Domain growth and scaling in the two-dimensional Langevin model

E. T. Gawlinski
Department of Physics and Center for Advanced Computational Science, Temple University, Philadelphia, Pennsylvania 19122

Jorge Viñals
Department of Physics, Carnegie Mellon University, Pittsburgh, Pennsylvania 15213

J. D. Gunton
Department of Physics, Lehigh University, Bethlehem, Pennsylvania 18015
(Received 22 December 1988)

A numerical simulation of the Langevin model for spinodal decomposition is carried out in two dimensions. The pair correlation function is found to exhibit dynamical scaling to a good first approximation. It is found that the domain growth law asymptotically approaches $t^{1/3}$, in agreement with previous results for the kinetic Ising model with conserved order parameter. The Langevin, cell dynamics, and spin exchange kinetic Ising models are shown to belong to the same dynamical universality class for the range of domain sizes studied here.

Numerical simulation methods have been used for some time to study the kinetics of first-order phase transitions.[1] These include Monte Carlo and molecular dynamics simulations and numerical integration of stochastic differential equations (such as the Langevin equation). It has been recognized for many years that such studies are useful in obtaining qualitative insights (e.g., the role that conservation laws play in ordering processes), but is has proved difficult to determine precisely the asymptotic universal properties such as domain growth laws and scaling functions. The main limitations are due both to finite-size effects and related finite-time limitations imposed by current computer capabilities.[2] The lack, in most models, of any theoretical predictions makes such a determination even more difficult. Recent theoretical developments, however, have led to renewed interest in systems which undergo spinodal decomposition and coarsening, such as occurs following a quench below a critical point at a critical value of the order parameter. On the one hand, it has been argued[3,4] that for such a quench the asymptotic growth is described by a modified Lifshitz-Slyozov law in which the characteristic length $R(t)$, which is related to the average size of ordered domains, behaves like $R(t) \approx t^x$, with $x = \frac{1}{3}$, and including an important correction term arising from surface diffusion along the interfaces of the interconnected structures. On the other hand, numerical simulations together with renormalization-group ideas have been used to study both the spin exchange kinetic Ising (SEKI) model with a conserved order parameter[5,6] and the continuum Langevin model[7] in two dimensions. The conclusions of that work were that the two models belong to different dynamical universality classes. The growth law obtained for the Langevin model is a power law with an exponent $x = \frac{1}{4}$.[7,8]

Recent extensive Monte Carlo studies[9,10] of the SEKI model show evidence for a power law with a growth exponent close to $x = \frac{1}{3}$, once the correction term for diffusion along the interfaces has been taken into account. Therefore, it is of interest to perform similar extensive studies of the Langevin model in order to clarify two major issues: (i) whether the model exhibits dynamical scaling, and (ii) whether the continuum Langevin model, the SEKI model, and perhaps other models of first-order phase transitions such as the Oono-Puri cell-dynamics model,[11] belong to the same dynamical universality class.

In this Brief Report we present the results of a comprehensive numerical simulation of spinodal decomposition and coarsening for the two-dimensional Langevin model. As discussed below, we calculate the pair-correlation function and show that to a good first approximation dynamical scaling is satisfied. We also find that our data for the characteristic length scale is consistent with the modified Lifshitz-Slyozov law. Furthermore, the scaling function that we obtain for the Langevin model is almost identical to the scaling functions for the SEKI (Ref. 10) and the Oono-Puri cell dynamics[11] models, both at a critical value of the order parameter. The three scaling functions differ by, at most, 2% in the range of domain sizes studied (see Fig. 1 inset). Thus, within this uncertainty, these three models appear to be in the same dynamical universality class. The same scaling function for the Langevin model has been obtained in an independent work by Rogers, Elder, and Desai.[12] Our largest simulation time exceeds theirs by nearly an order of magnitude. However, in the range of overlap the scaling functions are found to agree. We have also compared our scaled structure factor with that of Amar, Sullivan, and Mountain[9] and find very good agreement.[13]

