Monte Carlo renormalization-group study of spinodal decomposition: Scaling and growth

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The kinetics of domain growth during the late stages of spinodal decomposition is studied by the Monte Carlo renormalization-group technique. A block-spin transformation is applied to the evolving configurations of the two-dimensional kinetic Ising model with conserved order parameter. This acts to renormalize the growing domains, the moving interfaces between them, and the coupled long-range diffusion fields. The growth law for the average size of domains, $R(t)\sim t^n$, where t is time, is determined self-consistently by a matching condition. The result for the growth exponent, $n=0.338\pm0.008$, is consistent with the classical result of Lifshitz and Slyozov for Ostwald ripening, namely, $n=\frac{1}{3}$. A scaling form for the structure factor is obtained which is invariant under the renormalization-group transformation, to the accuracy of our study. For large wave numbers k, it is found that the scaled form of the structure factor F is in good agreement with Porod's law; i.e., $F(kR)\sim 1/(kR)^{d+1}$, in $d=2$ dimensions.

I. INTRODUCTION

There is considerable interest, both theoretically and experimentally, in the growth kinetics which occur during first-order phase transitions.[1] Typically, a system is rapidly quenched from a high-temperature disordered state to a temperature well below its ordering temperature T_c. The system then evolves from this initial state to its final equilibrium state, consisting of two coexisting phases. If phase separation involves the growth of order from an *unstable* state, the dynamical process is called spinodal decomposition.

In spinodal decomposition, a long-wavelength instability creates a network of interpenetrating and interconnected domains which coarsen and grow to macroscopic size as time evolves. This is shown in Fig. 1. Experiments and computer simulations find that, for late times, the growth of ordered domains involves a single, time-dependent length to which all spatial dependencies scale.[1] This length is the average domain size $R(t)$. For example, the order-parameter correlation function $g(r,t)$, defined explicitly below, which is a function of spatial position r and time t, satisfies $g(r,t)\approx G(r/R(t))$. Indeed, one can see in Fig. 1 that if one were to shrink one of the configurations at late times, the system is similar to itself at an earlier time. Furthermore, domain growth often satisfies power-law growth,

$$R(t)\sim t^n , \tag{1}$$

for late times, where n is the growth exponent. It is generally believed that n and time-independent shape functions like G are universal features of the growth kinetics of first-order phase transitions. In other words, many physical systems, if they are members of the same universality class, share these kinetic characteristics. Here we study the universality class of "model B," where the scalar order parameter is a conserved quantity. Theoretical representations of this class include the spin-exchange kinetic Ising model, and a nonlinear Langevin equation which is sometimes called the Cahn-Hilliard-Cook equation; one important experimental representation is provided by phase-separating binary alloys. At the present time, the nature of scaling and growth for spinodal decomposition are controversial. To address these issues from first principles, we have carried out a Monte Carlo renormalization-group study of spinodal decomposition.

The Monte Carlo renormalization-group method was originally developed by Ma[2] and extended by Swendsen

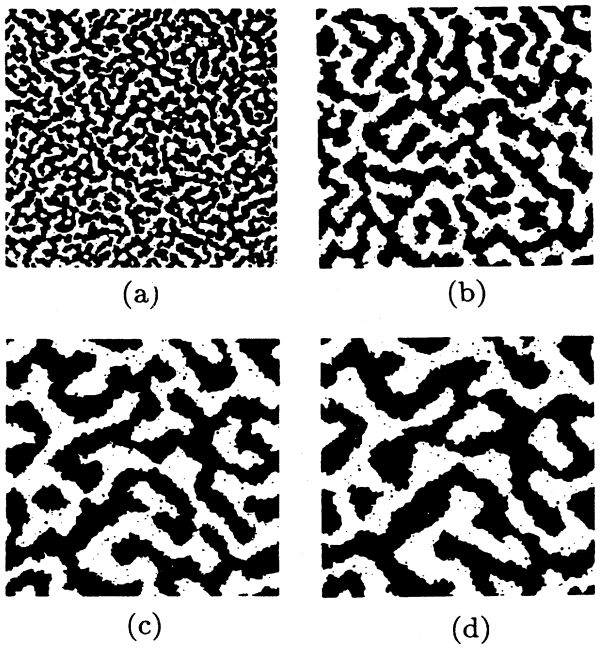

FIG. 1. Configurations at times 1000, 20 000, 100 000, and 160 000 MCS's.

and others,[3] who applied it to a variety of equilibrium problems. Tobochnik, Sarker, and Cordery[4] have extended this technique to the study of dynamical critical behavior. It was also used to study order-disorder transitions in the two-dimensional kinetic Ising model[5] and the Potts model on a square lattice.[6] The method involves matching correlation functions at different levels of renormalization using a conventional block-spin transformation. We generalize and extend it to the present problem below.

In contrast to this standard renormalization group, a novel renormalization-group method has been developed by Mazenko, Valls, and co-workers[7,8] for growth kinetics. They consider the behavior of finite-size systems embedded in an "infinite" lattice. Monte Carlo simulations are then used with analytic theory to both verify a scaling relation for the average domain size and to determine a time-rescaling factor. Use of this method initially predicted a logarithmic growth law $R(t)\sim \ln t$ for the kinetic Ising model of spinodal decomposition[7] (this result has now been withdrawn in favor of $n=\frac{1}{3}$). A more recent perturbation expansion approach for Langevin-type models yielded $R(t)\sim t^{1/4}$.[8] While it has been thought that both of these models would describe the same phenomena (at least so far as universal long-wavelength, long-time properties are concerned), the implication of these results is that the two models belong to different universality classes. We should note, however, that although a recent paper by Mazenko, Valls, and Zannetti[8] obtains $n=\frac{1}{4}$ in a more detailed account of their theory, they mention they cannot rule out the possibility that this is a problem for which their perturbation theory could fail, and the growth law could then differ from $n=\frac{1}{4}$.

These results are controversial, because the classical theory of Lifshitz and Slyozov[9] for Ostwald ripening during the late stages of nucleation and growth predicts $n=\frac{1}{3}$. While this a theory for the late-stage growth from an initially *metastable* state, and so is different from spinodal decomposition, the problems are related: it is expected that growth is determined by long-range diffusion at late times, which gives $n=\frac{1}{3}$ as we shall discuss below, rather than by the nature of the initial instability. Recently, several groups have studied this issue by different methods.[10] In a Monte Carlo study of the kinetic Ising model, Huse[11] obtained $n=0.29\pm0.04$ by extrapolating his results for finite domain size with a Gibbs-Thomson form for n, $n(R)=n(R\rightarrow\infty)+C/R$, where C is a constant. He suggested that the finite-domain-size correction is due to short-range diffusion along interfaces. Amar, Sullivan, and Mountain[12] obtained $n=0.330\pm0.005$ in a careful and extensive Monte Carlo study. A finite-size scaling study by Viñals and Jasnow[13] was also consistent with $n\approx\frac{1}{3}$. For Langevin models, recent work in dimension $d=2$ by Gawlinski, Viñals, and Gunton,[14] Rodgers, Elder, and Desai,[15] and in three dimensions by Toral, Chakrabarti, and Gunton[16] found $n\approx\frac{1}{3}$. Oono and Puri[17] have found the same result in studies of cell-dynamical systems, which are computationally optimized Langevin-like models. Finally, and we believe most significantly, recent neutron-scattering work on Mn-Cu by Gaulin, Spooner, and Morii[18,19] observed $n=\frac{1}{3}$.

