Numerical study of late-stage coarsening for off-critical quenches in the Cahn-Hilliard equation of phase separation

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Off-critical quenches of a two-dimensional system with a continuous, conserved order parameter are studied numerically. Domain growth and scaling are investigated by monitoring several measures of the morphology. For quenches within the mean-field spinodal, circular domains coarsen through an evaporation-condensation mechanism. The dynamics of this process are compared with the theory of Lifshitz and Slyosov. Surface smoothening plays an important role for nearly critical quenches and extends the timescale for the onset of self similar growth.

I. INTRODUCTION

During a first-order phase transition, coexisting macroscopic domains of different phases emerge from initially small fluctuations in a homogeneous, single-phase state. Late stages of this process are often dominated by the motion of interfaces separating the domains. The average domain size emerges as a time-dependent length for scale-invariant coarsening of the morphology. The growth of this length (usually a power law in time) and the scaling morphology are important characterizations of the dynamics. Similarities between many diverse processes have led to the identification of several universality classes for domain growth.[1,2] Systems with a conserved order parameter (which is not coupled to other hydrodynamic modes) form one of the simplest and most studied classes. Examples of such systems include binary alloys, polymers, and the spin exchange kinetic Ising model. Recently, a great deal of interest has centered on the late-stage growth law for this class.[3–21] As a result of extensive experiments and computer simulations, there is mounting evidence[22] of an asymptotic growth exponent of $\frac{1}{3}$. For a very dilute quench, where the volume fraction of one phase greatly exceeds the other, the growth is well understood in terms of the theoretical work of Lifshitz and Slyosov (LS).[3] However, for quenches through the critical point, where the phases are symmetric, the assumptions of LS are no longer valid. A theoretical understanding of domain growth and scaling in this regime remains somewhat controversial.[4,8,11,14]

The archetypal field theory for a conserved system is model *B*. The time evolution of the order parameter is described by a stochastic partial differential equation (the Cahn-Hilliard-Cook equation[23]) which can be expressed in the dimensionless form[24]

$$\frac{\partial\phi(\mathbf{r},\tau)}{\partial\tau}=\tfrac{1}{2}\nabla^2[-\phi(\mathbf{r},\tau)+\phi^3(\mathbf{r},\tau)-\nabla^2\phi(\mathbf{r},\tau)]+\epsilon^{1/2}\mu(\mathbf{r},\tau)\ , \tag{1}$$

with

$$\langle\mu(\mathbf{r},\tau)\mu(\mathbf{r}',\tau')\rangle=-\nabla^2\delta(\mathbf{r}-\mathbf{r}')\delta(\tau-\tau')\ .$$

Here $\phi(\mathbf{r},\tau)$ refers to the value of the order parameter at position $\mathbf{r}$ at time τ. (ϕ has been normalized to ±1 in the equilibrium phase). The only dimensionless parameter ϵ characterizes the strength of thermal fluctuations μ in the final state.

Equation (1) must be supplemented with appropriate initial conditions describing the system prior to the onset of phase separation. For quenches from a single-phase state

$$\phi(\mathbf{r},\tau=0)=\psi_0+\psi(\mathbf{r})\ , \tag{2}$$

where ψ represents fluctuations in the initial state which are symmetrically distributed about a mean value of zero. ψ_0 is a measure of how far off-critical the system is. For $\psi_0=\pm1$ the system will be quenched to the coexistence curve, while $\psi_0=0$ corresponds to a quench through the critical point. According to the "lever rule," nonzero ψ_0 reflects asymmetry in the amount of each phase present in the final state.

In this paper, numerical simulations are used to study the dynamics of phase separation for *off-critical* quenches of model *B* in two dimensions. Specifically we address the role that the asymmetry in phases plays in the growth law and in scaling by monitoring the time evolution of several measures of domain size. For large ψ_0, growth is dominated by competition between circular domains as described by Lifshitz and Slyosov. We compare the growth at $\psi_0=0.4$ with a simple extension of LS to two dimensions. Convoluted domains are formed during the early stages for quenches at $\psi_0=0.1$ and 0.2. Smoothening of the surface to form circular domains plays an important role initially and extends the time scale for the onset of scaling of the morphology. For quenches through the critical point, percolating domains are formed, leading to a different type of scaling morphology. The growth exponent of $\frac{1}{3}$ measured here is due to relaxation of surface corrugations. The study is restricted to the case $\epsilon=0$, which corresponds to a low-temperature quench of a mean-field system. However, we expect the conclusions to be applicable for small values of ϵ as well.

II. COMPUTER SIMULATIONS

In order to simulate Eq. (1), spatial gradients were approximated by a finite difference scheme. Following Oono and Puri,[13] an "isotropic" form for the discrete Laplacian was used which couples each cell to nearest and next-nearest neighbors. The mesh size Δx introduced in the discretization is bounded by the sharp gradients inherent to the interfaces as discussed in Ref. 16. The morphology of the clusters also seems to play an important role, with circular clusters being more sensitive to Δx than the convoluted domains seen in critical quenches. Large mesh sizes typically generate anisotropic clusters which align with the underlying lattice imposed in the difference scheme. In addition, cluster growth is slowed down.[25] However, a trade off in computational effort exists, since the time required for a simulation[16] varies approximately a Δx^{-4}. For the simulations reported here, the mesh size was set at 1.0.