The Langevin equation for the case of a conserved order parameter $c(\mathbf{x},t)$ is

$$\partial_t c(\mathbf{x},t) = M\nabla_{\mathbf{x}}^2[(-K'\nabla_{\mathbf{x}}^2 - a)c(\mathbf{x},t) + uc^3(\mathbf{x},t)] + \xi(\mathbf{x},t)\,, \tag{1}$$

where M is a kinetic coefficient, K', a, and u are positive,

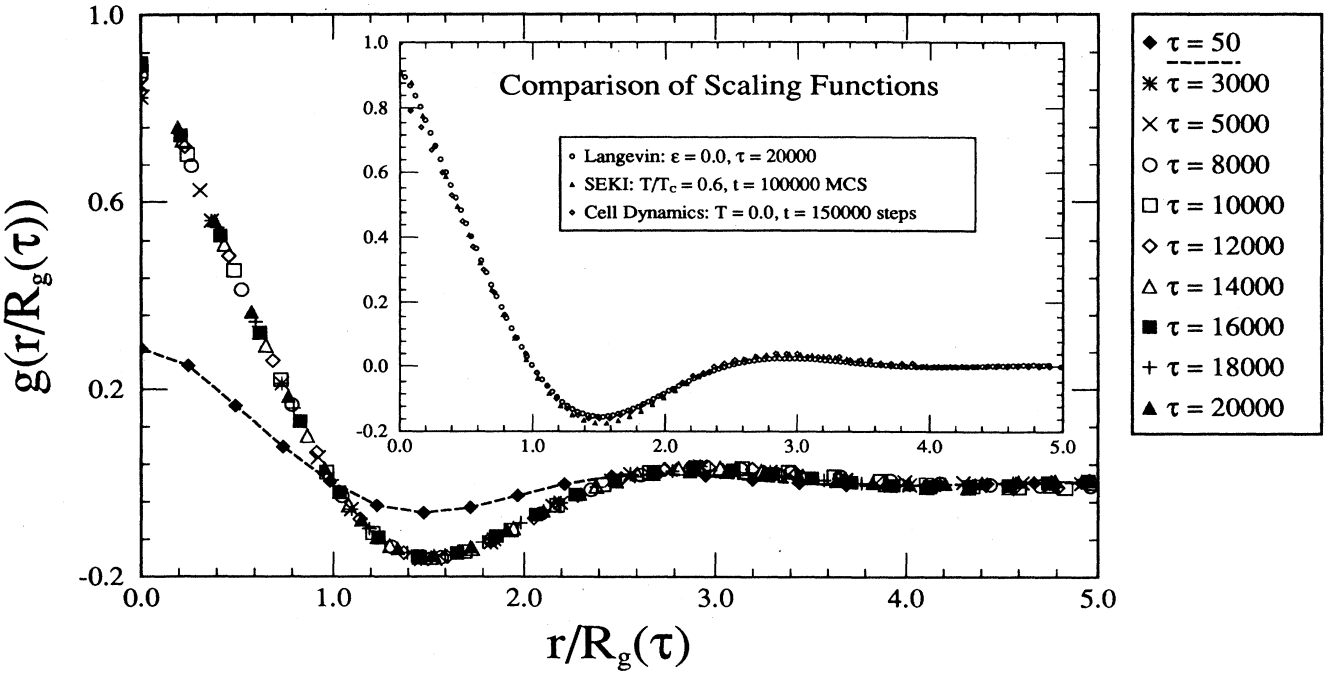

FIG. 1. The pair-correlation function $g(r,\tau)$ plotted with respect to $r/R_g(\tau)$, to test the scaling ansatz. The dimensionless times τ are indicated in the legend. Note the early time ($\tau=50$) breakdown of scaling. We find that only when $\tau \geq 1000$ are we safely in the scaling regime. Note also that $g(r,\tau)$ is close to but not exactly one for $r/R_g(\tau) \rightarrow 0$, even at the latest times studied. The inset shows a comparison of our results with those found for the SEKI and the cell dynamics models of Refs. 10 and 17, respectively.