Nevertheless, while this work addresses some aspects of scaling, it does not do so from first principles, as the work by Mazenko, Valls, and co-workers attempted to do. This situation underscores the need for a more extensive study, which directly addresses scaling and the nature of the renormalization group for spinodal decomposition. Therefore we have carried out a Monte Carlo renormalization-group study of the two-dimensional kinetic Ising ferromagnet with a conserved order parameter.[20] We consider the standard block-spin transformation. A description of our method is given in Sec. II.

In Sec. III we give our results for the growth law. We motivate our results with a physical picture, and present arguments for the value of n. Our analysis makes use of several measures of domain size: the inverse perimeter density, the first zero of the pair-correlation function, and the first and second moments of the structure factor. All of these exhibit a growth law consistent with the classical value of $n=\frac{1}{3}$, although some measures of domain size are sensitive to transient behavior, which is iterated away by successive application of the renormalization-group transformation. Our result for the growth law, $n=0.338\pm0.008$, is consistent with the classical result of Lifshitz and Slyozov, namely $n=\frac{1}{3}$. We do, however, observe strong transients, which can give an effective exponent of $n\approx\frac{1}{4}$ for analysis over a limited time regime. This transient is shared by the Langevin model for spinodal decomposition, and suggests that the kinetic Ising model and the Langevin equation may indeed share the same universality class. Our results are therefore consistent with recent theoretical, numerical, and experimental studies.[11–18]

The dynamic scaling of the structure factor is studied in Sec. IV. A scaling form for the structure factor is obtained, which is invariant under the renormalization-group transformation. We discuss its behavior for small and large wave numbers, and analyze the shape of the scaling function. For large wave numbers k, it is found that the scaled form of the structure factor F is in good agreement with Porod's law, i.e., $F(kR)\sim 1/(kR)^{d+1}$ in $d=2$ dimensions, which is a form factor for scattering from thin structureless interfaces. Finally, in Sec. V, we conclude the paper with a brief discussion of the fixed points of the renormalization group, and our results.

II. METHOD

The Hamiltonian of the two-dimensional ferromagnetic Ising model is

$$\mathcal{H}=-J\sum_{\langle ij\rangle}\sigma_i\sigma_j\ , \tag{2}$$

where J is the interaction constant, the sums run over distinct nearest-neighbor pairs on a square lattice, and the N spins can take on values of $\sigma_i=\pm1$. The system is quenched from infinite temperature to a low temperature T. Following the quench, the system evolves by Kawasaki spin-exchange dynamics: a pair of randomly selected nearest-neighbor pairs are exchanged if there is

no increase in energy, or with probability $\exp(-\delta E/T)$, if the exchange increases the lattice energy by δE, where Boltzmann's constant has been set to unity.

The simulations were performed on a Cray XMP. To utilize the vector capabilities of the Cray, a multispin-coding algorithm similar to that used in Refs. 12 and 21 was used. Using the binary representation of an Ising spin as one bit of a 64-bit word, we were able to simultaneously process 64 independent lattices. Logical operations, with "demon bits"[21] drawn from a large table of random numbers, were used to store and update the spins. Each of the 64 lattices was broken up into a 4×4 sublattice. Sets of corresponding spins on each sublattice made up one of 16 vectors. One vectorized update then corresponded to attempting to exchange the spins from one randomly chosen vector with the spins from a nearest-neighbor vector. Updates of N spins then made up one Monte Carlo step (MCS). Periodic boundary conditions were imposed on the square lattice.

Lattices of size $N=256^2$ and $N=128^2$ were simulated at $T=0.9T_c$. At this temperature, the interfaces are significantly rough, although the system is still far from the critical region. Furthermore, since the domain walls are rather diffuse, we expect the Ising model and Langevin model to be most similar at this temperature. At lower temperatures, the underlying lattice in the Ising model can cause important, though nonuniversal, differences between the two models. The results for the smaller system, which we studied over 60 000 MCS's, were averaged over 128 independent runs. On the larger lattice, results are averaged over 128 runs for the first 46 000 MCS's and 64 runs thereafter out to 280 000 MCS's. We also studied $N=128^2$ systems at $T=0.7T_c$, $T=0.4T_c$, and $T=0.1T_c$ out to 60 000 MCS's. Each of these systems was averaged over 64 independent runs.

The average domain size $R(t)$ was monitored in several different ways: from the inverse perimeter density, the first zero of the pair-correlation function, and the first and second moments of the circularly averaged structure function. The inverse perimeter density $R_e(t)$ is defined by[22]

$$R_e(t)=\frac{2}{2+E/J}\ , \tag{3}$$

where $E=\langle \mathcal{H}/N\rangle$ is the average energy per bond, and the angular brackets denote an ensemble average. Since R_e determines a length scale from the number of broken bonds, it measures the domain size explicitly in terms of the thermal correlation length ξ. It is sensitive to short-range effects. The pair-correlation function $g(r,t)=\langle\sigma(r,t)\sigma(0,t)\rangle$ was calculated along the x and y directions and along the two body diagonals where $x=y$. The first zero for each direction was then estimated by fitting $g(r,t)$ to a quadratic function. An average of these was taken as an estimate of the domain size[11] $R_c(t)$ from

$$g(R_c,t)=0\ . \tag{4}$$

This length scale was relatively insensitive to short-range fluctuations. Data for $R_e(t)$ and $R_c(t)$ were sampled ever 50 Monte Carlo steps.

We also obtained the two-dimensional structure factor, which is the Fourier transform of $g(\mathbf{r},t)$,

$$s(\mathbf{k},t)=\left\langle \frac{1}{N}\left[\sum_{\mathbf{r}_i}\sigma(\mathbf{r}_i,t)e^{i\mathbf{k}\cdot\mathbf{r}_i}\right]^2\right\rangle , \tag{5}$$

where $\mathbf{k}=(2\pi/L)(m\hat{\mathbf{i}}+n\hat{\mathbf{j}})$, $m,n=1,2,\ldots,L$, and L is the system size. The circularly averaged structure factor, which we examined in more detail, is

$$S(k,t)=\sum_{\hat{\mathbf{k}}}s(\mathbf{k},t)/\sum_{\hat{\mathbf{k}}}1\ , \tag{6}$$

with $k=2\pi n/L$, $n=0,1,2,\ldots,L$ and the sum $\sum_{\hat{\mathbf{k}}}$ is over a spherical shell defined by $n-\frac{1}{2}\le|\mathbf{k}|L/(2\pi)<n+\frac{1}{2}$. Note that the resolution of $S(k,t)$ depends on k_c, the cutoff frequency associated with the lattice. In our case, $k_c=(2\pi/L)n_c$, where we have chosen n_c to be the maximum possible value which is half the lattice size. From the first and second moments of $S(k,t)$, two further measures of domain size are defined as $R_1(t)=2\pi/k_1(t)$ and $R_2(t)=2\pi/k_2(t)$. The pth moment is defined as

$$[k_p(t)]^p=\frac{\sum_{k=0}^{k_c}k^pS(k,t)}{\sum_{k=0}^{k_c}S(k,t)}\ , \tag{7}$$

where $k=2\pi n/L$, and $n=0,1,2,\ldots,L$. Finally, for all the measures of domain size, we calculated the variance from the fluctuations of each run: $\Delta R(t)=(\langle R^2\rangle-\langle R\rangle^2)^{1/2}$. From this, the estimated statistical error δR was taken to be $\delta R=\Delta R(t)/\sqrt{\mathcal{N}-1}$, where $\mathcal{N}$ is the number of independent runs.

