), \quad (10)$$

where J_1 is a Bessel function of the first kind. For large x , this is in agreement with Porod's law, for which $F(x) \sim 1/x^3$. These results are well established for systems with short-range interactions,^{19,20} here we wish to clarify the effects of the interaction range, R , on domain growth. It is natural to expect that R only affects nonuniversal amplitudes in $L(t)$ and $F(x)$.

In order to study the late stages of order-disorder transitions for long-range interactions, we investigated systems of size 128×128 lattice sites, and $R^2 = 40, 60, 84, 144, 220$, and 312 . These systems were quenched to $T = 0.4T_c$, whereas the systems with interaction ranges $R^2 = 84$ and 220 were quenched to several temperatures, ranging from 0 to $0.95T_c$, in order to study the dependence of scaling and growth on temperature. The results were averaged over 20 independent runs, and the simulations were performed out to 100 MCS. Figure 10 shows that L^2 grows linearly with time, so $L(t) \sim t^{1/2}$, as ex-

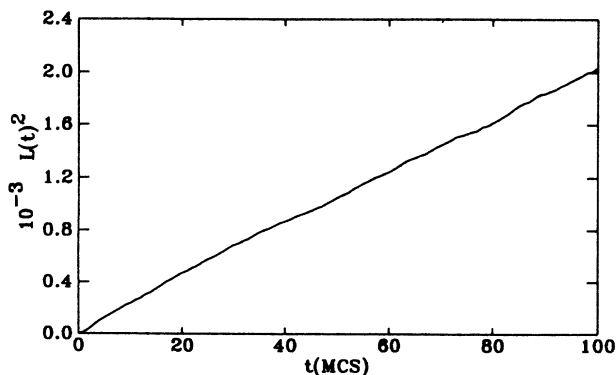


FIG. 10. Domain size squared $L(t)^2$ vs time t for $R^2 = 40$. (Nonconserved order parameter.)

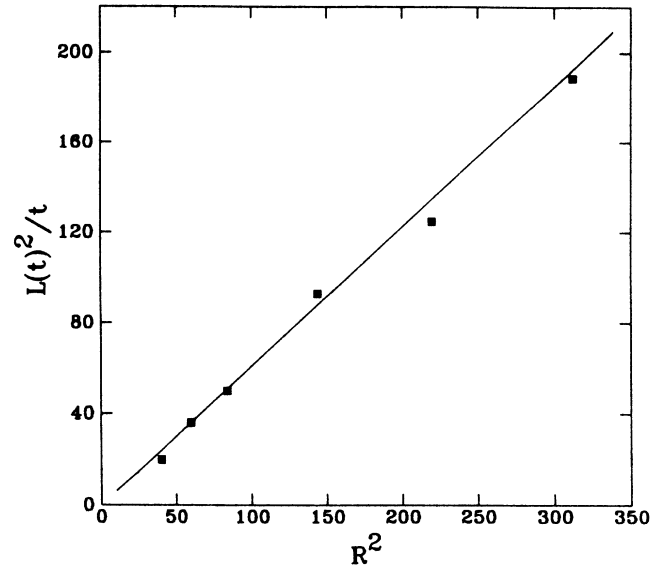


FIG. 11. Amplitude of growth law $L(t)^2/t$ vs R^2 . (Nonconserved order parameter.)

pected. For systems with ranges of interaction $R^2 > 220$, finite-size effects become important before we can observe the late-stage scaling regime. To show the dependence on the range of interaction on domain size, we plot the fitted values of L^2/t versus R^2 in Fig. 11. These fits are consistent with the form $L(t) \sim Rt^{1/2}$.

In addition, we calculated the scaling function,

$$F(x, t) = \frac{S(k, t)}{L^2(t)},$$

where $x = kL(t)$. For the systems we have studied, F becomes time independent for approximately $t > 40$ MCS. In Fig. 12 the scaled functions are plotted for all the sys-

