Spinodal Decomposition for Langevin Equations

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(Received 2 March 1987)

The time-dependent Ginzburg-Landau model for spinodal decomposition is studied by use of both a numerical simulation of the associated Langevin equation and a new low-temperature expansion. Both methods lead to a growth law for the characteristic domain size $L(t) \sim t^{1/4}$.

PACS numbers: 64.60.Cn, 64.60.My

Considerable insight into the growth kinetics of systems subjected to temperature quenches to unstable regions of the phase diagram has been gained through the study of time-dependent Ginzburg-Landau (TDGL) models. We study here the Langevin equation associated with a TDGL model for spinodal decomposition with a conserved, scalar order parameter.[1] We report results obtained using two different approaches. In the first, we have carried out direct numerical simulations of the Langevin equation, and we have analyzed the results using our recently developed[2,3] renormalization-group methods for nonequilibrium phenomena. For the first time, data of sufficiently good quality have been obtained to make it possible to estimate the growth law for this model. In the second approach we use a novel analytic low-temperature expansion technique which is applicable to the same model. The leading term in this expansion corresponds to a quench to very low temperatures. The problem then reduces to an integro-differential equation which can, at worst, be solved very accurately numerically. In the analytic approach, quantities of interest can be calculated at arbitrarily long times. Both approaches lead to the same result for the growth law of the characteristic domain size, $L(t) \sim t^n$, with $n = \frac{1}{4}$.

Our results have implications concerning universality classes for growth kinetics problems. It has often been assumed that the model studied here would fall in the same "universality class" as the spin-exchange kinetic Ising model. It appears that this assumption may not be justified, and that the discrete nature of a conserved[4] order-parameter variable may be relevant to the determination of the appropriate growth kinetics class. While we find here an exponent of $\frac{1}{4}$ for a very wide range of quenching parameters, previous work on the spin-exchange kinetic Ising model indicates freezing behavior[2] for quenches to zero temperature and an exponent[5-7] $n = \frac{1}{3}$ (the Lifshitz-Slyozov[8] result) for quenches to a final quenching temperature $T_F > 0$. Further investigations of this significance of these differences is under investigation.

The dynamics[9] of a conserved scalar order parameter $\psi_i(t)$, defined at site i and time t, are governed by the Langevin equation:

$$\partial \psi_i / \partial t = \Gamma \nabla^2 \delta F[\psi] / \delta \psi_i + \eta_i(t), \tag{1}$$

where Γ is a kinetic coefficient and $\eta_i(t)$ is a Gaussian noise field which satisfies

$$\langle \eta_i(t) \eta_j(t') \rangle = -k_B T \Gamma \nabla^2 \delta_{ij} \delta(t - t'), \tag{2}$$

and the total order-parameter conservation law is ensured by the discrete Laplacians. The free energy, F, governing our model is given by

$$F[\psi] = \tfrac{1}{2} J \textstyle\sum_i [r\psi_i^2 + (\nabla \psi_i)^2 + \tfrac{1}{2} u \psi_i^4] \tag{3}$$

and we shall use the parametrization introduced by Beale, Sarker, and Krumhansl,[10] where $r = -\theta$, $u = 1 + \theta$, and $K = J/k_B T$. As $\theta \to \infty$, the variables ψ_i become Ising type. The numerical calculations described below have been performed on a square lattice. We refer the reader to previous work[11] with a nonconserved order parameter for a detailed discussion of the techniques employed to solve equations of this type. To average over the noise the equation is solved a large number of times. We speak of each solution as a "run."