We used the renormalization group to exploit the scale invariance of the evolving system: the system is invariant, provided we rescale both length and time in an appropriate way. The relationship between these rescalings is, of course, implicit in the growth law. The renormalized lattices were obtained by a block-spin transformation, with length rescaling factor $b=2$. The majority-rule transformation was used to generate new cell spin variables from the original spin configurations. There one chooses a renormalized spin from a block of b^d spins by letting the spins vote, and the majority rule. "Ties" were broken by randomly assigning block spins the value ±1.[23] Figure 2 shows some typical results. This Wilson-type transformation explicitly renormalizes the domains, the moving interfaces between them, and the coupled long-range diffusion fields. It iterates away behavior on short length scales, thus allowing one to investigate the asymptotic large-length-scale properties of the system. Roughly speaking, spinodal decomposition involves at least two length scales: the average domain size $R(t)$ and the width $w(t)$ of the interfaces. As the domains get larger, w/R tends to zero, and corrections to scaling due to w become negligible. The renormalization-group transformation iterates away the small length scale w. This can be seen in Fig. 2. In this sense, the evolving system approaches a zero-temperature fixed point, since the nonzero width of w is essentially due to thermal fluctua-

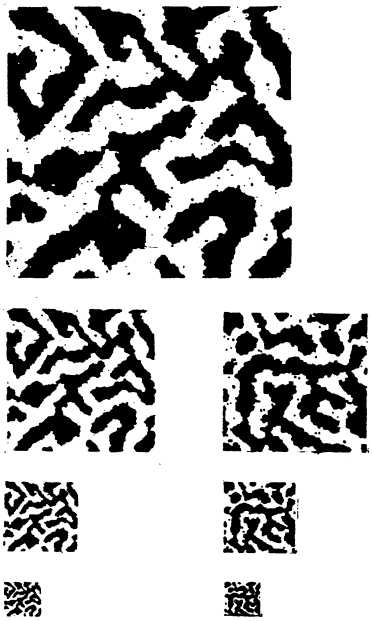

FIG. 2. Configurations on the left for $N=256^2$ system at $t=160\,000$ MCS's as it is renormalized. Configurations on right for $N=128^2$ system at $t=20\,000$ MCS's as it is renormalized. Note the similarity of the configurations with this choice of time rescaling factor.

tions which roughen the interface, i.e., $w\sim\xi$. It is important to note, however, that there can be microscopic activated processes at low temperatures in the Ising model, which are not generated by successive iterations of the renormalization group as one approaches the zero-temperature, strong-coupling fixed point.

We determine the growth law with a matching procedure. In principle, after the irrelevant variables have been iterated away, the probability distribution function will remain invariant under further renormalization-group transformations. It is expected that, after a finite number of iterations, contributions from the irrelevant variables will be negligible. Then, any quantity determined after m blockings of an N-spin system should be identical to those determined after $m+1$ blockings of a system of Nb^d spins. However, since the larger lattice has been renormalized once more, quantities will be at different times[24] t and t'. Hence, close to the fixed point, one can expect a matching condition to hold:

$$R(N,m,t)=R(Nb^d,m+1,t') . \tag{8}$$

From this the time rescaling factor t'/t can be calculated, and the growth exponent can be obtained, since

$$\frac{t'}{t}=b^{1/n} . \tag{9}$$

III. THE GROWTH LAW

The numerical value of the growth exponent n is the signature of the mechanism driving phase separation. During the late stages of spinodal decomposition the system is in a far-from-equilibrium state with many interfaces and thus a large amount of surface free energy. The system decreases its free energy as domains of ordered phase grow. The dynamics of this process is sensitive to the presence or absence of conservation laws.

If the order parameter is nonconserved, *unlike* the situation we are considering, it is relatively easy to obtain n. Then, the interfacial motion acts to reduce local surface area; curved interfaces move, and when part of an interface becomes flat, it stops moving. The interface velocity v is determined by the natural thermodynamic variables of the interface. In the most simple case, v is a function of only the local curvature K. (In two dimensions, the mean curvature K equals $1/R_K$, where R_K is the radius of curvature of an interface, while K is the sum of the inverses of the two radii of curvature in three dimensions.) At late times interfaces become gently curved, so K is small. Thus one has, $v(K)=DK+\cdots$, to leading order, where D is a positive constant.[25] Since the units of K and v are cm^{-1} and cm/sec, respectively, D has units of cm^2/sec. Therefore any length made with D, the only parameter in the equation of motion, must obey, $R(t)=(Dt)^{1/2}$, and $n=\frac{1}{2}$ for the nonconserved system.

In the case of a system with a conserved order parameter, some of these consideration still apply. The velocity of the interfaces is such that, on average, the curvature is reduced, since the motion of the interfaces is driven by the minimization of surface free energy. However, when the order parameter is conserved, the interfacial motion is coupled to the motion of material "under" the interface. For an interface to flatten, material within curved regions must diffuse away. Thus, although $v=DK$, now the diffusion constant itself must become, say, a function of curvature. In their most general form, the interface equations of motion for a conserved order parameter can involve the coupled nonlocal motion of many widely separated domains, and are quite interactable.[26] There are two important situations, however, where simplifications occur: long-range and short-range diffusion.

During nucleation and growth, all domains are circles in $d=2$ or spheres in three dimensions. Clearly, if all domains are of the same size, say $R_a=1/K_a$, the conservation law forbids growth by minimization of local surface free energy. Thus, to leading order, $D=D(K)=D_0(K-K_a)+\cdots$, where D_0 is a constant, and K_a is related to the average curvature of the domains in the system. The equation of motion becomes

$$v=D_0(K-K_a)K+\cdots . \tag{10}$$

This straightforwardly yields $n=\frac{1}{3}$. Note that large droplets, with $K<K_a$, grow through the accretion of material diffusing through the background matrix from disappearing small droplets, $K>K_a$. Thus this mechanism for phase separation is long-range diffusion between domains by evaporation and condensation. It was identified and explained in the elegant theory of Ostwald ripening due to Lifshitz and Slyozov.[9]

Although we expect long-range diffusion to be the dominant mechanism of phase separation for late times,

it is likely that other kinetic mechanisms are important during the intermediate stages of growth. In particular, we expect transient behavior involving short-range diffusion of material along interfaces. As shown in a study of sintering by Mullins,[27] this leads to an $n=\frac{1}{4}$ growth law. Consider a particle moving along an interface. It changes its chemical potential according to $\mu=\mu_0+CK$, where μ_0 is the chemical potential of a flat interface and C is a constant. Gradients of μ along the interface are therefore associated with gradients of curvature. The flux of particles, $\mathcal{J}$, is obtained through use of $\mathcal{J}\sim-\nabla_s\mu\sim-\nabla_s K$, where the gradient is taken with respect to the arc length of the surface. The surface divergence of $-\mathcal{J}$ yields the increase in the number of particles per unit area per unit time. Therefore the velocity of a surface element along its normal due to surface diffusion is

$$v=D_s\nabla_s^2 K\ , \tag{11}$$

where D_s is a constant. Counting dimensions gives $n=\frac{1}{4}$ for short-range diffusion.