phenomenological coefficients, and the noise $\xi(\mathbf{x},t)$ is assumed to be Gaussian distributed, with zero mean and $\langle \xi(\mathbf{x},t)\xi(\mathbf{x}',t')\rangle = -2k_BT\nabla_x^2\delta(\mathbf{x}-\mathbf{x}')\delta(t-t')$. The first term on the right-hand side of Eq. (1) is $M\nabla_x^2(\delta F/\delta c)$, where $F(c)$ has been taken to be the usual Ginzburg-Landau free-energy functional with two independent parameters. We have chosen[14] to introduce dimensionless variables as follows: $\psi=\sqrt{u/a}\,(c-c_0)$, $\mathbf{r}=\sqrt{a/K'}\,\mathbf{x}$, and $\tau=(2Ma^2/K')t$, where c_0 is the initial average value of the order parameter and $\sqrt{a/u}$ and $\sqrt{K'/a}$ are proportional to the mean-field values of the order parameter and correlation length, respectively. Equation (1) then becomes

$$\partial_\tau\psi(\mathbf{r},\tau) = \tfrac{1}{2}\nabla_r^2[(-\nabla_r^2-1)\psi(\mathbf{r},\tau)+\psi^3(\mathbf{r},\tau)] + \sqrt{\epsilon\eta}(\mathbf{r},\tau)\,, \tag{2}$$

where we have set the constant $c_0=0$ corresponding to the critical quench studied in this paper. The scaled noise $\eta(\mathbf{r},\tau)$ satisfies a scaled version of the fluctuation-dissipation relation, with the intensity given by $\epsilon=k_BTu/aK'$. The parameter ϵ is the quantity normally used in the Ginzburg criterion to determine the validity of mean-field theory in critical phenomena. Note that with this particular scaling, ϵ contains the only dependence on the original parameters of the Ginzburg-Landau free energy. This parametrization is useful for small ϵ, i.e., for quenches well below the critical temperature T_c. On the other hand, the parametrization which was used in Ref. 7 to also study the Langevin model is related to ours in the following way: $\Theta=a/K'$ and $K=K'^2(1+\Theta)/u$. Dynamical properties such as the growth law of both the conserved and nonconserved Langevin models were studied for different pairs of values of K and Θ; the results obtained there suggested that the domain growth law is independent of K and Θ (except for the special case $\Theta\rightarrow 0$ when the double-well potential disappears). If the noise term in Eq. (2) is irrelevant to the universal growth law (and possibly to the scaling function) for this model (and correspondingly for the nonconserved model), then our parametrization would appear to be the natural one to use in order to determine these properties, at least at low temperatures. The universal features of the domain growth law, correlation function, and structure factor would then be obtained for different values of K and Θ by the simple rescaling introduced above. When the noise term is important, such as in the early time behavior, one might expect nonuniversal behavior for different K since $\epsilon=(\Theta+1)/K\Theta$. This picture is qualitatively supported by the work described in Ref. 12. In fact, larger values of ϵ result in a more extended initial transient regime and require longer simulation times in order to reach the scaling regime. One should also point out that the largest deviation between the scaling functions of the Langevin model and the SEKI model appear at short distances. This is due to the fact that the average value of the local order parameter $\langle\psi^2(\tau)\rangle$ is identically equal to one in the case of the SEKI model but is a function of time in our present case. Its value approaches a constant when the scaling regime is reached but the time required to reach saturation depends

on ϵ (see Fig. 1 inset for comparison). The same transient behavior has also been discussed in Ref. 7.

In our numerical solution we have discretized Eq. (2). For the bulk of our work we have examined a square lattice of 256×256 points at $\epsilon=0.0$, with the order parameter $\psi(\mathbf{r}_i)$ given at each of the lattice points. We have also examined a 128×128 system at $\epsilon=0.1$ and out to a maximum time of $\tau=1000$. We have since established that $\tau=1000$ is not a sufficiently long time to be convincingly into the scaling regime. Therefore, for the case of $\epsilon=0.1$, we will present an expanded result of a much longer simulation in a later paper.