A fourth-order Adams-Bashford predictor and Adams-Moulton corrector algorithm approximated the time derivative.[26] A timestep of 0.1 was used, which led to an average fractional correction to ϕ of approximately 10^{-6} at each iteration for the late stages. (This degree of accuracy is probably not necessary for the quantities we are interested in, since previous studies at the critical point show that the late-stage growth is not sensitive to the timestep.) The late stages of some runs were repeated with a timestep of 0.01 to ensure convergence.

Most of the simulations were performed on square lattices of size 256^2 using periodic boundary conditions. However, smaller lattices (128^2) were also examined to ensure that there were no finite-size effects. The system was initially prepared in a homogeneous state by assigning a random number to each lattice site. The random numbers were uniformly distributed, with a mean value of ψ_0, and a width of 0.1. (The width of the distribution reflects the strength of thermal fluctuations in the initial state.) Averages were performed over many realizations using a different set of random numbers for each initial state.

III. GROWTH LAWS

The separation process was examined for various quenches within the mean-field spinodal ($\psi_0 < 3^{-1/2}$) as summarized in Table I. All simulations were run up to a time of $\tau = 5000$. Figure 1 compares the order-parameter evolution for $\psi_0 = 0$ and 0.4. Domain coarsening is evident in both cases, although the morphology depends on ψ_0. For the critical quench ($\psi_0 = 0$) there is symmetry between the two phases resulting in convoluted percolating interfaces. When the system moves off-critical this symmetry is broken. At $\psi_0 = 0.4$ the minority phase consists of isolated, compact clusters. Our studies show that the morphology plays an important role in the dynamics. In order to make a quantitative analysis of the growth, several different quantities were measured as a function of time.

Correlations of fluctuations about ψ_0 were examined through the radial pair correlation function. This function exhibits characteristic oscillations about zero, reflecting the domain structure. The time dependence of the first zero $R_1(\tau)$, which is a common gauge of domain coarsening, is shown in Fig. 2 for each ψ_0. (The details of this calculation are described in Ref. 16.) Many important features of the separation process are evident in the plot. Initially small fluctuations in the order parameter grow as a result of the linear instability in Eq. (1). During this time there is little change in R_1. As the magnitude of the fluctuations increases, nonlinearities in the equation lead to the formation of domains of each phase separated by sharp interfaces.[27] In the final stage, which is the focus of this paper, the domain structure coarsens as a result of surface tension. The time scale for the transition to this stage increases with ψ_0. In Fig. 2 the late stages have been reached for all ψ_0 when $\tau > 500$ [$\ln(\tau) > 6$].

TABLE I. Parameters used in the simulations.

ψ_0	Size	N_{trials}[a]
0.0	$(256)^2$	5
0.1	$(128)^2$	6
	$(256)^2$	3
0.2	$(128)^2$	5
	$(256)^2$	9
0.4	$(128)^2$	3
	$(256)^2$	5

[a] N_{trials} refers to the number of different realizations used to average over the fluctuations in the initial state. Each realization used a different seed for the random number generator.

The slope of the logarithmic plot can be interpreted as an effective exponent n_{eff} for power law growth of R_1. For both $\psi_0 = 0$ and 0.1, the plot is linear for $\tau > 200$, with a slope of approximately $\frac{1}{3}$ (which is approached monotonically from below). This result is consistent with previous studies of the critical quench at these late times.[13,15–18,22] The data for $\psi_0 = 0.2$ initially follow the same trend as with smaller ψ_0. There appears to be convergence towards a slope of $\frac{1}{3}$ for $\tau < 1000$. Beyond this time, however, there is a systematic curvature in the plot which lies beyond the statistical scatter in these data. This curvature can be interpreted as a decrease in n_{eff} with time. Like the critical quench, an increasing slope is seen in the late stages ($\tau > 500$) for $\psi_0 = 0.4$. However, the data yield an effective exponent which is smaller than $\frac{1}{3}$ (see Table II).

One of the hallmarks of late-stage coarsening is the emergence of interfaces with a characteristic width which is time independent. As a consequence, the inverse perimeter density P forms another coarsening length scale, where

$$P \equiv (1-\psi_0)^{1/2} \frac{A}{N_p} . \tag{3}$$

A is the total area of the system and N_p refers to the area covered by interfaces. The prefactor is included to make

