Monte Carlo Renormalization-Group Study of the Late-Stage Dynamics of Spinodal Decomposition

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The kinetics of spinodal decomposition has been studied by a Monte Carlo renormalization-group method. Using the standard blocking transformation, we numerically renormalize the evolving configurations during phase separation of a two-dimensional kinetic Ising ferromagnet with spin-exchange dynamics. We find that, as the scaling regime is approached, the domain size R grows in time t as $R \sim t^n$, where we obtain $n = 0.338 \pm 0.008$. This is consistent with the classical result of Lifshitz and Slyozov for Ostwald ripening, namely $n = \frac{1}{3}$.

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Spinodal decomposition, the dynamics of the growth of order from an unstable state during a first-order phase transition, is a problem of long-lasting interest and importance.[1] One prepares a system by rapidly quenching it from a high-temperature disordered state to a temperature well below its critical temperature T_c. A long-wavelength instability creates a morphology of interconnected interpenetrating domains of ordered phases (see Fig. 1). These domains grow as time goes on. Experiments and computer simulations indicate that, for late times, domain growth often involves a time-dependent length, the average domain size $R(t)$, to which scale all spatial dependences.[1] For example, the order-parameter correlation function $g(r,t)$ as a function of spatial position r and time t often satisfies $g(r,t) \approx G(r/R(t))$, for late times. Furthermore, $R(t)$ often satisfies a power-law form, $R(t) \sim t^n$, where n is the growth exponent. It is generally believed that n and the shape function G are two features of the kinetics of domain growth in first-order transitions which characterize a universality class. At the present time, the nature of scaling and the growth law for spinodal decomposition (domain growth with a conserved scalar order parameter, e.g., phase separation in binary alloys) are controversial. In this Letter, we present a Monte Carlo renormalization-group study of spinodal decomposition. This addresses the nature of growth and scaling from first principles.

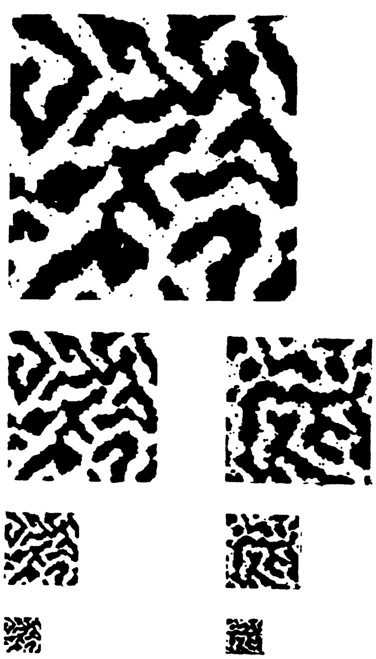

FIG. 1. Configurations on left for $N = 256^2$ system at $t = 160000$ Monte Carlo steps as it is renormalized. Configurations on right for $N = 128^2$ system at $t = 20000$ Monte Carlo steps as it is renormalized. Note the similarity of the configurations as they are renormalized with this choice of the time rescaling factor ($n = \frac{1}{3}$).

The Monte Carlo renormalization-group approach was introduced by Ma.[2] It was developed and extended by Swendsen and others,[3] who applied it to second-order phase transitions. It has since been applied to the study of other problems, including critical slowing down[4] and the dynamics of an order-disorder transition.[5] The method, which we shall discuss and extend below, involves the matching of correlation functions on different sized lattices at different levels of renormalization, using the conventional Wilson-type blocking transformation.

The nature of scaling in spinodal decomposition is not well understood. Recently, a number of interesting issues have been raised by Mazenko and co-workers.[6] They have invented a novel renormalization group for this problem which studies the behavior of small subsystems embedded in a larger lattice at different temperatures. Monte Carlo simulations are then used in conjunction with results for the thermal correlation length $\xi(T)$ to determine the growth law in the scaling regime. They predict a logarithmic growth law,[6a] $R(t) \sim \ln t$, for the kinetic Ising model of spinodal decomposition, and $R(t) \sim t^{1/4}$ for the corresponding Langevin model (which is often called model B).[6b] While it has been thought that both these models would describe the same phenomena (at least over long length scales and long times[1]), the implication of Mazenko and Valls's results is that the two models could belong to different universality classes.

