Universality and scaling for the structure factor in dynamic order-disorder transitions

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The universal form for the average scattering intensity from systems undergoing order-disorder transitions is found by numerical integration of the Langevin dynamics. The result is nearly identical for simulations involving two different forms of the local contribution to the free energy, supporting the idea that the model A dynamical universality class includes a wide range of local free-energy forms. An absolute comparison with no adjustable parameters is made to the forms predicted by theories of Kawasaki-Yalabik-Gunton, Ohta-Jasnow-Kawasaki, and Mazenko. The numerical results are well described by the Ohta-Jasnow-Kawasaki theory, except in the crossover region between scattering dominated by domain geometry and scattering determined by Porod’s law. [S1063-651X(98)04411-0]

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I. INTRODUCTION

Phase ordering by quenching from a region of the phase diagram where a material is uniform to one where several phases coexist at equilibrium provides an important technique for creating multiphase materials that are inhomogeneous on mesoscopic length scales. Since the macroscopic properties of such materials can be quite different from those of the constituent phases, and depend sensitively on the mesoscopic structure, a solid understanding of pattern formation in phase-ordering systems is important to several branches of materials science. Examples include precipitation strengthening in metals [1] and fabrication of glasses [2].

Universality allows one to describe the structure and dynamical properties of diverse phase-ordering systems using models that only take into account properties such as conservation laws and order-parameter symmetries. Two important universality classes involve only local relaxational dynamics and an order parameter that can be represented as a scalar field [3,4]. The order parameter is conserved in the first class, called model A. This can be used, for example, to model anisotropic magnets and alloys undergoing order-disorder transitions. In the second class, called model B, the order parameter is a locally conserved quantity, and relaxation proceeds by diffusion away from regions of high chemical potential. When hydrodynamic modes and strain effects can be ignored, binary mixtures and alloys are described by this model.

Scaling is the hypothesis that the behavior of the system over a large range of length scales can be described in terms of a single characteristic length R. A necessary condition for scaling is that R must be well separated from any other microscopic or macroscopic length scales present in the system. For many phase-ordering processes, the characteristic length has been found to have a power-law dependence on the time t elapsed since the quench, \( R \sim t^\alpha \). In nonconserved systems the power law is readily observable with \( n = \frac{1}{2} \). For conserved systems the late-time growth exponent is \( n = \frac{3}{2} \), although processes occurring at early times may mask this behavior. Both of these universality classes are important. However, the work reported here is restricted to nonconserved systems, for which more detailed theoretical results are available.

Here scaling and universality in phase ordering are tested by comparing numerical solutions of the Langevin equation involving two different forms for the local part of the free-energy functional: the \( \phi^4 \) Ginzburg-Landau form and a piecewise-linear triangular form. The numerical details are presented in Sec. II. The agreement between the structure factors found for both forms is discussed in Sec. III. Combined with a mapping to sharpen the interfaces, numerical solution of the Langevin equation allows comparison to analytic theories for the structure factor, without fitting parameters. In Sec. IV four theories are reviewed and compared to the numerical results. The Ohta-Jasnow-Kawasaki (OJK) theory agrees with the simulations at small and large wave vectors, with noticeable deviations only for intermediate wave vectors. The simulation results clearly support this theory over the Kawasaki-Yalabik-Gunton theory, which only differs from OJK by a scale factor. The two theories by Mazenko are qualitatively correct, but the zeroth-order theory agrees with the simulations better than the second-order theory. Section V is a brief summary of our results.

II. NUMERICAL MODEL

A general model of phase ordering can be constructed from a free energy composed of a local term with two degenerate minima and a nonlocal term representing the contribution from spatial fluctuations [3],

\[
F[\psi(r, \tau)] = \int d\mathbf{r} \{ f(\psi(r, \tau)) + \frac{1}{2} |\nabla \psi(r, \tau)|^2 \},
\]

where the scalar order-parameter field \( \psi(r, \tau) \) depends on position \( \mathbf{r} \) and time \( \tau \). The dynamics of the model are governed by a Langevin equation.
The strength of the noise is given by the normalized temperature $\epsilon$, which is the only parameter in the model after rescaling the order parameter, space, and time [5]. The primary effect of thermal fluctuations is to introduce randomness at early times [6]. In the late-time scaling regime the dynamics of this model are controlled by a zero-temperature master curve can be expressed in terms of a time-independent function can be expressed in terms of a time-independent function.