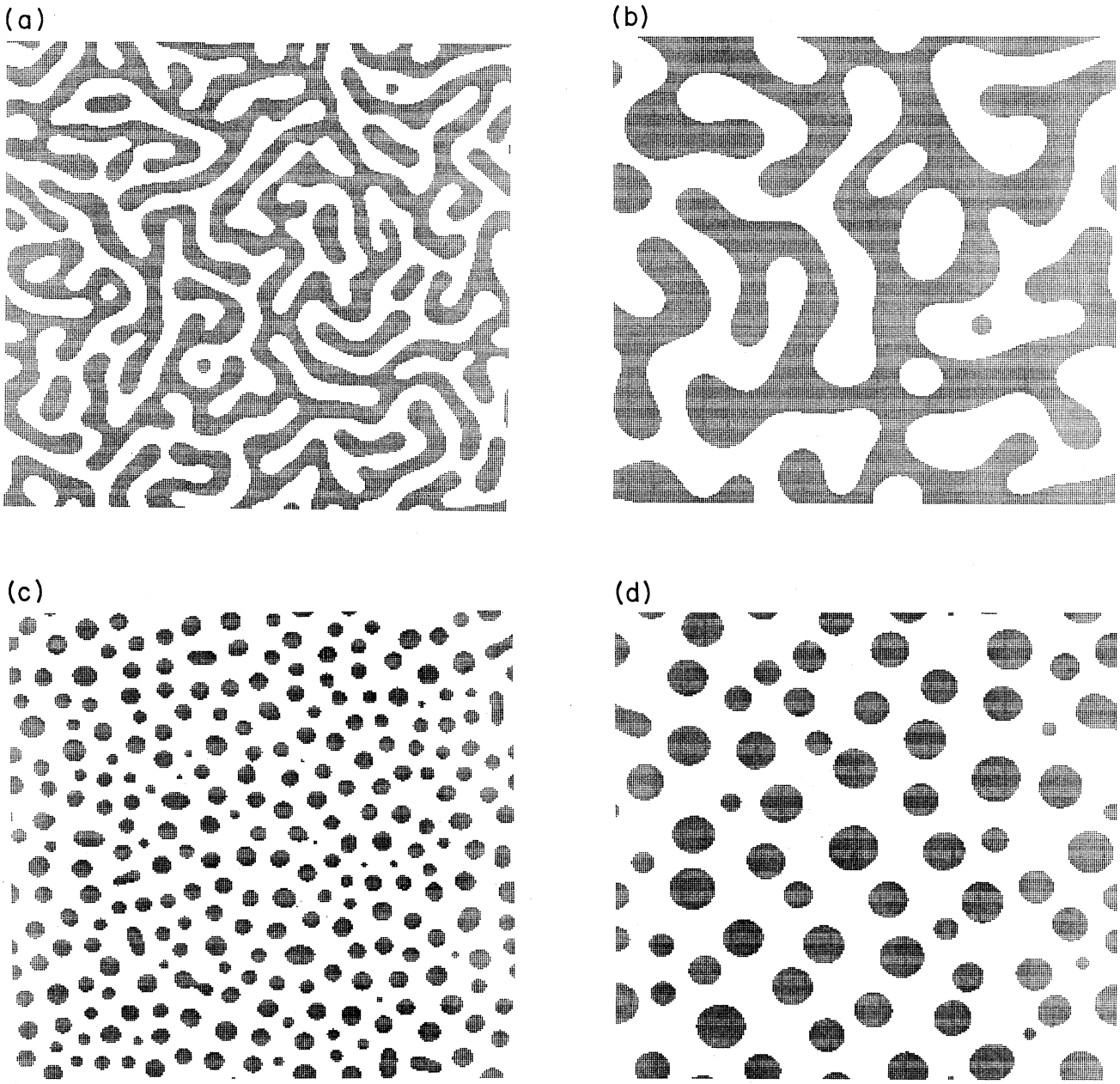

FIG. 1. Morphology of the order-parameter field (ϕ) for $\psi_0=0.0$ at $\tau=500$ (a) and 5000 (b); and for $\psi_0=0.4$ at $\tau=500$ (c) and 5000 (d). The shaded regions correspond to $\phi<0$.

P independent of ψ_0 in the final equilibrium state (assuming the domains have the same shape). N_p was calculated in the simulations by considering each cell as a part of an interface if $|\phi|<0.75$. (Note, however, that P is relatively insensitive to the cutoff because of the sharpness of the interface.) Figure 3 shows the time evolution of P for the different quenches. The effective exponent describing the growth of P is given in Table II.

For the off-critical simulations performed in this study, the minority phase forms a set of isolated domains. We used the working definition of a cluster as the set of connected lattice sites where $\phi<0$ (recall ϕ is negative in the minority phase when $\psi_0>0$). Using the clusters generated from all simulations at a given ψ_0, averaged properties were monitored. Figure 4 shows the time dependence of the cluster density N, where N is defined as the number of clusters *per unit area* in the system. For reasons that will become apparent in Sec. IV, it is useful to describe the

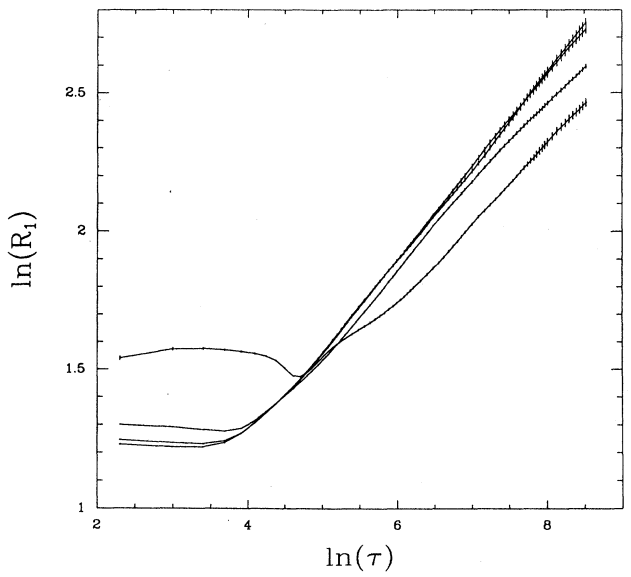

FIG. 2. Time dependence of the first zero of the pair correlation. At $\ln(\tau)=2$, from bottom to top the data correspond to $\psi_0=0$, 0.1, 0.2, and 0.4. [At $\ln(\tau)=8$, the ordering is reversed.]

