Possibilities and limitations of Gaussian-closure approximations for phase-ordering dynamics

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The nonlinear equations describing phase-ordering dynamics can be closed by assuming the existence of an underlying Gaussian stochastic field which is nonlinearly related to the observable order-parameter field. We discuss the relation between different implementations of the Gaussian assumption and consider the limitations of this assumption for phase-ordering dynamics. The fact that the different approaches give different results is a sign of the breakdown of the Gaussian assumption. We discuss both the nonconserved and conserved order-parameter cases. We demonstrate that the Gaussian assumption cannot describe the large length-scale behavior in the latter case.

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I. INTRODUCTION

After a system is quenched from the disordered to the ordered phase, domains of the ordered phases form and grow. At late stages, it is empirically known that the phase-ordering process obeys dynamical scaling, i.e., the spatial distribution of domains can be described by a single time-dependent length $L(t)$. On this length scale, the phase-ordering process depends only on a few general features of the dynamics. Due to the inherently nonlinear nature of the dynamics, understanding the phase-ordering process remains a challenge [1–3]. Analytic progress has been confined to the case of $O(n)$ component order parameter in the limit of large n [4–6]. For $n \leq d$, where d is the spatial dimension, topological defects become important and progress has been limited to dimensional analysis of the defect motion [2,7,8] and methods by which the nonlinear equations describing the dynamics are "closed" (closure approximations) [9–14]. Some progress is achieved by assuming that there exists an underlying Gaussian field which is nonlinearly related to $\psi(\mathbf{r},t)$ [12–14].

In this paper, we will explore the reliability of closure approximations based on the assumption of an underlying Gaussian stochastic field for phase-ordering dynamics both with and without conservation of order parameter. We clarify the relations among various approaches and the limitations of the Gaussian assumption. In particular, we discuss the interface approach for the non-conserved order parameter (NCOP). We clarify misunderstood points in Ref. [13] and more importantly, we discuss this in a manner easily generalizable to the conserved-order-parameter case. For the NCOP case we demonstrate that the various approaches use exactly the same assumptions. Thus the discrepancies among the results by different approaches indicate the limitation of the Gaussian approximation. Approximate relations between these approaches have also been discussed by Bray and Humayun [15] very recently.

The interface approach is readily generalizable to the conserved-order-parameter case. For this case, the interface approach gives a real-space correlation function which agrees well with the empirical result up to its second zero. However, the Gaussian approach is more fundamentally flawed in its description of the long-distance correlations in that, in order to reproduce the empirically observed scattering intensity, the Gaussian field must have a negative spectral density at small wave number. Therefore the Gaussian approximation cannot be used to recover the entire form factor.

In Sec. II we discuss the relations among the approaches for nonconserved-order-parameter case. In Sec. III we study the approximate Gaussian nature of $u(\mathbf{r},t)$ through direct numerical updating of the evolution equation for u. We find that the single-point probability distribution function $P(u)$ decays as a Gaussian at the tails but decays slower than a Gaussian function near $u = 0$. In Sec. IV we present our findings for the conserved-order-parameter case.

II. NONCONSERVED ORDER PARAMETER

The simplest model of phase ordering without conservation of order parameter is the time-dependent Ginzburg-Landau (TDGL) equation

$$\frac{\partial\psi}{\partial t} = -\mu_B(\psi) + \frac{\xi^2}{2}\nabla^2\psi, \tag{2.1}$$

where ξ is the interfacial width, $\psi(\mathbf{r},t)$ is the scalar order parameter, and $\mu_B(\psi)$ is the portion of the local chemical potential without gradient terms. We assume μ_B is an odd function of ψ, the equilibrium values of ψ are ± 1, and that there exist a stationary planar interface solution $f(z)$, obeying $0 = -\mu_B(f(z)) + (\xi^2/2)d^2f/dz^2$. The exact form of $f(z)$ is unimportant except that $f(z)$ in-

creases monotonically from -1 to 1 over length ξ [13,16]. This universality is closely related to the universality in the dispersion relation around the interface [17].