We have replaced the Laplacian in Eq. (2) by its discretized form $\nabla_{\mathbf{r}}^2\psi(\mathbf{r},\tau)=\sum_{\mathbf{m}}[\psi(\mathbf{r}_i+\mathbf{m})-\psi(\mathbf{r}_i)]/(\Delta r)^2$ where $\mathbf{m}$ is the nearest-neighbor lattice vector. We then numerically integrate Eq. (2) using Euler's method, with dimensionless mesh size $\Delta r=1.0$ and a discrete dimensionless time step $\Delta\tau=0.02$ for $\epsilon=0.0$. Our initial distribution for $\psi(\mathbf{r}_i,\tau=0)$ was specified by a random, uniform distribution in the range $[-1/\sqrt{3},+1/\sqrt{3}]$. To average the noise[15] and the initial distribution of $\psi(\mathbf{r}_i)$, we solved Eq. (2) a large number of times (runs), namely for $\epsilon=0.0$, 20 runs.

We have paid particular attention (for reasons that will become clearer below) to an accurate determination of the (nonequilibrium) pair correlation function $\hat{g}(\mathbf{r},\tau)=\langle\delta\psi(\mathbf{r},\tau)\delta\psi(\mathbf{0},\tau)\rangle$. We have performed a circular average on $\hat{g}(\mathbf{r},\tau)$ to calculate $g(r,\tau)$ and the resulting quantity has been averaged over the 20 independent runs at $\epsilon=0.0$. [We have found this number of runs sufficient for an accurate determination of $g(r,\tau)$.[16]]

Our first length $R_g(\tau)$ is defined[3] as the smallest value of r for which $g(r,\tau)=0.0$ at time τ. We have also used the conventional procedure of defining lengths from moments of the structure factor. The behavior of the different lengths is quite similar, although we have found that the lengths defined from moments of the structure factor are sensitive to the upper cutoff in k space.

First we address the validity and extent of the dynamical scaling ansatz, which states that the pair-correlation function satisfies a scaling relation $g(r,\tau)=F[r/R(\tau)]$ for $\tau>\tau_0$ (τ_0 is some initial transient time). In Fig. 1 we show the results obtained from plotting our data for $g(r,\tau)$ vs $r/R_g(\tau)$ for τ ranging from 3000 to 20000. It seems quite clear that for $\tau>3000$ we are fairly well into the scaling regime. We also show in Fig. 1 the results for $\tau=50$ (dashed line); this gives an idea of the maximum departure from scaling observed at very early times. For $\tau\leq1000$ the departure from scaling increased continuously with decreasing τ up to the degree seen for $\tau=50$. In the inset in Fig. (1) we show our results for the scaling function for $\epsilon=0.0$ plotted with those of the SEKI (Ref. 10) and the cell-dynamics[17] models. One can see that over the range of lengths studied the agreement is very good.

Finally, we consider the issue of the domain growth law for this system. As noted earlier, it has been argued,[3] that the growth law for the kinetic Ising model satisfies $R(\tau)=a+b\tau^{1/3}$, where the constant a arises from the surface diffusion.[18] The same argument would appear to hold for the Langevin model as well. On the other hand, it has been also argued[5] that the asymptotic behavior in the Langevin model is given by a different power law, i.e., $x=\frac{1}{4}$.[7] It is of course difficult to distinguish between two small exponents, such as $\frac{1}{3}$ and $\frac{1}{4}$. Nevertheless, we believe our results strongly suggest that $x=\frac{1}{3}$. For example, we have analyzed our data in terms of a nonlinear fit to $R(\tau)=a+b\tau^x$. We have found that the best fit corresponds to $x=0.38\pm0.04$ for $\epsilon=0.0$, so that our data are in reasonable agreement with the prediction of Ref. 3. It should be mentioned that these nonlinear fits are over the entire data range beginning at the earliest times and hence our value of x obtained in this way is probably affected by regions of the data that are not characterized by a pure power law. If we restrict the fitting procedure to $\tau>4000$ we obtain a value of $x=0.36\pm0.03$ in better agreement with the predicted value of $x=\frac{1}{3}$. The error bars are a crude estimate based upon the curvature of $\chi^2(a,b,x)$ near its minimum.

As a second illustration, we show in Fig. 2 the results of logarithmically plotting our data for $\epsilon=0.0$. The dashed line has a slope of $\frac{1}{3}$. One can see that the data is in agreement with an asymptotic approach of $x\rightarrow\frac{1}{3}$.