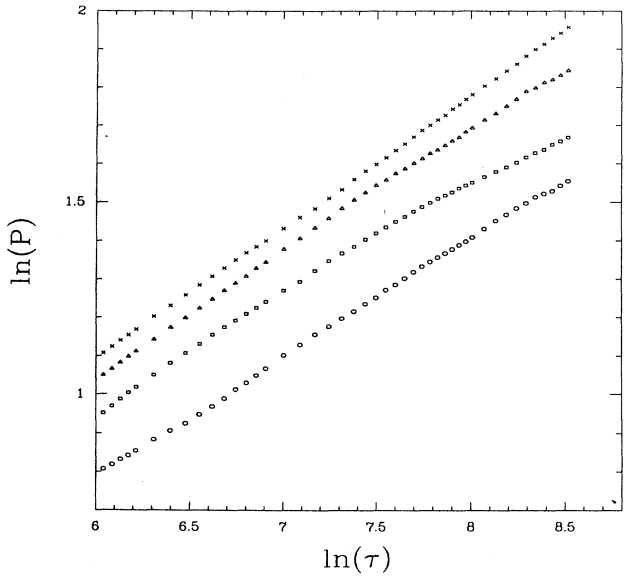

FIG. 3. Time dependence of the inverse perimeter density P for $\psi_0=0$ (crosses), 0.1 (triangles), 0.2 (squares), and 0.4 (circles).

evolution of N in terms of an effective exponent n, such that

$$N \sim N_0 t^{-2n} . \tag{4}$$

It can be seen from Table II that this effective exponent increases with increasing ψ_0.

A radius of gyration was used to define the cluster radius

$$R_c = \left[\frac{2}{N} \sum_i (\mathbf{r}_i - \mathbf{r}_{\text{c.m.}})^2 \right]^{1/2} . \tag{5}$$

The sum is over the N lattice sites of the cluster. $\mathbf{r}_i$ is the position of lattice site i and $\mathbf{r}_{\text{c.m.}}$ is the position of the cluster's center of mass. (The factor of 2 ensures that R_c reduces to the usual cluster radius for circular clusters.) In Fig. 5 the average radius is plotted for the off-critical quenches.

IV. DYNAMICAL SCALING

First-order phase transitions often exhibit dynamical scaling (or self-similarity), wherein the morphology at a given time can be made to (statistically) match that of an earlier time by a global change of scale. Consequently, the growth of the system is characterized by a single length (the average domain size) to which all other relevant lengths must scale. Dynamical scaling forms an important cornerstone of the emerging theoretical understanding of domain growth.

For quenches near the coexistence curve, the onset of scaling is predicted by the classic work of Lifshitz and Slyosov (LS).[3] In the LS analysis, clusters of the minority phase compete through an evaporation condensation mechanism, whereby larger clusters grow at the expense of smaller ones. Cluster interaction is mediated by diffusion in the majority phase. Although their theory was originally formulated in three dimensions, it can be extended to two dimensions as discussed in the Appendix. The LS analysis focuses on the time dependence of the cluster distribution function $f(R,t)$. [Here f is defined such that $f(R,t)dR$ is the probability of finding a cluster with radius between R and $R+dR$ and it satisfies the normalization $\int f(R,t)dR = 1$.] Scale invariance is manifested by the asymptotic result

TABLE II. Effective growth exponents for various measures of domain size.

Measure	$\psi_0=0$	$\psi_0=0.1$	$\psi_0=0.2$	$\psi_0=0.4$
R_1	0.34±0.01	0.33±0.01	0.29±0.01	0.29±0.02
P	0.34±0.01	0.32±0.02	0.29±0.01	0.31±0.02
$\langle N \rangle$[a]		0.20±0.03	0.23±0.02	0.28±0.02
$\langle R_c \rangle$		0.11±0.03	0.19±0.01	0.29±0.01

[a]In order to make a direct comparison with other measures of domain coarsening, the exponent reported for $\langle N \rangle$ is *half* the slope describing the data points in Fig. 4 [see (A14)].