These results are controversial, because the classical theory of Lifshitz and Slyozov for Ostwald ripening during the late stages of nucleation and growth,[7] a closely related problem, gives $n=\frac{1}{3}$. Recently, several groups have studied this issue. In a Monte Carlo study of the kinetic Ising model, Huse[8] obtained $n=0.29\pm0.04$, by extrapolating his results for finite domain size with a Gibbs-Thomson-type[9] form for n. He suggested that, like all local thermodynamic quantities on domains of finite size, n should be modified by dependence on the local curvature $1/R$ of a domain of finite size, i.e., $n(R\to\infty)=n(R)+C/R$, where C is a constant. In neutron scattering work on Mn-Cu, Gaulin, Spooner, and Morii[10] see agreement with Lifshitz-Slyozov behavior (using Huse's form for n). Amar, Sullivan, and Mountain[11] obtained $n=0.330\pm0.005$ in an extensive and careful Monte Carlo study. Finite-size scaling done by Viñals and Jasnow[12] also appears to be consistent with $n=\frac{1}{3}$. For Langevin models, recent work[13] by Oono and Puri, Rodgers, Elder, and Desai, and Gawlinski, Viñals, and Gunton finds $n\approx\frac{1}{3}$.

Nevertheless, while this work[8,11-13] addresses some aspects of scaling, it does not do so from first principles, as Ref. 6 attempts 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.

In this Letter, we apply the standard Monte Carlo block-spin-transformation renormalization group to spinodal decomposition for the first time. We study the dimension $d=2$ kinetic Ising model with a conserved order parameter. Our result for the growth law, $n=0.338\pm0.008$, is consistent with the classical result of Lifshitz and Slyozov, namely $n=\frac{1}{3}$. However we do observe strong transients, which can give an effective exponent of $n\approx\frac{1}{4}$ for analysis over a limited time regime.

Spinodal decomposition involves at least two length scales: the domain size and the width w of the interface. 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. 1, and has been discussed elsewhere.[5] In this sense, the evolving system approaches a zero-temperature fixed point, since the nonzero width of w is essentially due to thermal fluctuations which roughen the interface, i.e., $w\sim\xi$.

The Hamiltonian of the two-dimensional ferromagnetic Ising model is $H=-J\sum_{\langle ij\rangle}\sigma_i\sigma_j$, where J is the interaction constant, the sums run over distinct nearest-neighbor pairs, and the N spins take on values of $\sigma_i=\pm1$. After a quench from an infinite temperature to $T=0.9T_c$, the system evolves by Kawasaki spin-exchange dynamics. At this temperature, the interface is significantly rough, although the system is still far from the critical region. We note that it is natural to expect that the Ising model and the Langevin model would be most similar at these temperatures where the domain walls are rather diffuse. The simulations were performed on a Cray model X-MP with a multispin-coding algorithm similar to those described in the literature.[14] We studied lattices of sizes $N=256^2$ and 128^2. Results have been averaged over 128 runs for the smaller lattice over 60000 Monte Carlo steps. On the larger lattice, results are averaged over at least 64 runs out to 280000 Monte Carlo steps. Two measures of domain size were used. The inverse perimeter density is defined by $R_e(t)=2/(2+E/J)$, where E is the average energy per spin. Since this gives a length scale determined from the number of broken bonds, it measures the domain size explicitly in terms of ξ, and is sensitive to short-range effects.[15] The first zero of the spin-spin correlation function $g(r,t)$ (along the x,y, and $x=y$ directions of the lattice)[16] allows us to define a length R_c via $g(R_c,t)=0$. This length scale is relatively insensitive to short-range fluctuations. We used extrapolations of the form proposed by Huse, since, even after 280000 Monte Carlo steps, the domains have only grown to a size of $R\approx10$–20 lattice constants.