At late times, the domain growth is determined by the motion of the sharp interfaces. The interface dynamics can be derived from the TDGL [7,8,18],

$$v_n(\mathbf{r},t) = -\frac{\xi^2}{2}\,\kappa(\mathbf{r},t). \tag{2.2}$$

The normal velocity of the interface v_n is positive if the "minus" phase moves into the "plus" phase, the normal $\hat{\mathbf{n}}$ points into the plus phase, and the local curvature $\kappa = \boldsymbol{\nabla}\cdot\hat{\mathbf{n}}$ is positive for a bump of the minus phase into the plus phase.

These equations are the basis of different analytic approaches. Ohta, Jasnow, and Kawasaki [12] used the interface dynamics (2.2) while Oono and Puri [13] and Mazenko [14] started with the full bulk dynamics (2.1). In the bulk approach, an auxiliary field u is introduced through $\psi = f(u)$, and is assumed to be Gaussian. One then seeks a closure approximation in terms of correlations of u (bulk-u approach) [13] or in terms of correlations of ψ (bulk-ψ approach) [14]. We concentrate on the interface approach as a similar method can be attempted for the conserved order parameter case. We clarify misunderstood points in Ref. [13] and show that a suitably corrected interface and bulk-u approach uses identical assumptions. The discrepancy between the two bulk approaches then is a test of the reliability of the Gaussian assumption.

The essence of the interface approach is to write the interfacial dynamics using an indicator field $u(\mathbf{r},t)$ [12]. The indicator field is a smooth function such that $u>0$ ($u<0$) in the plus (minus) phase and $u=0$ at the interface. In terms of u, Eq. (2.2) becomes $\partial_t u - |\boldsymbol{\nabla}u|\boldsymbol{\nabla}\cdot(\boldsymbol{\nabla}u/|\boldsymbol{\nabla}u|) = 0$. It has recently been proven that, independent of the choice of u in the bulk, this equation correctly describes the dynamics of infinitesimally thin interfaces driven by mean curvature [19,20]. However, further progress requires the preaveraging of the dynamical equations via a random orientation assumption for the interfaces [12]. Unfortunately, the results obtained by the preaveraged version, $\partial_t u = (d-1)\nabla^2 u/d$, where d is the spatial dimensionality, depends crucially on the choice of u in the bulk, so some auxiliary condition on the meaning of u must be imposed. The original interpretation was that $|u|$ is the distance to the interface so that u must grow as $L(t)$. However, it was found that u decays as $t^{-1/2}$. These difficulties motivated the bulk approach in Ref. [13]. Here we emphasize a derivation without these difficulties as a similar approach will be attempted for the conserved-order-parameter case.

We define u in the same manner as Ohta, Jasnow, and Kawasaki (OJK) enforcing the condition $\boldsymbol{\nabla}u(\mathbf{r},t) = \hat{\mathbf{n}}(\mathbf{r},t)$ near $u=0$ [12,13]. In terms of u, the interfacial dynamics [Eq. (2.2)] is

$$\frac{\partial u}{\partial t} = \frac{\xi^2}{2}\nabla^2 u. \tag{2.3}$$

However, Eq. (2.3) can only hold at $u=0$, since, if it were true in the bulk, the condition that $|\boldsymbol{\nabla}u| = 1$ near the interface would be violated [13]. We assume that this condition can be met by extending Eq. (2.3) into the bulk by adding a "Lagrange multiplier function" $\tilde{P}(u,\boldsymbol{\nabla}u)$ to the right-hand side of Eq. (2.3),

$$\frac{\partial u}{\partial t} = \frac{\xi^2}{2}\left[\nabla^2 u + \tilde{P}(u,\boldsymbol{\nabla}u)\right]. \tag{2.4}$$