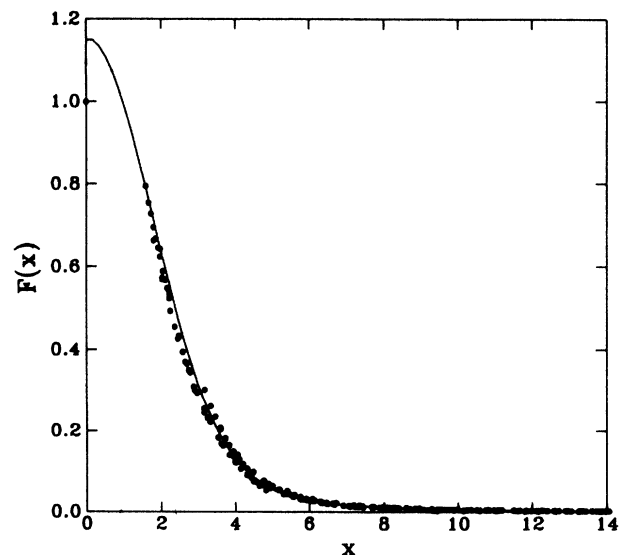


FIG. 12. Scaling function $F(x = kL(t))$ vs x , for different R . Solid-line fit is to Eq. (10). (Nonconserved order parameter.)

tems investigated at $0.4T_c$, from which it can be concluded that the scaled function does not depend on the range of interaction. The solid line corresponds to the prediction of Ohta, Jasnow, and Kawasaki,¹⁷ which is in good agreement with our numerical results. We also fitted F for large wave numbers to Eq. (11), and found agreement with Porod's law. Furthermore, we have studied the temperature dependence of scaling of two interactions $R^2=84$ and 220 , for several temperatures ranging from 0 to $0.95T_c$. For $T \leq 0.4T_c$ the scaling function is almost temperature independent, but for larger temperatures F becomes strongly dependent on T , and narrows as T increases. However, when x is rescaled with a temperature-dependent prefactor $\Omega(T)$ as in $x \rightarrow x/\Omega(T)$, $F(x)$ is again given by the form calculated by Ohta, Jasnow, and Kawasaki to the accuracy of our calculations. This factor, $\Omega(T)$, decreases with increasing temperature as is qualitatively expected from theoretical arguments.²¹ This is reasonable since $\Omega(T)$ can be regarded as the temperature-dependent amplitude of the growth law.

C. Conclusions

In summary, we have investigated the effects of long-range interactions on the kinetics of first-order phase transitions. This is the first study of phase separation for

both the *two-dimensional* Ising model with long-range interactions and the early stages of the order-disorder transition. We have found that long-range interactions are sufficient to allow the observation of a linear regime, in agreement with Binder's predictions. The critical wave number was found to be independent of the dynamical model studied, which gives us confidence in both our simulation results and the use of coarse-grained models to describe kinetic Ising models, even during the early stages of growth. An unanswered question, which is not addressed by our work, concerns the existence of a linear regime for systems with short-range interactions. We also studied the late stages of growth of an order-disorder transition. Our results imply that, for any large but finite range of interaction, there is an eventual crossover to late-stage growth. In this late-stage growth regime, the range of the interaction only appears to affect nonuniversal amplitudes.

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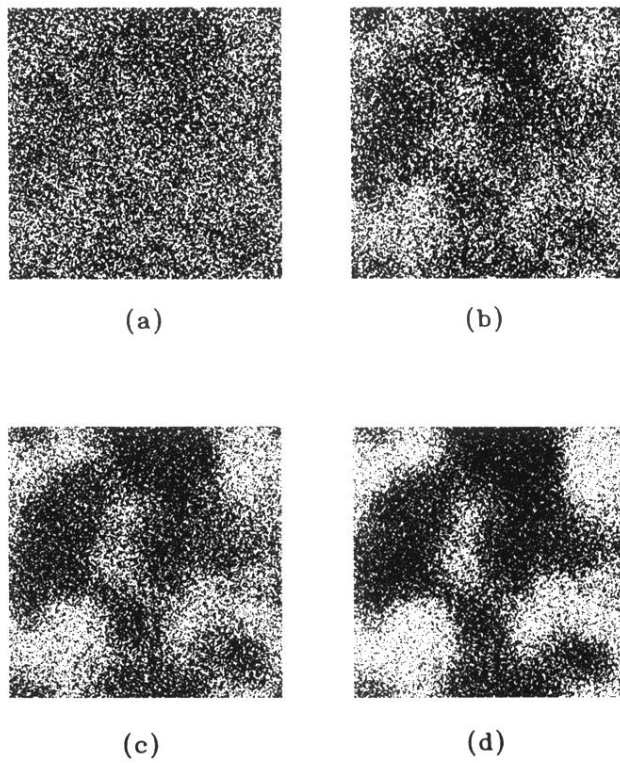


FIG. 1. Configurations for $R^2=840$. Times are $t=0.5$, 1.25, 1.875, and 2.5 MCS. (Nonconserved order parameter.)