Our renormalized lattices were obtained by a majority-rule block-spin transformation of the evolving spin configurations. The length rescaling factor b was chosen to be 2. "Ties" were broken by our randomly assigning block spins the value ±1.[17] Since the system is approximately invariant under a change of length scales given a corresponding change in time scales, the relationship between the two rescalings gives the growth law. In principle, after the irrelevant variables have been iterated away, the system will be invariant under a renormalization-group transformation. 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 N spins should be identical to those determined after $m+1$ blockings of Nb^d spins. However, since the time scale in the larger lattice has been renormalized once more, quantities will be at different times t and t', i.e., $R(N,m,t)=R(Nb^d,m+1,t')$. This is our matching criterion. The ratio of times gives the time rescaling factor from which we obtain the growth exponent, i.e., $t'/t=b^{1/n}$.

Before doing the matching, we have simply looked for the best fits to the renormalized data. The R_c data, at all levels of renormalization down to $m=3$, are fitted exceedingly well by $R(t)=A+Bt^{1/3}$. Fits by the same form with $n=\frac{1}{4}$ are significantly worse; two-parameter fits by $R\sim\ln t$ are much worse. The R_e data show more interesting behavior because R_e is more sensitive to short-range correlations than R_c. Before renormalization, the R_e data are fitted better with $n=\frac{1}{4}$ than with $n=\frac{1}{3}$ (see Fig. 2). Note that the data are fairly noisy for $m=0$. After renormalization, however, the data are

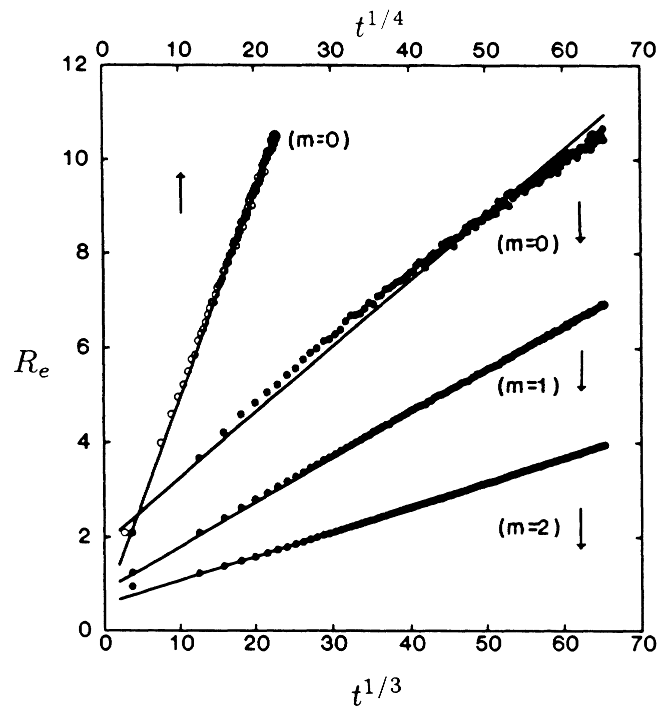

FIG. 2. R_e vs $t^{1/4}$ or $t^{1/3}$, as the configurations are renormalized. Best fits are shown. Lines are identified by the number of rescalings m.

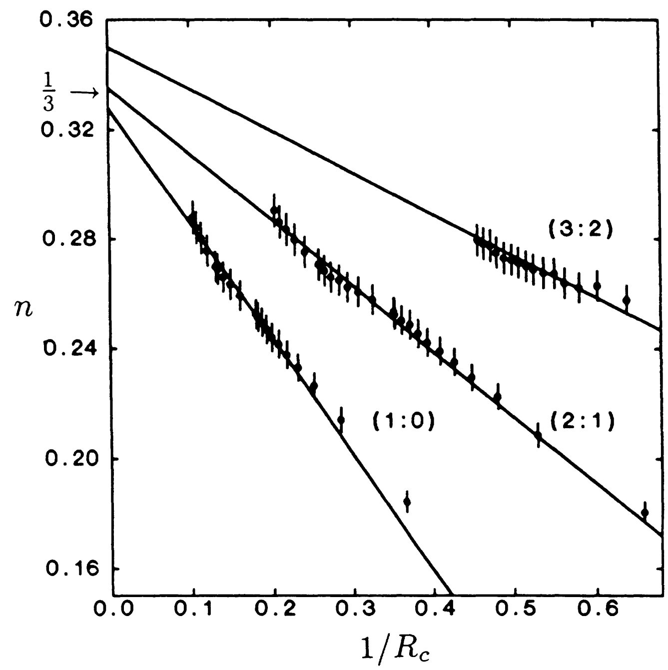

FIG. 3. Results of matching to estimate n with R_c. Lines are identified by level of matching $(m:m')$, which are the numbers of rescalings on the large and small lattice, respectively. Every fifth datum point is shown to 2500 Monte Carlo steps, then every twentieth to 9500 time steps, and every seventieth thereafter. Fits are obtained from the full data set. Error bars are 3 standard deviations.