For spinodal decomposition, then, we expect short-range transient behavior involving $n=\frac{1}{4}$, crossing over to long-range behavior with $n=\frac{1}{3}$, for late times. This scenario is consistent with Huse's picture[11] of growth during phase separation. He proposes that the equation of motion is

$$v=\frac{\partial R(t)}{\partial t}=\frac{C}{R^2(t)}+\frac{D}{R^3(t)}+\cdots\ , \tag{12}$$

where C and D are constants. The first term is due to the Lifshitz-Slyozov long-range diffusion, while the correction term is due to short-range diffusion. The effective exponent, $n_{\rm eff}=d[\ln R(t)]/d(\ln t)$, then has the form $n_{\rm eff}=\frac{1}{3}-D/[6CR(t)]+\cdots$. This Gibbs-Thomson[28] form is natural since, like all thermodynamic quantities on domains of finite size, n should be modified by its dependence on the local curvature $1/R$.

It is also of interest to know if inequalities can be found for the growth exponent. An argument has been given by one of us[29] for a bound on n in terms of the dynamical critical exponent z. If one quenches close to T_c, time scales are affected by critical slowing down. Milchev, Binder, and Heermann[30] have shown this can be incorporated into the growth law, $R\sim t^n$, using a simple scaling argument: near T_c time scales must be measured in units of the correlation time τ, and length scales in units of the correlation length ξ. This gives $R/\xi\sim(t/\tau)^n$. The dynamical scaling assumption is[31] $\tau\sim\xi^z$. Thus[30] $R/t^n\sim(T_c-T)^{\nu n(z-1/n)}$, where ν is the correlation length exponent. It is clear, however, that since the driving force for a first-order phase transition is due to thermodynamic forces, and not fluctuations, thermal fluctuations can only slow down domain growth, or leave it unaffected. Certainly, fluctuations will not speed up growth. Thus one obtains the inequality, $z\geq 1/n$. For model B there is an exact result from critical dynamics:[31] $z=4-\eta$, where η is the correlation function exponent. This give $n\geq 1/(4-\eta)$. This provides a good bound on n, since $\eta=\frac{1}{4}$ in two dimensions.

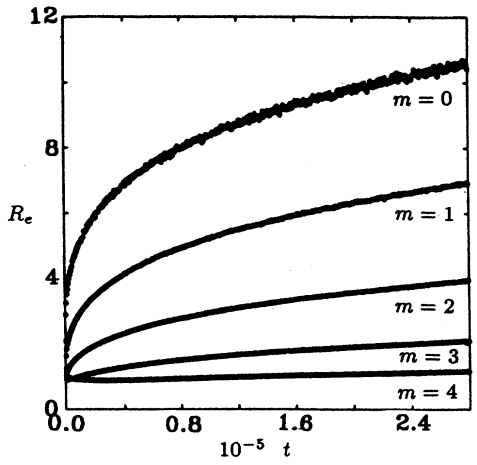

FIG. 3. $R_e(t)$ vs time for $N=256^2$ at $T=0.9T_c$. Levels of renormalization indicated by m. Note that renormalization reduces noise in the data.

An upper bound on the value of n has been given in a nice study by Yeung.[32] He considered the nonlinear Langevin equation for spinodal decomposition. He found that a self-consistent requirement of being in the scaling regime was that $n\leq\frac{1}{3}$. He also obtained a strong bound on the behavior of the scaling function at small values of its argument, which is discussed in the next section. Finally, it should be noted that Oono and Bahiana[33] have recently argued that the power law for copolymer lamellar thickness implies that $n=\frac{1}{3}$.

In order to obtain independent estimates of the nature of growth in the scaling regime, we performed our analysis in several ways. First, we looked for the best fits to the renormalized data. Second, we matched R's on different lattices at different levels of renormalization as

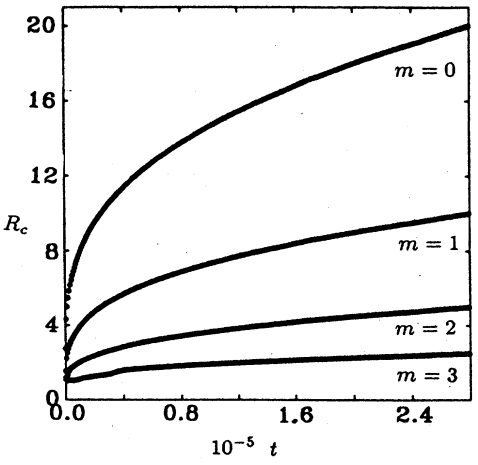

FIG. 4. $R_c(t)$ vs time for $N=256^2$ system at $T=0.9T_c$. All levels of renormalization shown, indicated by m, fit well to $R_c(t)=A+Bt^{1/3}$.

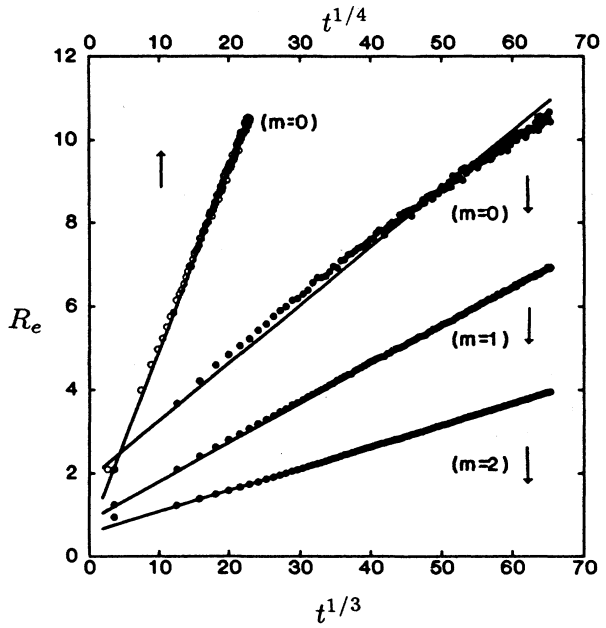

FIG. 5. R_e vs $t^{1/4}$ or $t^{1/3}$, as the configurations are renormalized. Best fits are shown. Lines are identified by the number of rescalings m.