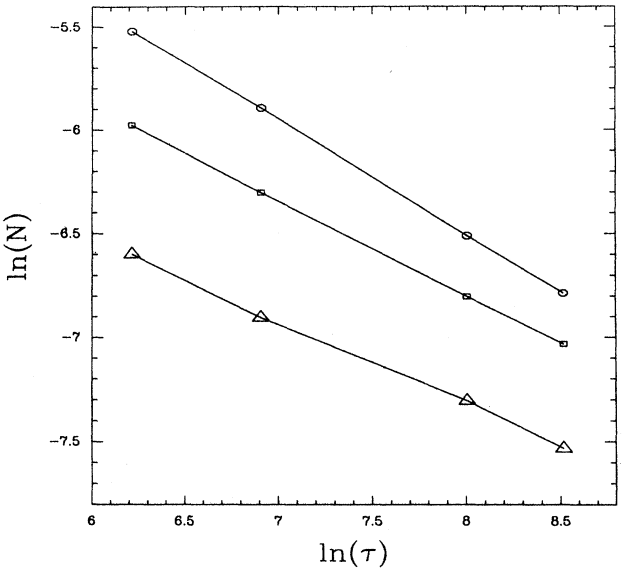

FIG. 4. Time dependence of the cluster number density for $\psi_0=0.1$ (triangles), 0.2 (squares), and 0.4 (circles).

$$f(R,t)=f_0(R/\langle R_c\rangle)\,, \tag{6}$$

where $\langle R_c\rangle$ is the average cluster radius and f_0 is a time-independent function.

In our study the closest quench to the coexistence curve is $\psi_0=0.4$. Table II shows that for this simulation all measures of domain coarsening give the *same exponent,* which is a necessary condition for scaling. With a value of 0.29 ± 0.01, the growth exponent is less than the conventional LS prediction of $\frac{1}{3}$. It should be noted, however, that the lack of a steady-state solution to the diffusion field in two dimensions invalidates a direct application of the LS assumptions. In the Appendix this singular behavior is handled by employing an asymptotic analysis of the diffusion equation. As a result, the scaling length grows as $(t/\ln 4t)^{1/3}$. The logarithmic time correction, which does not occur in three dimensions, is a consequence of the long-range nature of the diffusion field. This correction underscores the slow approach to $\frac{1}{3}$ in two dimensions.[28]

Figure 6 compares the cluster distribution at $\tau=500$ and $\tau=5000$ for $\psi_0=0.4$. Instabilities in the early stages lead to nearly circular clusters with a relatively sharp distribution of radii. As time progresses, the width of the distribution increases slightly. The late-stage cluster distribution function is much broader than the LS prediction. The discrepancy can be traced back to the mean-field nature of LS. Specifically, in deriving the growth rate of a given cluster LS assume that the influence of all other clusters in the system can be modeled by a simple boundary condition. Correlations between clusters, which can affect the growth, are precluded by this assumption. A large cluster, for example, grows more quickly when surrounded by smaller clusters, than when surrounded by clusters of comparable size. These correlations will act to spread out the distribution function.

In contrast to the above case, scaling has not been obtained for $\psi_0=0.1$ and 0.2 since different measures give different exponents. In particular, the average cluster radius grows with a much smaller exponent than the first

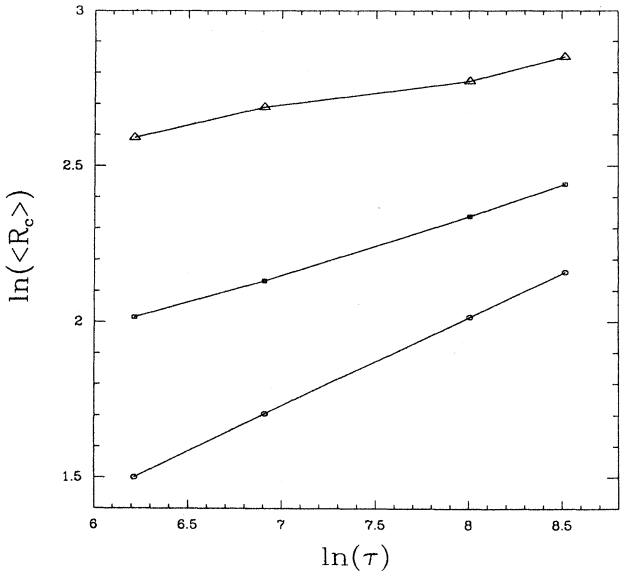

FIG. 5. Time dependence of the average cluster radius for $\psi_0=0.1$ (triangles), 0.2 (squares), and 0.4 (circles).

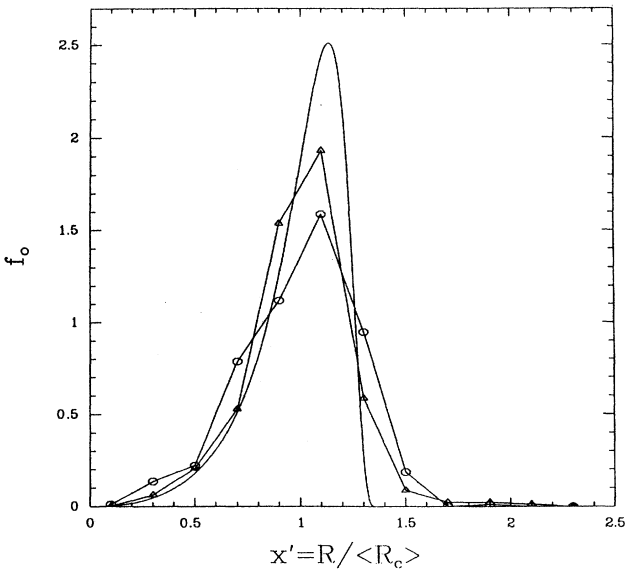

FIG. 6. The (normalized) cluster probability function for $\psi_0=0.4$. The triangles correspond to $\tau=500$; the circles to $\tau=5000$. The upper line is the Lifshitz-Slyosov prediction for two dimensions. Note that all distributions have an average of $\langle x'\rangle=1$. [To compare with (A11), $x'=x/1.0665$.]