The local part of the free-energy functional $F$ is usually chosen to have the Ginzburg-Landau form

$$f[\psi(\mathbf{r}, \tau)] = -\frac{1}{2} \psi^2(\mathbf{r}, \tau) + \frac{1}{4} \psi^4(\mathbf{r}, \tau),$$

which is a double well with degenerate minima. The corresponding dynamical equation is

$$\frac{\partial \psi(\mathbf{r}, \tau)}{\partial \tau} = (1 + \nabla^2) \psi(\mathbf{r}, \tau) - \psi^3(\mathbf{r}, \tau) + \sqrt{\epsilon} \eta(\mathbf{r}, \tau),$$

which is commonly called the time-dependent Ginzburg-Landau (TDGL) equation.

To test universality for nonconserved scalar order-parameter systems, comparisons need to be made with the standard TDGL model. The new model still needs to follow the nonconserved dynamics of Eq. (2) for a scalar order parameter with two degenerate equilibrium values. One obvious aspect to change is the form of the local part of the free-energy functional, $f[\psi(\mathbf{r}, \tau)]$. A choice quite different from the Ginzburg-Landau form is a piecewise-linear triangular double well, which is not harmonic near the minima. This form is compared to the $\psi^3$ Ginzburg-Landau potential in Fig. 1. It has been chosen to match the Ginzburg-Landau potential at the extrema, i.e., $f(\pm 1) = -\frac{1}{4}$ and $f(0) = 0$. The Langevin equation for this potential is

$$\frac{\partial \psi(\mathbf{r}, \tau)}{\partial \tau} = \sqrt{\epsilon} \eta(\mathbf{r}, \tau) + \nabla^2 \psi(\mathbf{r}, \tau) - \left\{ \begin{array}{ll} -\frac{1}{4}, & \psi < -1 \\ \frac{1}{4}, & -1 \leq \psi < 0 \\ -\frac{1}{4}, & 0 \leq \psi < +1 \\ \frac{1}{4}, & \psi \geq +1. \end{array} \right.$$  

The simulations with both potentials were conducted on square lattices with periodic boundary conditions and lattice constant $\Delta \tau = 1$. The system size considered was $L_x = L_y = L = 1024$. The Laplacian was approximated by an eight-point form [7], and a simple Euler integration scheme with $\Delta \tau = 0.05$ was used to collect data every 25 time units up to a maximum of $\tau = 2000$. Results for the TDGL equation were averaged over 100 realizations; only ten realizations of the piecewise-linear model were needed to confirm its agreement with the TDGL results.

**III. UNIVERSALITY**

The average structure of the system at a specific time after the quench can be described by the order-parameter correlation function

$$C(|\mathbf{r} - \mathbf{r}'|, \tau) = \langle \psi(\mathbf{r}, \tau) \psi(\mathbf{r}', \tau) \rangle,$$

where $\langle \cdots \rangle$ denotes averaging over the ensemble of quenches as well as the volume. When scaling holds, the correlation function can be expressed in terms of a time-independent master curve.
where $\bar{r} = r/R(\tau)$. While microscopy techniques can be used to measure $C$, scattering experiments probe its Fourier transform $S(k, \tau)$, called the structure factor. The structure factor is also related to a time-independent form

$$S(k, \tau) = R^d(\tau)F(q),$$

where $q = kR(\tau)$ and $d$ is the dimension of the system.