In the case of a conserved order parameter, particular attention must be paid to the analysis of the results, and to the choice of correlation functions to be collected and analyzed. The quasistatic structure factor, $C(\mathbf{q},t)$, is often chosen. Although we have studied this quantity (see below, and also Ref. 9), one knows[7,12] that it is extremely difficult to extract a growth law from $C(\mathbf{q},t)$ data obtained over limited time ranges for a finite system. We have, therefore, adopted the much more efficient procedure of performing a renormalization-group analysis on our data, using the methods of Refs. 2 and 3, for the block correlation functions introduced there. We sketch below the salient points of the pro-

cedure. We consider blocks of size $M\times M$ (where $M<N$, the size of our system) and define the block "magnetization" m_M as the sum of the variables ψ_i within the block. The theory is formulated in terms of the quantities,

$$R_M(t)=\langle m_M^2\rangle_t/[M^2S(t)], \tag{4}$$

where the $\langle\ \rangle_t$ represent the time-dependent averages over noise and initial conditions, and $S(t)=N^{-1}\sum_i\langle\psi_i^2\rangle_t$ is the "local" order parameter. We start our quenches, for convenience, from the initial condition $\{\psi_i\equiv 0\}$. In general, $R_M(t)$ will depend on the location of the final state in the ordered region of the phase diagram[10] as parametrized by K and θ. In the region of parameter space we have explored [twenty (K,θ) pairs including five values of θ ranging from 1.5 to 12, and ten values of K ranging from 0.8 to 14] this dependence appears not to affect the growth law, and for brevity and simplicity it will be omitted here.[9] Central to our renormalization-group analysis, as shown in Ref. 3, is the quantity

$$D=-\frac{[\partial\ln R_M(t)/\partial\ln M]_t}{[\partial\ln R_M(t)/\partial\ln t]_M}, \tag{5}$$

which, in the scaling regime, is independent of M. Furthermore, given a spatial rescaling factor b, there is a corresponding rescaling factor for the time which satisfies the partial differential equation

$$\left[b\frac{\partial}{\partial b}+Dt\frac{\partial}{\partial t}+D\right]\Delta=0;\quad \Delta(b=1,t)=1. \tag{6}$$

In particular, if D is independent of t, $D\equiv D_0$, (6) has the solution $\Delta=b^{-(1/D_0)}$, which implies[2,3] a growth law $L(t)=L_0t^{1/D_0}$, and scaling under simultaneous rescaling of space and time.

Our calculations for the functions $R_M(t)$ and D have been for a 32×32 system, with checks at larger and smaller sizes to verify that our results are free of finite size effects for the range of M ($<\frac{1}{2}N$) of interest. We have chosen our units of time t by taking $\Gamma k_BT=1$. To plot data obtained for different values on K on the same time scale it is convenient, since $K\sim T^{-1}$, to use the variable $\tau\equiv tK$. We have data for times up to $\tau_M\approx 10$–20 in all cases and with spot checks up to $\tau_m=180$. The connection between Langevin and Monte Carlo times for this model was discussed by Meakin *et al.*[13] The results there show that 2200 Monte Carlo steps correspond to $\tau=0.4\times 7=2.8$. To compare Ising dynamics is much more difficult. One can form an estimate by comparing the degree of order reached by the system at time τ, as measured by moments of $C(\mathbf{q},t)$ or the absolute values of $R_M(t)$, with corresponding Ising result. In this way we find corresponding Ising results. Estimates obtained in this way depend on the quantity chosen but $\tau_m=20$ corresponds to at the very least 10^3 Monte Carlo steps[14] and 10^4 would be a reasonable guess. Thus we have results for rather long times. All of our results are averages over 100 runs. A representative sample of our results for $R_M(t)$ is given in Fig. 1. We find that for a wide range of M values the product R_MM^3 is independent of M. This holds also[3] in the Ising case and it is a consequence of the conservation law. Thus the numerator in (5) equals 3. The denominator appears to be independent of M in the same range. We are, therefore, in the scaling regime (D independent of M). To study the remaining dependence of D on time and other parameters we have considered the value of D averaged over the weak M fluctuations as a function of time, for all (K,θ) pairs. A portion of these data is shown in Fig. 2. Within the statistical fluctuations, which are appreciable, the results appear to be independent of time. Furthermore, they depend only weakly on the values of (K,θ) as stated above. We are led, therefore, to the result $D_0=4$, which implies $L(t)\sim t^{1/4}$.