zero of the pair correlation function. The breakdown of scaling is related to the cluster morphology. An important element of the LS analysis is the assumption of compact (circular) clusters. However, for small ψ_0, early-stage instabilities lead to convoluted domains. For such morphologies, smoothening of surface corrugations forms another coarsening mechanism in which the system acts to eliminate changes in the curvature as a function of arc length. Consequently, there is an evolution of the cluster shapes towards circles. Figure 7 illustrates this process for a typical simulation at $\psi_0=0.2$. As a result of changes in the cluster shape, the evolution of the cluster distribution differs qualitatively from the quench at $\psi_0=0.4$. Initially a large spread in the distribution is created. With the progression of time there is a change in the *skew* as the maximum shifts towards a larger radius. (Note that the very asymmetric LS prediction has a sharp upper cutoff.) This phenomenon has also been observed in studies of the spin exchange kinetic Ising model.[5] Surface smoothening and LS cluster competition are both driven by surface tension. However, the former can be a single cluster interaction which changes the cluster shape, whereas the latter results from interactions between different clusters. Both processes are initially operative when $\psi_0=0.1$ and 0.2.

For a cluster with area A, it is useful to define a shape factor

$$S=\frac{A}{\pi R_c^2}\ , \tag{7}$$

where R_c is defined in (5). For circular clusters $S=1$, while convoluted clusters are characterized by $S<1$. In Fig. 8, the average shape factor is plotted as a function of time for the off-critical quenches. At $\psi_0=0.4$, the factor remains unity throughout the late stages, which is a necessary consequence of scaling. For smaller ψ_0 there is a systematic increase in $\langle S\rangle$ as the cluster surface is minimized. Here scaling of the morphology has not been reached for the time scales we have probed because the clusters are not yet circular.

At the critical quench, on the other hand, the system forms large percolating domains. In two dimensions, breakup of these domains will not occur during the late

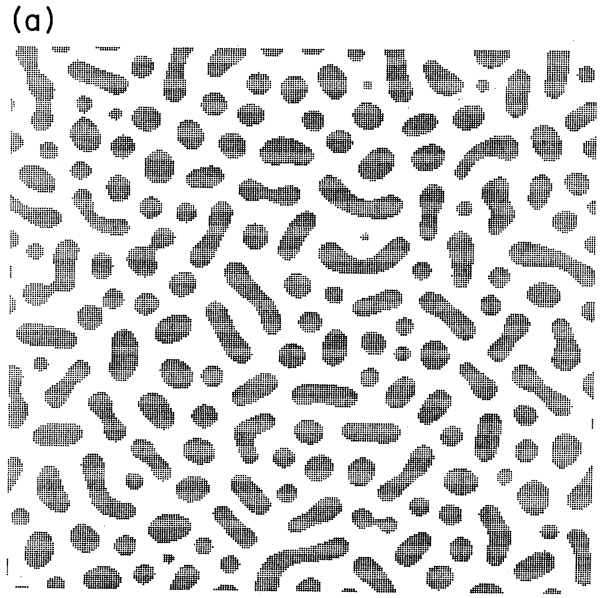

FIG. 7. Morphology of the order-parameter field (ϕ) at $\tau=500$ and 5000. The shaded regions correspond to $\phi<0$. Here $\psi_0=0.2$.

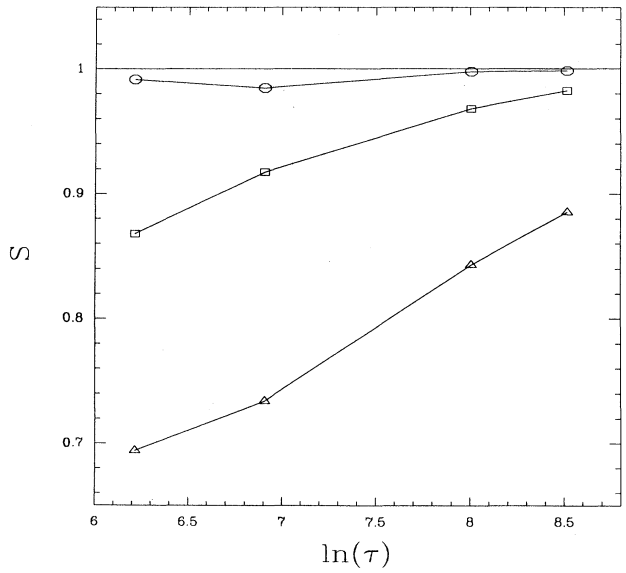

FIG. 8. Time dependence of the average cluster shape factor for $\psi_0=0.1$ (triangles), 0.2 (squares), and 0.4 (circles). The horizontal line at $S=1$ corresponds to circular clusters.

stages because the system is stable to "necking and pinching".[29] A crossover to the LS regime of circular clusters does not occur and surface relaxation remains an important growth mechanism. The data show that this process also leads to a growth exponent of $\frac{1}{3}$ for R_1 and P.