When infinitely sharp domain interfaces are randomly oriented throughout the system, the small-$r$ form of the correlation function is linear, $\bar{C}(r) = 1 - \alpha r + \cdots$, where $\alpha$ is proportional to the surface area to volume ratio for the domains [8]. This corresponds to a power-law decay of the scaled structure factor at large $q$, $F(q) = (2\pi/\alpha)q^{-(d + 1)}$, called Porod’s law [4,8]. These results are valid for $1/q$ much smaller than the characteristic length, but much larger than the domain interface width.

The assumption of infinitely sharp interfaces is used in most situations, e.g., experimental and analytic studies. By design, the domain interface in the simulations has a width of about $\sqrt{2}$. We compensate for this by using a nonlinear mapping [9] of $\psi(r, \tau)$ to $\pm 1$ before finding the Fourier transform $\hat{\psi}(k, \tau)$ [7]. The structure factor is

$$S(k, \tau) = \langle |\hat{\psi}(k, \tau)|^2 \rangle,$$

where $|\hat{\psi}(k, \tau)|^2$ is the scattering intensity associated with a particular domain pattern $\psi(r, \tau)$, and $\langle \cdots \rangle$ represents averaging over the ensemble of quenches. The simulation results used to estimate the structure factor were found by “onion-shell” binning of the two-dimensional scattering intensity into a one-dimensional function [7] of $k$, averaging over all trials, and then scaling the result using the average wave vector of each bin.

Including noise in the simulation has three drawbacks. The first is purely practical: generating Gaussian random numbers for $\eta$ is computationally expensive. The second drawback has a physical basis. Ohta [10] showed that thermal fluctuations in the TDGL equation retard growth of the characteristic length and cause a broadening of $F(q)$. Third, the thermal noise causes the structure factor to cross over to the equilibrium $q^{-2}$ behavior for large $q$, thus masking the $q^{-(d + 1)}$ Porod tail. For these reasons, we have simulated the TDGL equation at zero temperature with the early-time fluctuations implemented by an initial condition where $\psi(r, \tau = 0)$ consists of independent random numbers distributed uniformly on $[-0.1, 0.1]$.

On the other hand, thermal fluctuations are essential for simulating the piecewise-linear model. For this model at $T = 0$, the domain growth stops after a finite time. The time until the evolution stops, as well as the corresponding characteristic length, depends on the random initial condition. If a system which has stopped coarsening is briefly heated, the growth continues for a while and then stops again. In addition, making the potential shallower prolongs growth and yields larger domains. The arrested growth appears to result from trapping into metastable configurations associated with finite wavelength corrugations on the interfaces. A constant, moderate level of thermal noise completely prevents this trapping phenomenon, so noise with $\epsilon = 0.1$ was included in simulations of the piecewise-linear model.

An estimate of $F(q)$ was constructed for each local form by combining the estimates at different simulation times with the restrictions $\tau > 500$ and $k < 0.5$. For $k$ larger than this, lattice effects become important. The estimates of $F(q)$ for both models are presented in Fig. 2, with $R(\tau) = \sqrt{2\tau}$ (see Sec. IV). The agreement seen in Fig. 2(a), a log-log plot of $F$ vs $q$, is very good. The Porod tail at large $q$ can be highlighted by plotting $q^{d+1}F(q)$ against $q$, as shown in Fig. 2(b). This was found independently of the structure factor, using $q = kR(\tau)$ before the binning and averaging. The agreement is quite good well into the power-law tail. The claim that the details of the local part of the free-energy functional do not influence the universal behavior appears well supported by the numerical models.
IV. ANALYTIC THEORIES

Several theories for the universal form of the scaling function for nonconserved systems exist. They can be compared to the numerical integration of the TDGL equation without adjustable parameters.