outlined in Sec. II. Finally, we assumed that $n = \frac{1}{3}$, and seen with this choice of time rescaling factor, whether or not the R's at different levels of renormalization approached each other as we extrapolated to $1/R \to 0$. In the regime we investigated, the domains had grown to a size $R \sim 10$–20 lattice constants. We therefore made use

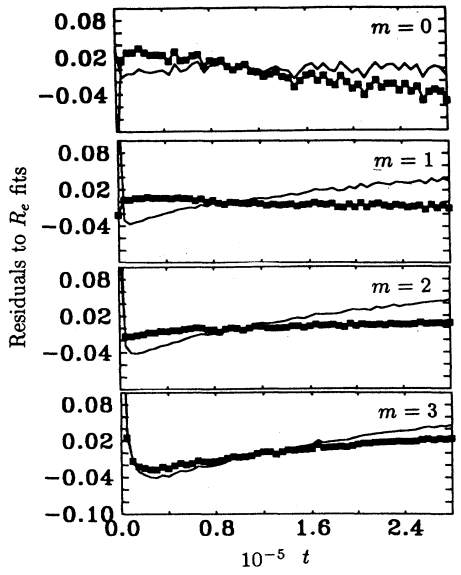

FIG. 6. Residuals of R_e data to fits of $n = \frac{1}{4}$ (lines) and $n = \frac{1}{3}$ (solid squares) for various levels of renormalization. Note R_e initially fits well to an $n = \frac{1}{4}$ power law, but as data is renormalized, fit to $n = \frac{1}{3}$ better.

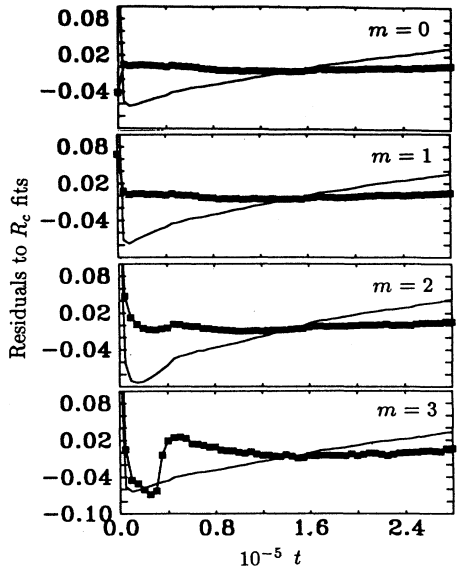

FIG. 7. Residuals of R_c data to fits to power laws $n = \frac{1}{4}$ (lines) and $n = \frac{1}{3}$ (solid squares) at various levels of renormalization. Fit by $n = \frac{1}{3}$ is significantly better.

of Gibbs-Thomson extrapolations of the form suggested by Huse. Our results for R_c, R_1, and R_2 were sensible down to 32^2 systems, and to 16^2 systems for R_e. Below this point, domain sizes were too small to obtain meaningful results.

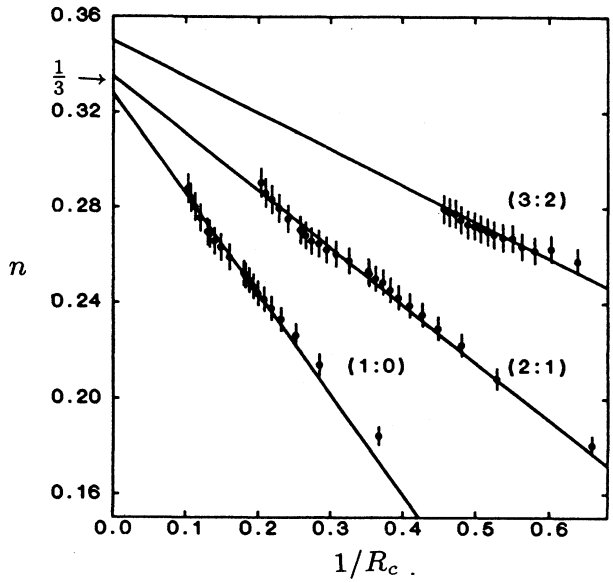

FIG. 8. Results of matching to estimate n with R_c. Lines are identified by levels of matching (m:m'), which are the number of rescalings on the large and small lattice, respectively. Every fifth data point is shown to 2500 MCS's, then every twentieth to 9500 MCS's, and every seventieth thereafter. Fits are obtained from the full data set. Error bars are estimated from three standard deviations.

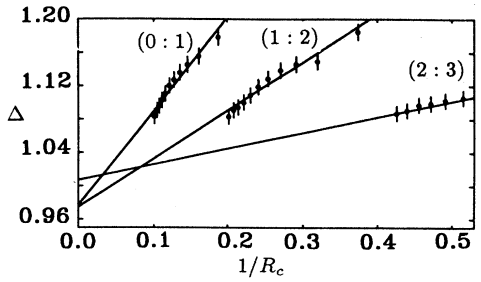

FIG. 9. Matching for R_c data with $n=\frac{1}{3}$. We calculate $\Delta=R(N,m,t)/R(Nb^d,m+1,tb^3)$ and fit this to $\Delta(R)=\Delta(R\rightarrow\infty)+C/R$. Lines are identified by levels of matching $(m:m')$, which are the number of rescalings on the large and small lattice, respectively.

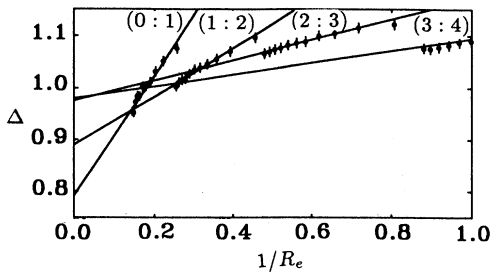

FIG. 10. Matching for R_e data with $n=\frac{1}{3}$, as in Fig. 9. Lines are identified by levels of matching $(m:m')$, which are the number of rescalings on the large and small lattice, respectively.

Figure 3 and 4 show plots of $R_e(t)$ and $R_c(t)$ as a function of time. The R_c data, at all levels of renormalization down to $m=3$, were fit exceedingly well by $R(t)=A+Bt^{1/3}$. Fits of the same form by $n=\frac{1}{4}$ and $R\sim \ln t$ were much worse. The R_e data showed more interesting behavior, because R_e is more sensitive to short-range fluctuations than R_c. Before renormalization, the R_e data were fitted better by $n=\frac{1}{4}$ than $n=\frac{1}{3}$. (See Fig. 5.) Note that the data are fairly noisy for $m=0$. After renormalization, the data are significantly smoother and fit $n=\frac{1}{3}$ better than $n=\frac{1}{4}$. Direct comparisons of the fits are given in Fig. 6 and 7, where the residuals of the data to the two fits are shown. The implication is that there exists a strong transient, which leads to an effective exponent of $n\approx\frac{1}{4}$, to which R_e is sensitive. This is in accord with the picture outlined above. To a lesser extent this trend is also observed in the R_1 and R_2 data. There, fits of the unrenormalized data by $n=\frac{1}{3}$ or $n=\frac{1}{4}$ were roughly equivalent. However, after renormalization, the data fitted better to $n=\frac{1}{3}$. A similar transient was also observed in simulations for Langevin equations.[14–17] While this does not prove that the Ising and Langevin models belong to the same universality class, it suggests that this may indeed be the case.