Finally, in Fig. 3 we show the effective exponent $N_{\text{eff}}=d\log_{10}[R_g(\tau)]/d\log_{10}(\tau)$ plotted as a function of $1/R_g$ for the $\epsilon=0.0$ data. Assuming the Huse[3] prediction, this effective exponent should approach $\frac{1}{3}$ for $R_g\rightarrow\infty$. Our results clearly support Huse's prediction. The large fluctuations for large R_g are due to the numerical differencing done when computing the logarithmic derivative. (See Fig. 1 caption.)

These results are to be contrasted with those of Ref. 7, which also involves a numerical simulation of this model but where a growth exponent of $x=\frac{1}{4}$ is obtained. We find that the relationship between our time scale τ and the time scale in that reference τ_{MV}, is $\tau=2\Theta^2\tau_{\text{MV}}$.[19] The renormalization-group analysis presented therein (see Fig. 2 in Ref. 7 and also Fig. 6 in Ref. 6) is based on times $\tau\leq300$ for $\epsilon=0.5$ and $\tau\leq500$ for $\epsilon=0.7$. The longest simulation time, however, appears to be $\tau\approx800$ with $\epsilon=0.17$ (Fig. 3, Ref. 7). We conclude that most of the

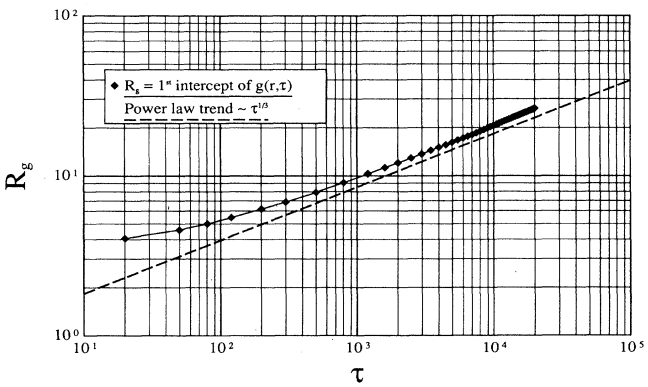

FIG. 2. The first intercept of the pair-correlation function, R_g, is shown on a log-log axis plotted vs the dimensionless time τ. The dashed line is a power law with exponent identically equal to $\frac{1}{3}$. Note the very slow asymptotic approach to this line demonstrated by the raw data.

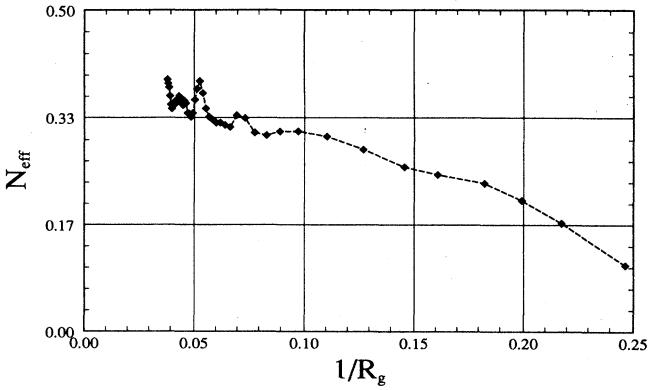

FIG. 3. The effective exponent, $N_{\rm eff}=d\log_{10}[R_g(\tau)]/d\log_{10}(\tau)$, is plotted vs $1/R_g$. The extrapolated value of $N_{\rm eff}$ as $1/R_g\rightarrow 0$ should give the asymptotic late time effective power law behavior. For $1/R_g\geq 0.08$ the data is smooth and continuously approaches $\frac{1}{3}$ from below. For smaller values of $1/R_g$ the data become rather noisy due to the numerical round-off error incurred while taking the logarithmic derivative (which is proportional to the ratio of small differences between large, nearly equal, numbers).

data presented in Refs. 6 and 7, and especially the data used to determine the exponent x, are probably effected by transient behavior prior to the asymptotic scaling regime.

In conclusion, our results for the domain growth are consistent with a modified Lifshitz-Slyozov law and the scaling function which we have presented appears to coincide with the recently obtained scaling function for both the cell dynamics (at $T=0.0$) and SEKI (at $T=0.6T_c$) models, both for quenches at a critical value of the order parameter. As a consequence, and within the precision of our study, all three models appear to belong to the same dynamical universality class.