significantly smoother and are fitted by $n=\frac{1}{3}$ better than $n=\frac{1}{4}$. The implication is that there is strong short-range transient behavior to which R_e is sensitive. Note that the transient is iterated away by the successive application of the renormalization-group transformation. Indeed, short-range diffusion gives $n=\frac{1}{4}$,[18] although the long-range Lifshitz-Slyozov mechanism gives $n=\frac{1}{3}$. It should be noted that the appearance of a strong $\frac{1}{4}$ transient was also seen in the work on Langevin models[13]; this similarity indicates that the kinetic Ising model and the Langevin model of spinodal decomposition may share a universality class.

Our simply fitting each curve does not necessarily determine the nature of scaling near the fixed point. To find n in the scaling regime we do a matching analysis as described above. We estimate an effective n by determining the ratio of times on the two lattices which gives equal domain sizes. Using this $n(R)$, we estimate the asymptotic exponent by generalizing Huse's formula, i.e., by $n(R\rightarrow\infty)=n(R)+C/R$. The data for R_c are shown in Fig. 3. The estimates for n from the first three levels of matching are 0.328, 0.335, and 0.349. Somewhat surprisingly, R_c is such a good measure of length that all the extrapolated n's are equal within our statistical error. Averaging them gives our estimate for the growth exponent: $n=0.338\pm0.008$, which is in good agreement with the classical result 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 become smaller, as we would expect. The results for R_e are more sensitive to short-ranged effects (the renormalization group has to "work harder" to both iterate away the transient $n=\frac{1}{4}$ behavior and cause the amplitudes to converge): While the results at the third level of matching are consistent with those above, we cannot improve that estimate for n. We have also assumed $n=\frac{1}{3}$, and found that the R's at different levels of renormalization approach each other as the renormalizations are done.[19]

To conclude, the results of our Wilson-type Monte Carlo renormalizaton-group study indicate that the $d=2$ kinetic Ising model with a conserved order parameter obeys an asymptotic power law with growth exponent close to $\frac{1}{3}$ in the scaling regime. We see no evidence for logarithmic growth. Furthermore, while long transients involving an effective $n\approx\frac{1}{4}$ were seen in the R_e data, these were iterated away by the renormalization-group transformation leaving $n\approx\frac{1}{3}$. While more study is needed—for example, we had to make use of Gibbs-Thomson-type relations to obtain the asymptotic growth exponent—it is clear that these results are in agreement with the classical theory of Lifshitz and Slyozov, as well as the recent Monte Carlo and Langevin-model work.[8,11-13] Although this does not demonstrate that the Langevin model and the kinetic Ising model share the same universality class, it strongly indicates that this may be the case.[20] In the future, we shall present our results for the correlation function and studies done at other temperatures, as well as a more detailed account of the work reported here.[19]

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[20]The renormalization group of Mazenko and co-workers looked at low temperatures and short times with a rather different approach. It may be, as Huse (Ref. 8) has suggested, that they never enter the scaling regime; i.e., their data could be affected by microscopic activated processes which limit the growth at early times. See also the discussion of low temperatures in Ref. 15. We have seen evidence of such processes at low temperatures (Ref. 19). Also, it may be worth noting that in the nonconserved Potts model on a square lattice, there exists a complicated $T=0$ fixed-point structure. [See J. Viñals and J. D. Gunton, Phys. Rev. B **33**, 7795 (1986); J. Viñals and M. Grant, Phys. Rev. B **36**, 7036 (1987).] For that system, a freezing fixed point at $T=0$ caused long "logarithmlike" transients at $T>0$. In the future, we intend to investigate further the nature of the zero-temperature fixed point for spinodal decomposition.