We have also examined the growth of R_e and R_c at quench temperatures of $T=0.4T_c$ and $T=0.7T_c$, finding that data for both measures of length give excellent fits to form $R=A+Bt^{1/3}$. Furthermore, we found the $n=\frac{1}{4}$ transient was considerably weaker at lower temperature. This is due to the reduced diffuseness of the domain walls at lower temperatures. The results of our fits are presented in Table I. Investigating spinodal decomposition at $T=0.1T_c$, we found that our data were dominated by early-time activated processes, and the asymptotic regime was never reached.

TABLE I. Fits by $R(t)=A+Bt^{1/3}$.

T	A_e	B_e	A_c	B_c
$0.9T_c$	1.96	0.14	1.84	0.28
$0.7T_c$	1.61	0.15	1.85	0.21
$0.4T_c$	1.85	0.06	2.14	0.07

Our most reliable estimate for the value of the growth exponent comes from the matching condition described in the preceding section. Note that the matching requires not only the exponent of growth to be the same as the evolving configurations are iterated, but also that the amplitude A of the growth becomes identical, where $R\sim At^n$. We obtained an effective n by determining the ratio of times on two lattices at different levels of renormalization, which gave equal domain sizes. Using this $n(R)$, we estimated the asymptotic exponent by generalizing Huse's formula, i.e., by fitting to $n(R)=n(R\rightarrow\infty)+C/R$. The data for R_c are presented in Fig. 8. The estimates for n from the first three levels of matching are approximately 0.328, 0.335, and 0.349. Averaging these gives us our best estimate for the growth exponent, 0.338 ± 0.008, which is in good agreement with the classical value of $\frac{1}{3}$. The error is estimated to be 3 standard deviations of the statistics in the data. We can also see that the renormalization-group transformation makes the constant C smaller as one would expect. The results for R_e are more sensitive to short-range effects (the renormalization group has to "work harder" to iterate away both the transient $n=\frac{1}{4}$ behavior and cause the amplitudes to converge). The first two levels of matching do not approach the same value of n. The third level of matching gives $n=0.36$, which is consistent with results for R_c, but unfortunately does not allow us to improve our estimate for the growth exponent. We cannot extrapolate a result from the fourth level of renormalization.

Finally, we assumed $n=\frac{1}{3}$, and checked whether R's at different levels of renormalization approached each other as $R\rightarrow\infty$. We calculated

$$\Delta=R(N,m,t)/R(Nb^d,m+1,tb^3)\ . \tag{13}$$

Motivated by Huse's formula, we fit this data to $\Delta(R)=\Delta(R\rightarrow\infty)+C/R$. See Figs. 9 and 10. The estimates for Δ from the first three levels of matching of R_c are 0.98, 0.98, and 1.01. The average is 0.99 ± 0.02 in good agreement with the result expected for perfect matching, $\Delta=1$. The R_e's give similar results. The first two levels do not converge, while the third and fourth

levels give $\Delta=0.98$. There is no convergence for $n=\frac{1}{4}$ or $n=0(\ln t)$.

IV. THE STRUCTURE FACTOR

For late times, all length scales have their time dependence set by the size of growing domains. Thus the pair-correlation function satisfies $g(r,t)\sim G(r/R(t))$, which implies that the structure factor obeys

$$S(k,t)=R^{d}(t)F(x)\ , \tag{14}$$

where $x=kR(t)$. This is because g is dimensionless, so S has units of volume. We know of no analytic theory for the shape of the scaling function $F(x)$.[34] Nevertheless, arguments can be given for its behavior at large[35,36] and small[32] values of x, and a physically motivated form can be obtained[37] which interpolates between these regimes. For small x, the conservation law causes $F(x)$ to vanish, or more precisely to be fixed to its initial value. This can be written in the form

$$F(x)\sim x^{\delta}\ . \tag{15}$$

Yeung[32] has obtained the inequality $\delta\geq 4$ in two and three dimensions, in the scaling study mentioned above. He finds that $\delta=4$ is satisfied by experiments he considers.

For large x, Porod's law,[35] which we derive below, gives

$$F(x)\sim\frac{1}{x^{\theta}}\ . \tag{16}$$

If the scattering is due to thin flat interfaces, as is the case here on small lengths scales, $\theta=d+1$. More generally, scattering from an interface of dimension d_A, which is embedded in a volume of dimension d_V, gives $\theta=2d_V-d_A$.

The arguments for Porod's law are straightforward, so we review them here. It is most simple to consider $g(r)$ for small r. The $g(r)$ function is correlated so long as the two points separated by r are in the bulk. Only the contributions near interfaces, $\delta g(r)$, affect this. Those contributions depend on the relative amount of surface to bulk, i.e., they scale as $\delta g(r)\sim(A/V)r^{d_V-d_A}$, where the area $A\sim R^{d_A}$, and the volume $V\sim R^{d_V}$. The factor of $r^{d_V-d_A}$ ensures that $g(r)$ is dimensionless. Fourier transforming introduces another factor of volume, and one obtains $S(k)\sim 1/k^{2d_V-d_A}$, which implies the scaling form above. Note that, for small r's as the lattice structure becomes important, the interface is flat so that $d_V=d$ and $d_A=d-1$, which implies $\theta=d+1$.

Roughening can give rise to a different effective value for θ on larger length scales, and thus smaller values of x. These correspond only to logarithmic corrections in three dimensions, but there are large effects in $d=2$. For roughening $A\sim R^{d-1}w(R)$, where w is the interface width. It is well known that $w\sim R^{(3-d)/2}$, on *large* length scales for $1<d<3$ (note, however, that we shall apply this result to behavior on *small* length scales to obtain an effective exponent). Thus, we estimate an effective $d_A=(d+1)/2$, so that $\theta=(3d-1)/2$, for $1<d<3$. In two dimensions this implies that, if a sufficiently large k is not considered, one may obtain $\frac{5}{2}\leq\theta_{\rm eff}<3$ rather than the asymptotic value of 3. In fact, some effective values for θ in this range have been reported.[37]

Fratzl and Lebowitz[36] have given a semiempirical form for the scaling function in *three* dimensions. It is of the form

$$F(x)=\frac{Ax^4}{B+x^4}f(x)\ , \tag{17}$$

where A and B are constants. The prefactor is to account for Yeung's result. $f(x)$ is obtained from the Fourier transformation, with respect to the kernel $e^{i\mathbf{x}\cdot\mathbf{r}}$, of

$$\hat{f}(r)\propto e^{-\lambda r}\frac{\sin\alpha r}{\alpha r}\ , \tag{18}$$

where λ and α are constants. The exponential and oscillatory factors are to account for both the randomness and modulation of the interfaces, respectively. In the absence of a result which has been explicitly derived for $d=2$, we have taken the *two-dimensional* Fourier transform of Fratzl and Lebowitz's expression to obtain

$$f(x)=\left[\frac{[(x^2-C)^2+D]^{1/2}-(x^2-C)}{[(x^2-C)^2+D]}\right]^{1/2}\ , \tag{19}$$

where C and D are constants. Note that Eqs. (17) and (19) then interpolate between Yeung's result and Porod's law in two dimensions.