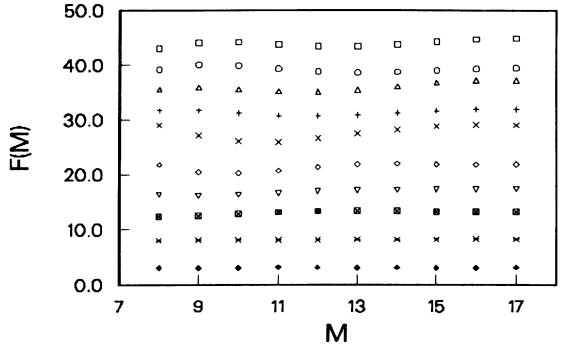

FIG. 1. The quantity $F(M)\equiv R_M(t)M^3$ plotted as a function of M for several values of θ, K, and the time parameter τ. From top to bottom the ten values of (θ,K,τ) are (2.5, 1.75, 14.0), (1.5, 2.5, 13.75), (2.5, 1.75, 10.5), (1.5, 2.5, 10.0), (2.5, 2.5, 7.5), (5.0, 1.75, 5.25), (12.0, 0.8, 3.4), (12.0, 1.6, 3.2), (12.0, 0.8, 1.2), and (12.0, 3.2, 0.6).

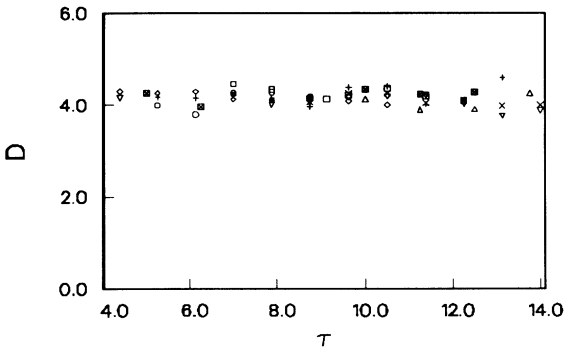

FIG. 2. The quantity D, defined in Eq. (5), as a function of τ, for several values of (θ,K) as follows: open squares, (2.5, 14); circles, (5, 1.75); triangles, (2.5, 2.5); plus signs, (2.5, 1.75); crosses, (2.5, 7); lozenges, (3.5, 14); inverted triangles, (2.5, 3.5); and filled squares, (1.5, 2.5).

We have endeavored to check this against direct simulations. In Fig. 3 we plot results for $q_1(t)$, the first moment of $C(q,t)$, in a more extended time region ($\tau_M = 180$). The linear best fit to this log-log plot gives an exponent of $\simeq 0.27$, which is quite compatible with $\frac{1}{4}$. Similar results[9] are obtained for other (K,θ) pairs. These direct results, while supportive of the evidence for a $\frac{1}{4}$ exponent, would not in themselves be conclusive since good fits to the form $(t-t_0)^{-n}$ with t_0 small and $n \neq \frac{1}{4}$ can also be obtained.