One theory was developed by Ohta, Jasnow, and Kawasaki [10,11]. The OJK theory starts from a diffusion equation for a Gaussian auxiliary field \( u(\mathbf{r},\tau) \),

\[
\frac{\partial u(\mathbf{r},\tau)}{\partial \tau} = D \nabla^2 u(\mathbf{r},\tau),
\]

where the interfaces in the inhomogeneous material are defined to be the set of \( \mathbf{r} \) such that \( u(\mathbf{r},\tau) = 0 \), and the order-parameter field is obtained by the mapping \( \psi(\mathbf{r},\tau) = \text{sgn}[u(\mathbf{r},\tau)] \). The coefficient \( D = 4 \rho_d \) is the diffusion constant for the interface. The factor \( \rho_d = (d-1)/d \) results from assuming that the interfaces are randomly oriented. As time progresses, the characteristic size of the single-phase domain grows as \( R_{OJK}(\tau) = \sqrt{D \tau} \), in accordance with the Lifshitz-Slyozov-Wagner theory of domain growth driven by surface tension [12,13]. The two-point correlation function for the order-parameter field in this model has the simple analytic form

\[
C_{OJK}(r,\tau) = \frac{2}{\pi} \arcsin \left[ \exp \left( -\frac{r^2}{2R_{OJK}^2(\tau)} \right) \right].
\]

Since the system is isotropic, the Fourier transform of this correlation function can be written in terms of a radial integral over a Bessel function, and the scaling form of the structure factor is [11,14]

\[
F_{OJK}(q) = \frac{2}{\pi} \frac{(2\pi)^{d/2}}{q} \int_0^\infty dw \, w^d [\exp(w^2) - 1]^{-1/2} \times (qw)^{1-d/2} J_{d/2}(qw).
\]

Except for the dimensional dependence of \( \rho_d \) in \( R(\tau) \), this result agrees with the perturbation calculation by Kawasaki, Yalabik, and Gunton (KYG) [15].

Another theoretical approach to phase ordering has been developed by Mazenko and co-workers [16–19]. This theory also starts with a Gaussian auxiliary field \( u(\mathbf{r},\tau) \). In contrast to the OJK theory, \( u(\mathbf{r},\tau) \) is interpreted as the distance from \( \mathbf{r} \) to the closest interface, and it is mapped onto the order-parameter field using the equilibrium interface profile \( \psi(\mathbf{r},\tau) = \psi_{eq}(u(\mathbf{r},\tau)) \). The characteristic length is then defined in terms of the rms distance to the nearest interface, \( R_{0}(\tau) = \frac{1}{\sqrt{\pi}} u(\mathbf{r},\tau)^2 \). Here the subscript 0 is used to denote the zeroth-order theory developed in Refs. [16–18]. Using the Gaussian properties of \( u(\mathbf{r},\tau) \), the TDGL equation can be used to find a closed, nonlinear partial differential equation which describes the scaling form of the correlation function

\[
\frac{d^2 C_0(v)}{dv^2} + \left( \frac{d-1}{v} + \mu_0 v \right) \frac{dC_0(v)}{dv} + \tan \left[ \frac{\pi}{2} C_0(v) \right] = 0,
\]

where \( v = r/R_0(\tau) \) is the scaled length. The characteristic length is \( R_0(\tau) = 4 \mu_0^2 \tau \) with \( \mu_0 \) an undetermined parameter. The correlation function must obey the boundary condition \( C_0(0) = 1 \). As a consequence, the third term in Eq. (14) acquires a \( 1/v \) singularity at \( v = 0 \). This singularity must cancel against the \((d-1)/v\) factor in the second term, resulting in the requirement that \( dC_0(v)/dv = \sqrt{2/\pi(d-1)} \) at \( v = 0 \). Therefore, the first derivative of \( C_0(v) \) at \( v = 0 \) is not available to be used as the second, independent constant in the solution of Eq. (14). Instead, one must determine the value of \( \mu_0 \) such that \( C_0(v) \) becomes integrable as \( v \to \infty \). For \( d = 1 \) the asymptotic form agrees with that found analytically for the kinetic Ising model with Glauber dynamics [20,21], while in the limit \( d \to \infty \) the function \( C_0 \) becomes identical with the OJK result [18]. For finite \( d > 1 \), determining \( C_0 \) is equivalent to the numerical solution of a nonlinear eigenvalue problem [17,22,23].