Figure 11 shows the circularly averaged structure factor we have obtained. Note the increase in peak height and decrease of the peak position with increasing time,

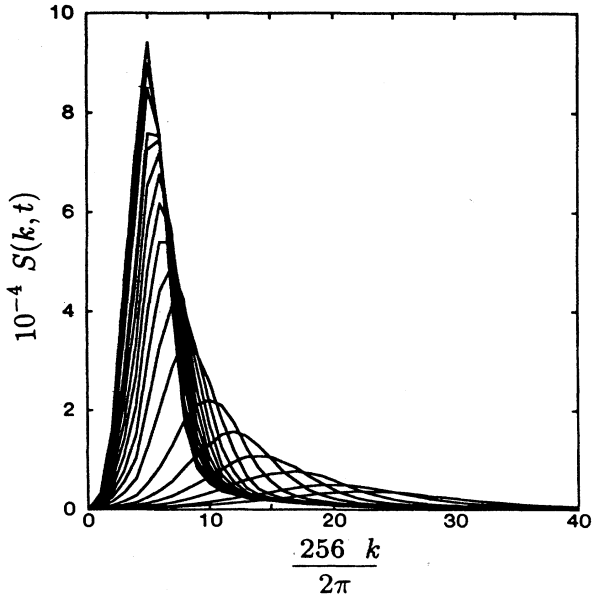

FIG. 11. Circularly averaged structure function for $N=256$ system at $T=0.9T_c$. Times shown are $t=500$, 1000, 2500, 5000, 10 000, 20 000, and then in steps of 20 000 MCS's up to 260 000 MCS's respectively.

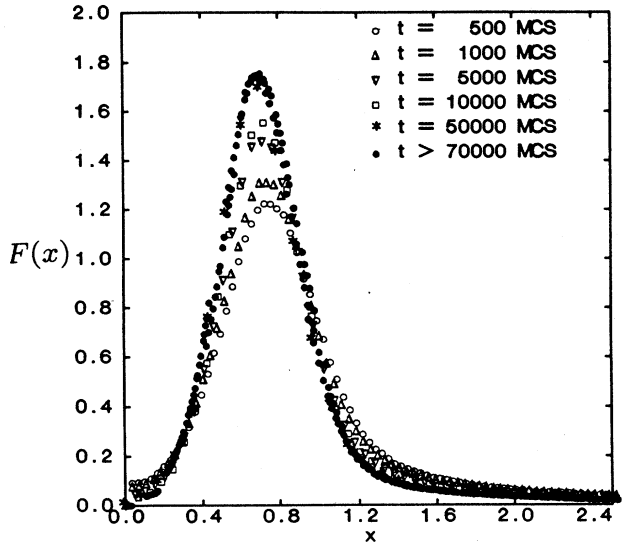

FIG. 12. $F(x)$ for various times showing the approach to scaling. From below, we show data for $t=500$, 1000, 5000, 10 000, and 50 000 MCS's. The top curve indicates data for times greater than 70 000.

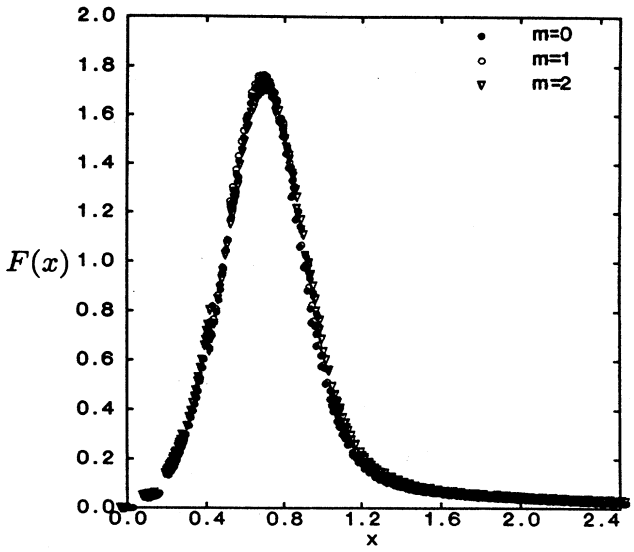

FIG. 14. Scaled structure factor $F(x)$ for late times at $m=0$, 1, and 2 levels of renormalization. Data are same as plotted in Fig. 13, except for a linear transformation applied to renormalized data. Note that $F(x)$ is invariant under renormalization.

while the $k=0$ mode is pinned due to the conservation law. These are characteristic features of spinodal decomposition. To test scaling, we have calculated

$$F(x,t)=(2\pi/L^2)k_1^2(t)S(xk_1(t)) \tag{20}$$

as a function of time. Following Amar, Sullivan, and Mountain,[12] the normalization constant for $F(x)$ was chosen by $\int xF(x)dx=1$. Figure 12 shows the approach to scaling. At $T=0.9T_c$, the peak height of $F(x,t)$ continues to increase for times up to about 70 000 MCS's. This indicates that up to this time, our system has not yet entered the scaling regime and that contributions due to the interface roughness are not negligible. After about 70 000 MCS's. $F(x)$ is independent of time, to the accuracy of our simulations. Figure 13 shows $F(x)$ for late times for $m=0$, 1, and 2 iterations of the renormal-

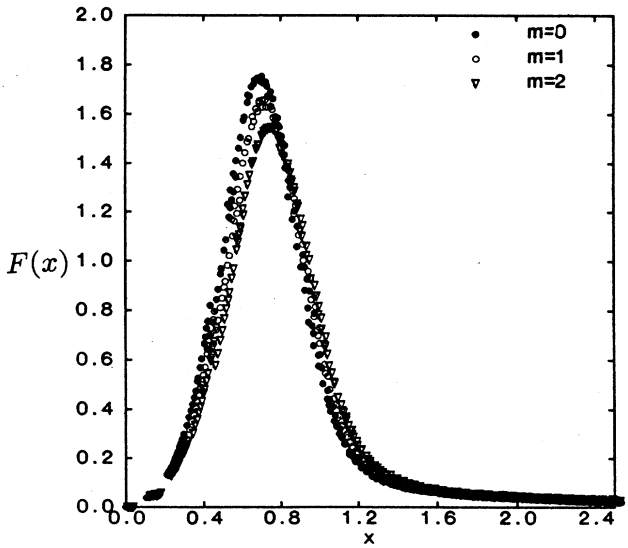

FIG. 13. Scaled structure factor $F(x)$ for late times at $m=0$, 1, and 2 levels of renormalization. Data plotted are in intervals of 10 000 MCS's between 100 000 and 250 000 MCS's.