We turn now to a description of the low-temperature expansion. A thorough discussion is technically involved and will be given elsewhere.[9] We outline the main points here. For the N-vector model in the large-N limit, the first systematic treatment of a TDGL model describing a quench from a stable to an unstable region was given by Mazenko and Zannetti (MZ).[15] The equation governing the structure factor in Ref. 15 is formally identical to that introduced by Langer, Bar-on, and Miller[1] for a scalar order parameter. The solution in MZ properly generates, for $N > 1$, massless Nambu-Goldstone (NG) modes $[C(\mathbf{q},t \to \infty) \sim 1/q^2]$ whose growth one can follow. As pointed out by Billotet and Binder,[16] a problem with the Langer, Bar-on, and Miller approximation, for $N = 1$, is that one still obtains NG modes in the final equilibrium structure factor; this should be replaced by a standard Ornstein-Zernlike form, $(q^2+\xi^{-2})^{-1}$, for a low final temperature. The difficulties become apparent when one attempts to set up a direct low-temperature expansion. This appears at first rather straightforward: After rescaling the fields $\psi \to K^{-1/2}\psi$, one realizes that all residual temperature dependence (except that associated with the initial conditions) can be absorbed into an effective quartic coupling $u' = u/K$. Thus a low-temperature expansion (large K) is an expansion in powers of u'. But things are not quite so simple for quenches into an unstable region since the system will try to grow a Bragg peak. The structure factor will eventually have a peak which has weight $m^2 = -r/u'$ which, for small u', is large. Thus one must treat the peak contribution to the structure factor as $O(1/u')$. This is reminiscent of equilibrium expansions in the ordered state where one must treat the average magnetization as $O(1/\sqrt{u})$ and $m^2 = O(1/u)$. It is not difficult to structure the calculation so that one can take this complication into account. The $O(1)$ calculation is then formally identical to Eq. (2.31) for $C(\mathbf{q},t)$ in MZ, except that one replaces $u \to 3u$ in Eq. (2.28) (MZ). This theory, when applied for $N = 1$, has two defects which are associated with the improper final state reached by the structure factor. The first defect, the existence of spurious NG modes, is discussed above: The second is that it gives the incorrect value for the final spontaneous magnetization $(-r/3u)^{1/2}$ rather than the correct value $(-r/u)^{1/2}$. These defects indicate an improper expansion in this case. One of the problems[9] is that the field contains a portion associated only with local ordering. We write $\psi_i(t) = \phi_i^0(t) + \phi_i(t)$, where $\phi_i^0(t)$ describes the local ordering (domains) and $\phi_i(t)$ describes fluctuations about the local ordering. For low temperatures and long times $\phi_i(t)$ can be treated as small compared with $\phi_i^0(t)$. The field $\phi_i^0(t)$ is written in the form $\phi_i^0(t) = [S_0(t)]^{1/2}\sigma_i(t)$, where $\sigma_i^2(t) = 1$ and $S_0(t)$ is to be determined. The difficult part of the analysis is to define $\phi_i^0(t)$ and $\phi_i(t)$ such that $\phi_i^0(t)$ gives the Bragg-peak component of the structure factor, without the offending q^{-2} tail, and such that $\phi_i(t)$ represents the [small $= O(k_B T)$] equilibrating thermal fluctuation contribution to the structure factor. The key[9] is to introduce a fictitious probability distribution governing the $\sigma_i(t)$ which facilitates the decoupling of ϕ^0 and ϕ. The strength of the ordering component, $S_0(t)$, is determined self-consistently by the requirement of $S_0(t) = \langle [\phi_i^0(t)]^2 \rangle$ evaluated at lowest order in $k_B T$. So $S_0(t)$ is the weight under the Bragg peak at time t. The resulting lowest-order equation for the peak contribution, $C_p(\mathbf{q},t) = \langle |\phi_0(\mathbf{q},t)|^2 \rangle$, is identical to that found by MZ [Eq. (2.31) of Ref. 15] with one important exception: The NG modes are subtracted off so $C_p(\mathbf{q},t) = D(\mathbf{q},t)$, which is given by MZ [Eq. (3.10) of Ref. 15]. The numerical solution for $C_p(\mathbf{q},t)$ is, therefore, the same as in MZ. For our purposes here the important result is that in the low-temperature limit $n = \frac{1}{4}$ (for both $d = 2$ and $d = 3$). This is the same result that we have obtained using the numerical method.

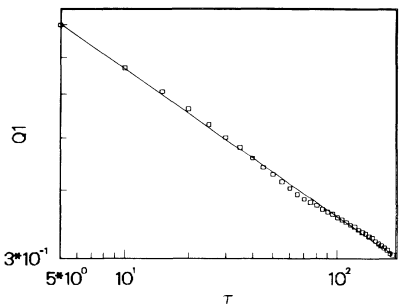

FIG. 3. The first moment of the structure factor plotted vs τ for $(\theta = 1.5, K = 10)$. The squares are the numerical results and the straight line the best power-law fit which corresponds to an exponent of 0.27.

This work has been supported in part by National Science Foundation Grant No. DMR-84-12901 at The University of Chicago, and by the Microelectronics and Information Sciences Center at the University of Minnesota. The numerical calculations were made possible by a grant of computer time from the University of Minnesota Supercomputer Institute.

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