Recently, Bray and co-workers [4,24] and Yeung, Oono, and Shinozaki [25] criticized the use of Gaussian fields in constructing theories of phase ordering. Noting this criticism, Mazenko [19] expanded his theory to include a \( u(\mathbf{r},\tau) \) whose distribution is an expansion around a Gaussian. The theory with the second-order correction, here marked with the subscript 2, produces a new differential equation for the scaled correlation function \( \tilde{C}_2(v) \),

\[
\frac{d^2 \tilde{C}_2(v)}{dv^2} + \left( \mu_2 v + \frac{d-1}{v} \right) \frac{d\tilde{C}_2(v)}{dv} + \tan \left[ \frac{\pi}{2} \tilde{C}_2(v) \right] = 0.
\]

Here \( \tilde{H}_2(v) \) is a new function governed by

\[
\frac{d\tilde{H}_2}{dv} = b \tilde{C}_2 - \sqrt{\tilde{C}_2^2 + \alpha_0 q_2 (1-b)}.
\]

A corrected eigenvalue \( \mu_2 \neq \mu_0 \) results, and the characteristic length becomes \( R_2(\tau) = 4 \mu_2^2 \tau \) with the scaled length \( v = r/R_2(\tau) \). A new eigenvalue \( q_2 \) is introduced, and the resulting double eigenvalue problem must solved numerically [26].

While experimental tests ultimately determine the validity of any theory, finding the characteristic length \textit{a priori} is difficult for experiments, as well as many simulation techniques. The characteristic length may be taken as a fitting parameter, with which data can be fit well to all three theories. Such fitted comparisons for Monte Carlo simulations made by OJK [11] showed quantitative disagreements in the tail of the correlation function. Later Oono and Puri [27]
used cell dynamics simulations to show that the deviations might be caused by the nonzero interface width present in simulations. Blundell, Bray, and Sattler [24] also used cell dynamics simulations to test the theories. To compare their simulation results to theory without fitting, they reduced $R(t)$ to a parametric variable by plotting values for the correlation function for the order parameter against values for the correlation function for the square of the order parameter for many $(r, t)$. The simulation results plotted this way show good scaling, but the two Gaussian theories give the same result for this particular scaling function [24,25]. As a consequence, this method does not provide a strong test of the two forms, which are, in fact, noticeably different, as we will see below. The question of how well these theories predict the scaling function $C(\tau)$ therefore remains open.

Numerical simulation of the TDGL equation can be compared to the theories without fitting parameters, since analytic forms for $R(t)$ are known. We have chosen to work in terms of $R_{OJK}$, since $R_0$ and $R_3$ depend on numerically determined eigenvalues. The TDGL estimates for the two-dimensional $F(q)$ for several times are presented in Fig. 3, along with the predictions of the theories. Figure 3(a) is a log-log plot, showing good collapse of the simulation data. Concerning the difference between the theories of OJK and

**FIG. 3.** Scaling of the structure factor for the two-dimensional time-dependent Ginzburg-Landau model. The scaling variable is $q = kR_{OJK}(\tau)$. The simulation data for different times scale quite well, using the characteristic length $R_{OJK}(\tau) = \sqrt{4\mu_0 \tau}$, except at large wave vectors where lattice effects are important. The curves represent the theoretical scaling forms discussed in the text: OJK (solid), Mazenko zeroth order (dashed), and Mazenko second order (dotted). (a) For small values of $q$ ($q < 3$), the simulations agree quantitatively with the OJK theory. (b) Plot emphasizing the Porod’s law tail at large $q$, the trend with time indicates that the OJK theory gives a better estimate of this amplitude than either of the Mazenko theories. The theoretical amplitude of the second-order theory is lower than that for the first-order theory.
KYG, much better agreement with the simulations is found using the factor $\rho_d$ predicted by OJK, so we only consider that theory. In Fig. 3(a) the obvious differences between the theories shown are the magnitude at small $q$ and the behavior near the shoulder at intermediate $q$. For both of these features, the simulation data agree significantly better with the OJK theory than with those of Mazenko.