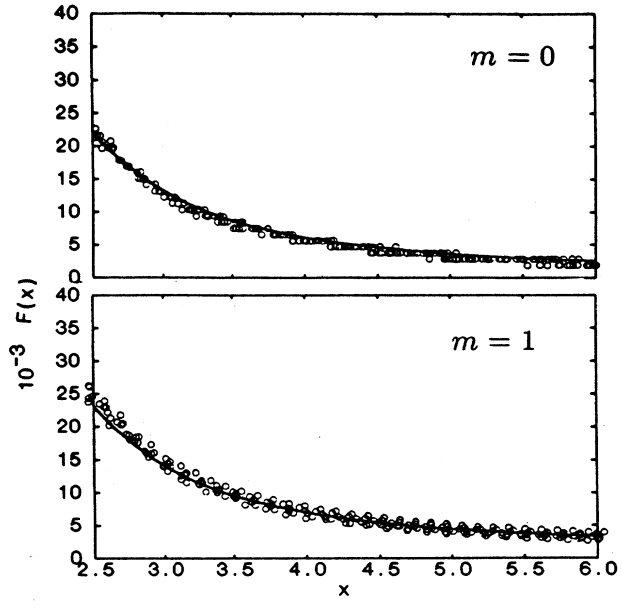

FIG. 15. Detailed view of the tail of the scaled structure factor $F(x)$ for $m=0$ and $m=1$. The solid curve shows the fitted $F(x)\sim 1/x^3$ behavior.

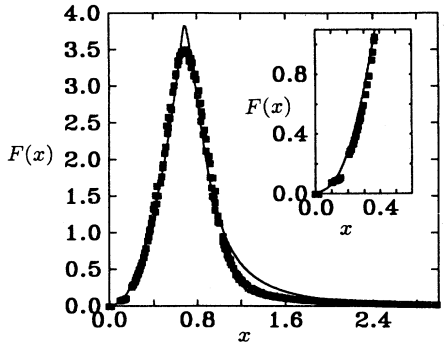

FIG. 16. Fit of $F(x)$ by Eqs. (17) and (19). Inset shows fit at small x.

ization-group transformation. We have not presented data for the $m=3$ level of renormalization due to the large amount of statistical error present there. The error is chiefly due to the loss of resolution incurred with each renormalization-group iteration, and is associated with the increasing cutoff frequency for the lattice. For any given level of renormalization, scaling appears to hold. However, the renormalized data exhibit a slight shift in peak position and a slight decrease in peak height. This involves only a linear transformation of the scaling function, as is shown in Fig. 14. To our accuracy, all the curves are coincident in that figure.

To investigate $F(x)$ in a more quantitative fashion, we have fitted our large x values ($x \geq 2.5$) to Eq. (16). Our results for $t > 100\,000$ and $m=0$ and 1 iterations of the renormalization-group transformation are in good agreement with Porod's law, $\theta = d+1$. We measure $\theta = 2.98 \pm 0.03$. See Fig. 15. For times less than 100 000 MCS's, our measured values of θ are somewhat below the expected value of 3. They range from 2.5 to 3.0. For $m=2$, and 3 levels of renormalization, our results are consistent with $F(x) \sim 1/x^3$ behavior. It is, however, difficult to unambiguously fit the large x data to a single power law. Tomita[35] has given a more detailed discussion of the conditions necessary to observe Porod's law.

We have also tried fitting the behavior of $F(x)$ at $x \to 0$. However, we have insufficient data close to the origin to make a good estimate of δ in Eq. (15). We find effective δ's in the range 2.5–3, which we attribute either to fitting too far from the origin or possibly to transients. (Recall that Yeung's inequality is $\delta \geq 4$). The insert in Fig. 16 shows that a fit by Eq. (17), where $\delta = 4$, is possible. Also in Fig. 16, we show a four-parameter fit by Eqs. (17) and (19) above. The fit is reasonably good, except for x approximately twice its value at the maximum of F. We emphasize again that the form was intended for three-dimensional scattering patterns; we have used it here to motivate further work on the two-dimensional problem. It should be noted that we have found that the fit is much better than one by a simple ratio of polynomials, i.e., $F(x) = Ax^4/(B+Cx^7)$, which also interpolates between small and large x regimes.

V. CONCLUSIONS

In this last section, we give a brief discussion of the fixed points of the renormalization group. The block-spin transformation renormalizes the master equation simulated by our Monte Carlo method. This leads to new non-Markovian equations of motion for the renormalized spins, which are exceedingly difficult to study analytically. Indeed, although we have made the natural assumption that scaling dominates the behavior at the fixed point, to our knowledge there exists no first-principles understanding of the nature of fixed points in first-order transitions. Nevertheless, some qualitative remarks can be made.

Under renormalization, one studies, say, the flow of coupling constants in a Hamiltonian or free energy. For spinodal decomposition, two sets of coupling constants, $\kappa_i = J/T_i$ and $\kappa_f = J/T_f$, must be taken into account, which respectively characterize the initial disordered state and the final low-temperature state. Clearly, as the renormalization group is iterated, the initial coupling constants κ_i flow to the infinite-temperature fixed point, while the final coupling constants κ_f flow to the zero-temperature fixed point.[7] In contrast to critical phenomena, the dynamics of first-order phase transitions involve stable and attractive fixed points.[38] However that may be, the system's dynamics at the zero-temperature fixed point are not the same as one would obtain by simply quenching to zero temperature. There can be activated processes as $T \to 0$ which are not generated by the successive application of the renormalization group. As has been suggested by Sadiq and Binder[22] and Huse,[11] these short-wavelength activated processes can cause long transients at low temperatures. In fact our data at $T = 0.1T_c$ are dominated by this transient behavior. In this regard, it should be noted that studies of domain growth in the nonconserved Potts model on a square lattice pointed to the existence of two fixed points at $T=0$.[6] The "freezing" fixed point led to an $R \sim \ln t$ growth, while the "equilibration" fixed point led to power-law growth. Only the equilibration fixed point was stable for quenches to nonzero temperatures, however, although the freezing fixed point could cause strong transients at low temperatures.

In conclusion, by using an extension of the standard Monte Carlo renormalization-group method, we were able to study two universal features of the growth kinetics of spinodal decomposition: the growth exponent n and the shape of the scaled structure function F. While more study is needed, for example we made use of Gibbs-Thomson relations to estimate the asymptotic growth exponent, it is clear that our results are in agreement with most of the recent work in the field. Our best estimate for the growth exponent, which we obtained through a matching condition, $n = 0.338 \pm 0.008$, is in agreement with the classical result of Lifshitz and Slyozov. We observed evidence of a strong transient involving an effective $n = \frac{1}{4}$, however. This transient behavior, which we attributed to short-range diffusion, was iterated away by the renormalization-group transformation, leaving $n = \frac{1}{3}$. Similar transients have been seen in studies of Langevin models.[14–17] While this does not demonstrate that the Langevin models and the kinetic Ising model share the same universality class, it indicates that this may indeed be the case. We investigated dynamical scal-

ing in the circularly averaged structure factor. We obtained the scaling function, and found that its shape remained invariant under the renormalization-group transformation. In the absence of predictions for the shape of this scaling function, our analysis has been essentially of a qualitative nature. However, a detailed investigation of the tail of the scaled structure function showed that its shape was consistent with Porod's law.

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