V. DISCUSSION

In the late stages of phase separation, surface tension is the driving force for domain coarsening. We believe that the LS theory captures the essential features of coarsening for $\psi_0=0.4$. The system seems to be approaching a scaling regime. The effective exponent, although less than $\frac{1}{3}$, is consistent with the slow approach to asymptotic growth in two dimensions. In detail, however, the LS predictions fail because they do not account for cluster correlations. Most notably, the LS distribution function is too narrow. (It should be noted that LS theory applies to an infinitely dilute minority phase, whereas for $\psi_0=0.4$ the minority phase is quite concentrated.)

For smaller values of ψ_0, cluster interaction is more intense in the early stages, resulting in convoluted, noncircular domains. Here, smoothening of the surface plays an important role. This coarsening mechanism is precluded by the assumptions of LS. Quenches through the critical point show that the process leads to a growth exponent of $\frac{1}{3}$ for the inverse perimeter density and for the pair correlations. At $\psi_0=0.1$ and 0.2 there is a crossover from growth dominated by surface smoothening to LS-type growth as the system evolves. Late-stage scaling of the *morphology* has not set in for these simulations. We conjecture that percolating clusters and circular domains form two types of scaling morphology. The former occurs for critical quenches, while the latter applies near the coexistence curve. When $\epsilon=0$, the crossover from percolation to isolated droplet growth occurs very close to the critical quench (i.e., $\psi_0<0.1$). (The mean-field spinodal plays no special role in late-stage coarsening since the circular domain topology for $\psi_0=0.4$ is consistent with a nucleating system despite the fact that the quench lies within the spinodal.) For all values of ψ_0, however, we believe that the asymptotic growth exponent is $\frac{1}{3}$ (with possible logarithmic corrections near the coexistence curve).

Previous studies of quenches at the critical point suggest that for small values of ϵ, thermal fluctuations in the final state will only play a minor role in late-stage coarsening. However, the introduction of thermal fluctuations allows for diffusive motion of the center of mass of the domains.[30] For large ϵ, this new mechanism may lead to qualitative changes in the separation process.

One of the most important quantities uniting theory, experiment, and computer simulation is the pair correlation function $g(r,\tau)$ (and its Fourier transform, the structure factor).[31] The first zero of g can be used to construct a scaling function g_0

$$g(r,\tau)=(1-\psi_0^2)^{-1}g_0(z)\ , \tag{8}$$

where $z=r/R_1(\tau)$. The normalization factor $(1-\psi_0^2)$ is included so that for an infinite system

$$\lim_{\tau\to\infty} g_0(z=0)=1\ . \tag{9}$$

"Scaling" is characterized by time independence of g_0. The late-stage scaling functions for $\psi_0=0$ and 0.4 are compared in Fig. 9. The identification of a scaling regime through examination of the pair correlation function depends on the statistics of the simulations since the changes with time become very small in the late stages. For the simulations reported here, the most notable change for $\tau>3000$ was a slight increase in g_0 with time near the origin. This increase will continue, as demonstrated in Fig. 9 by the fact that $g_0(z=0)\neq 1$. The behavior at the origin can be understood in terms of a correction to scaling due to the finite interfacial width l. Such corrections, which are of order l/R_1, arise because the (time-independent) interfacial width introduces a second length scale into the problem. (Note that $l/R_1\to 0$ as $\tau\to\infty$.)

Despite the differences in morphology (see Fig. 1), g_0 is remarkably similar for the two values of ψ_0. (It should be noted that differences near the origin are partly due to the fact that l/R_1 is different for two simulations.) Clearly, for this range of ψ_0, the pair correlation function is *not* very sensitive to the topological structure (e.g., circular clusters versus percolating domains). In an experiment or simulation, time independence of the scaled pair correlation function may not necessarily imply scaling of the morphology.

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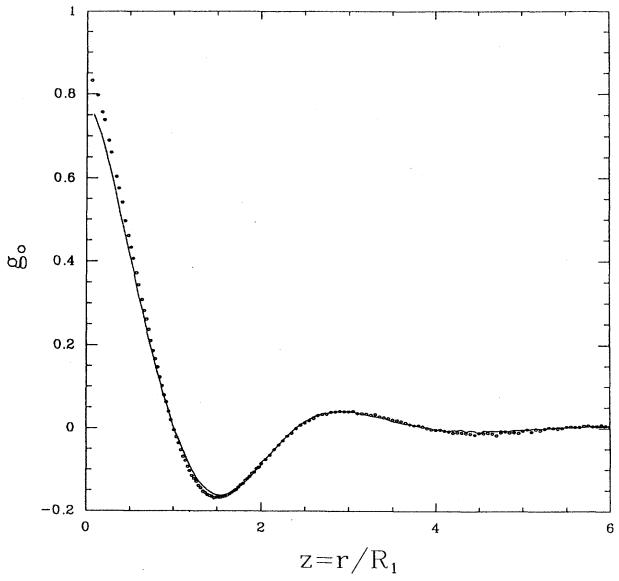

FIG. 9. Scaled pair correlation function at $\tau=5000$ for $\psi_0=0$ (circles) and 0.4 (solid line).

puter time on their CRAY XMP. This work is also supported by the Natural Sciences and Engineering Research Council of Canada.