This work was supported by the National Science Foundation (NSF) Grant No. DMR-8612609 and by the computing facilities at the Center for Advanced Computational Science at Temple University and those at the Pittsburgh Supercomputer Center. One of us, J.V., was supported by NSF Grant No. DMR-8409397. We are also grateful to Dr. A. Chakrabarti, Dr. J. Collins, Professor M. Grant, and Dr. R. Toral for helpful discussions.

[1]Reviews of the field are given in J. D. Gunton, M. San Miguel, and P. S. Sahni, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and J. L. Lebowitz (Academic, New York, 1983), Vol. 8; K. Binder, in *Condensed Matter Research Using Neutrons,* edited by S. W. Lovesy and R. Schem (Plenum, New York, 1984); J. D. Gunton, in *Proceedings of NATO ASI on Time Dependent Effects in Disordered Materials, Geilo, Norway, 1987,* edited by R. Pynn and T. Riste, NATO Advanced Study Institute, Series B Physics (Plenum, New York, 1987), Vol. 167.

[2]See, for example, O. Penrose, J. L. Lebowitz, J. Marro, M. H. Kalos, and A. Sur, J. Stat. Phys. **19**, 243 (1978); O. Penrose, J. L. Lebowitz, J. Marro, M. H. Kalos, and J. Tobochnik, J. Stat. Phys. **34**, 399 (1984); P. Fratzl, J. L. Lebowitz, J. Marro, and M. H. Kalos, Acta Metall. **31**, 1849 (1983), and references therein; K. Binder and D. W. Heermann, in *Scaling Phenomena in Disordered Systems,* edited by R. Pynn and A. Skjettorp (Plenum, New York, 1985); A. Milchev, K. Binder, and D. W. Heermann, Z. Phys. B **63**, 521 (1986).

[3]D. Huse, Phys. Rev. B **34**, 7845 (1986).

[4]H. Furakawa, Adv. Phys. **34**, 703 (1985).

[5]G. F. Mazenko, O. T. Valls, and F. C. Zhang, Phys. Rev. B **32**, 5807 (1985).

[6]Z. W. Lai, G. F. Mazenko, and O. T. Valls, Phys. Rev. B **37**, 9481 (1988).

[7]G. F. Mazenko and O. T. Valls, Phys. Rev. Lett. **59**, 680 (1987).

[8]R. Petschek and H. Metiu, J. Chem. Phys. **79**, 3443 (1983).

[9]J. G. Amar, F. E. Sullivan, and R. D. Mountain, Phys. Rev. B **37**, 196 (1988).

[10]K. Kaski and J. D. Gunton (unpublished); J. D. Gunton, E. T. Gawlinski, and K. Kaski, in *Dynamics of Ordering Processes at a Phase Transition,* edited by S. Komura (Plenum, New York, in press).

[11]Y. Oono and S. Puri, Phys. Rev. Lett. **58**, 836 (1987).

[12]T. M. Rogers, K. R. Elder, and R. C. Desai, Phys. Rev. B **37**, 9638 (1988).

[13]To compare these data we multiply both $R_g^2S(kR_g)$ and kR_g by constants that bring the peaks of the structure factors into coincidence. After doing so we find that the rest of the data agree within statistical fluctuations.

[14]M. Grant, M. San Miguel, J. Viñals, and J. D. Gunton, Phys. Rev. B **31**, 3027 (1985).

[15]We have calculated the noise term in the same manner as in Ref. 8.

[16]Previous "long-time" calculations (Refs. 3, 9, and 10) of $g(r,\tau)$ for the kinetic Ising model have been based on much less information (except for the work described in Ref. 12), namely on an average of $g(r,\tau)$ computed along the horizontal, vertical, and diagonal directions of the lattice only.

[17]A. Chakrabarti and J. D. Gunton, Phys. Rev. B **37**, 3798 (1988).

[18]It is interesting to note that surface diffusion by itself, in the absence of the Lifshitz-Slyozov mechanism, leads to a growth law with $x=\frac{1}{4}$.

[19]There is a misprint in Eq. (2), Ref. 7 [G. F. Mazenko and O. T. Valls, (private communication)]. A factor of 2 was apparently omitted.