Figure 3(b) highlights the power-law tail at large $q$. The scaling of the simulation data is quite good through the large peak for all of the times presented here. Where the simulation data scale, they agree very well with the OJK theory. There is a noticeable second peak in the OJK and second-order Mazenko theories that is absent in the zeroth-order Mazenko theory. Unfortunately, the simulation data cannot be used to test the presence of a second peak because they do not scale in this region for the practically attainable simulation times. Instead, a pronounced trough is seen, which decays with time. Simulations for significantly larger systems and longer times would be needed to achieve asymptotic scaling in this region.

At values of $q$ higher than this, the scaling regime also has not been reached, preventing an estimate of the Porod amplitude $(2\pi/\alpha)$ for the simulations. However, a clear trend in the data is apparent, indicating an amplitude which is significantly higher than those predicted by both of Mazenko’s theories. In fact, the second-order theory has the smaller Porod amplitude. This is a result of the relationship [19]

$$\alpha_2 = \alpha_0 \frac{\sqrt{q_2(q_2+2)}}{q_2+1},$$  

where $q_2$ is the second-order eigenvalue. Since $\alpha_2 > \alpha_0$ for any value of $q_2$, the predicted Porod amplitude is necessarily smaller for the second-order theory. The disagreement with simulations increasing with the order of the theory is also true for the decay of the order-parameter autocorrelation function, although estimates for both of Mazenko’s theories are better than that for OJK [7,19,28].

Very recently, Emmott presented a perturbation expansion about the OJK form for the correlation function, with a lowest-order term of order $1/d^2$ [29]. Our numerical results presented here suggest that the OJK theory may provide a good starting point for a more complete theory of the dynamics of phase separation in systems with nonconserved order parameter. However, we note that the lowest-order correction provided by Emmott’s approach would produce a change in the Porod amplitude (which is well described by OJK), while it shares with OJK the inability to correctly describe the decay of the order-parameter autocorrelation function [7,19,28].

V. CONCLUSIONS

Numerical simulations of the TDGL equation have been used to confirm the universality of model A systems with different forms for the local free-energy contribution. With no adjustable parameters, the scaling form of the structure factor from simulations is found to agree quantitatively with the OJK theory at scaled wavevectors $q<3$. The simulation results agree much better with OJK than KYG, indicating that the scale factor $\rho_q = (d-1)/d$ should be included in the characteristic length. At large $q$, the numerical results have not yet entered the asymptotic scaling regime, even at our latest simulation times. However, the trend is toward OJK and away from both of the Mazenko theories. Although the OJK theory has been found here to describe well the scaling form of the structure factor, it does not correctly describe the decay of the order-parameter autocorrelation function [7,28].

Mazenko’s theories describe the estimated scaling form only qualitatively. Interestingly, the zeroth-order theory gives better agreement with simulations than does the second-order theory. Since, aside from $d=1$, $d=2$ is expected to be the dimension where the theoretical results are the most different, computational cost has lead us to forgo three-dimensional simulations at this time. Another aspect of universality not considered here is the effect of anisotropy [30].

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In our numerical calculations of the scaling functions, we directly used the $d=2$ Fourier transform of Eq. (12) without the integration by parts, which leads to Eq. (13).

We constructed an implementation of this theory where values of $\mu_0$ were varied interactively. To guard against round-off errors, the eigenvalue problem was solved twice in 64-bit precision, once with a step size of $10^{-4}$ and once with $10^{-3}$. The solutions agreed to six significant digits for $v \leq 5.8$, beyond which point $\tilde{C}_0(v) < 10^{-8}$. We find $\mu_0 = 1.10420873646(1)$. The scattering intensity was constructed using a Romberg integration scheme and cubic spline interpolation from Ref. [23].

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