APPENDIX

In this appendix a simple extension of Lifshitz-Slyosov theory (LS) is presented for model B in two dimensions. The theory is appropriate for quenches near coexistence where one phase ($\phi^{\min}$) occupies a much smaller area fraction than the other (ϕ^{maj}) in the final equilibrium state. We continue to use the dimensionless variables introduced in Eq. (1). For convenience, the case $\phi^{\text{maj}}=1$ and $\phi^{\min}=-1$ is considered.

In the majority phase, small gradients in the order-parameter field are established which influence the growth of the clusters. Linearizing (1) about this phase generates a diffusion equation of the form

$$\frac{\partial \delta\psi}{\partial \tau}=\nabla^2\delta\psi \ , \tag{A1}$$

where $\delta\psi=1-\phi$. [Only the lowest-order gradient term has been retained in (A1).] It is assumed that the minority phase consists of circular clusters in which the order parameter assumes its equilibrium value. It is further assumed that the cluster radius is much larger than the interfacial width. The cluster interface can then be modeled as a boundary condition for diffusion in the majority phase. In particular, at the surface of each cluster a Gibbs-Thomson form of boundary condition results

$$\delta\psi|_{\text{surf}}=\frac{\sigma}{4R} \ . \tag{A2}$$

Here R is the radius of the cluster and σ is the surface-free energy.[4]

Diffusion gradients in the neighborhood of a given cluster lead to a flux, causing the cluster to grow or shrink. From mass conservation at the (sharp) cluster interface

$$|\Delta\psi|\frac{d(\pi R^2)}{d\tau}=-\Phi \ , \tag{A3}$$

where $\Delta\psi$ is the difference in the order parameter between the two equilibrium phases. Φ is the total flux *into* the surface of the cluster. The diffusion equation (A1) coupled with the moving boundary conditions (A2) and (A3) represent a formidable many-body problem. In order to proceed further, a mean-field approximation is made. Namely, the growth of a single cluster is considered, subjected to the conditions that the field satisfy (A2) at the cluster surface and that infinitely far from the cluster

$$\delta\psi|_{\infty}=\delta\bar{\psi} \ . \tag{A4}$$

$\delta\bar{\psi}$ is an average value of the field which reflects the presence of other clusters in the system. Under these conditions, the flux at the cluster surface is approximated by the asymptotic result[32]

$$\Phi=-(\delta\bar{\psi}-\delta\psi|_R)\frac{4\pi R}{R\ln 4\tau} \ , \tag{A5}$$

where it has been assumed that $R^2\ll 4\tau$. Combining (A2), (A3), and (A5) leads to an equation for the cluster radius

$$\frac{dR}{d\tau}=\frac{(\delta\bar{\psi}-\sigma/4R)}{R\ln 4\tau} \ . \tag{A6}$$

This expression reduces to the same form as LS if a transformation is made to a new unit of time defined

$$t=\text{li}(4\tau)/4\sim\frac{\tau}{\ln(4\tau)} \ , \tag{A7}$$

where $\text{li}(x)$ is the logarithmic integral.[33]

The dynamics of $\delta\bar{\psi}$ is established by considering the entire ensemble of clusters. It is useful to define a cluster distribution function $F(R,t)$ such that $F(R,t)dR$ measures the number of clusters per unit area with a radius between R and $R+dR$. Assuming no nucleation of new clusters and no coalescences, this function will obey a continuity equation

$$\frac{\partial F}{\partial t}+\frac{\partial}{\partial R}(\dot{R}F)=0 \ . \tag{A8}$$

Finally, the conservation law must be imposed on the entire system. This restriction leads to

$$\delta\bar{\psi}+\pi\int R^2F(R,t)dR=1-\psi_0 \ . \tag{A9}$$

Equations (A6), (A8), and (A9) can be solved using arguments similar to Ref. 3.

The conservation law imposes a unique scaling form to the cluster distribution function

$$F(R,t)=t^{-3n}f_0(x) \ , \tag{A10}$$

where

$$x=R\left[\frac{9\sigma t}{16}\right]^{-n}$$

and

$$n=\tfrac{1}{3} \ .$$

The time-independent scaling distribution function is

$$f_0(x)=\begin{cases}\dfrac{Cx^2}{(1.5-x)^{28/9}(3+x)^{17/9}}\exp\left[\dfrac{-1}{(1.5-x)}\right], & \text{for } x<1.5,\\ 0, & \text{for } x>1.5,\end{cases} \tag{A11}$$

where C is a normalization constant.

As a consequence of (A10), the time dependence of various moments of the cluster distribution function are simply related. For example, the average cluster radius satisfies

$$\langle R \rangle \equiv \frac{\int RF(R,t)dR}{\int F(R,t)dR} = t^n R_0 , \tag{A12}$$

where R_0 is a time-independent constant given by

$$R_0 = \left[\frac{9\sigma}{16} \right]^n \frac{\int x f_0(x)dx}{\int f_0 dx} . \tag{A13}$$

Similarly, the number of clusters per unit area decays according to

$$N(t) \equiv \int F(R,t)dR \sim N_0 t^{-2n} . \tag{A14}$$

(It should be noted that the scaling form of the distribution function is a more general ansatz for growth during phase transitions, applicable to systems in which the evaporation condensation mechanism described by LS is not applicable.)

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