The function $\tilde{P}$ has the following properties: $\tilde{P}(u,\boldsymbol{\nabla}u)|_{u=0} = 0$, since Eq. (2.3) must be recovered at $u=0$. Due to the symmetry of the TDGL, $\tilde{P}(u,\boldsymbol{\nabla}u)$ is an odd function of u and an isotropic function of derivatives of u. Since the interface dynamics depends only on local properties of the interface, $\tilde{P}(u,\boldsymbol{\nabla}u)$ is assumed to be local, i.e., $\tilde{P}(u,\boldsymbol{\nabla}u)$ depends only on a finite number of gradients of u. The two-point correlation function $\langle u_1 u_2\rangle$ obeys

$$\frac{\partial\langle u_1u_2\rangle}{\partial t_1} = \frac{\xi^2}{2}[\nabla_1^2\langle u_1u_2\rangle + \langle\tilde{P}(u_1,\boldsymbol{\nabla}_1u_1)u_2\rangle], \tag{2.5}$$

where $u_i = u(\mathbf{r}_i,t_i)$ and $t_1 \neq t_2$. Note that the local constraint $|\boldsymbol{\nabla}u| = 1$ at the interface forces $\langle u_1^2\rangle$ to grow as L_1^2, where $L_i = L(t_i)$.

Now, assume u is a Gaussian stochastic field. For any Euclidean symmetric Gaussian stochastic field and $\tilde{P}$ with the conditions above, $\langle\tilde{P}(u_1,\boldsymbol{\nabla}u_1)\,u_2\rangle$ has a simple form:

$$\langle\tilde{P}(u_1,\boldsymbol{\nabla}u_1)\,u_2\rangle = p(t_1)\,\langle u_1u_2\rangle, \tag{2.6}$$

since $p(t)$ depends only on $\langle u^2\rangle$ and $\langle|\boldsymbol{\nabla}u|\rangle$. Thus the correlation function obeys

$$\frac{\partial\langle u_1u_2\rangle}{\partial t} = \xi^2\left[\nabla^2\langle u_1u_2\rangle + p(t)\,\nabla^2\langle u_1u_2\rangle\right], \tag{2.7}$$

where we have assumed $t_1 = t_2 = t$. The Gaussian approximation means that the detailed local constraint $|\boldsymbol{\nabla}u| = 1$ is no longer met, but is replaced by a global constraint $\langle u^2\rangle \sim L^2$. In the scaling limit, this requires that $\xi^2 p(t) = (d+2)/(2t)$. The same result was obtained in Ref. [13] to enforce the condition that $\xi/L(t) \sim t^{-1/2}$. One difference between the above and the result in Ref. [13] is the presence of a factor of $(d-1)/d$ in front of the diffusive term, which is also present in the OJK approach. The reason for this difference is the lack of the local constraint $|\boldsymbol{\nabla}u| = 1$ at the interface. However, there is also a difference in the derivation. In OJK one makes a random interface assumption which results in u being a Gaussian variable while, in the above discussion, the Gaussian assumption is made directly rather than being the result of a secondary assumption. We take this more direct approach in our analysis of the conserved-order-parameter approach in Sec. IV.

Let us now discuss the bulk approaches. We demonstrate that the interface and bulk-u approaches are exactly equivalent. The approximate relation between these approaches has also been recently discussed by Bray and Humayun [15]. In terms of the auxiliary field u, application of the chain rule to the TDGL gives

$$\frac{\partial u}{\partial t} = \frac{\xi^2}{2}\left[\nabla^2 u + \left(1 - |\nabla u|^2\right) Q(u)\right], \tag{2.8}$$

where $Q(u) \equiv -(df/du)^{-1} d^2 f/du^2$. In general, $Q(u)$ is an odd function of u and $Q(u)$ approaches $d\mu_B/d\psi|_{\psi_{eq}}\ \mathrm{sgn}(u)$ in the limit $\xi \to 0$. Note that this is exactly of the form given by Eq. (2.4). In fact $\left(1 - |\nabla u|^2\right) Q(u)$ is the Lagrange multiplier function of the interface approach. Therefore it is clear that the bulk-u and interface approaches are exactly equivalent.

We complete the calculation to show that $p(t)$ in Eq. (2.6) can be obtained explicitly. Assuming that u is a Gaussian stochastic field, for general Q, the $\langle u_1 u_2 \rangle$ obeys

$$\frac{\partial \langle u_1 u_2 \rangle}{\partial t} = \xi^2\left[\nabla^2 \langle u_1 u_2 \rangle + p(t) \langle u_1 u_2 \rangle\right], \tag{2.9}$$

where $t_1 = t_2 = t$. It is clear that in the scaling limit this equation must be the same as that obtained in the interface approach. [Bray and Humayun have also recently pointed out the equivalence for a particular form of $Q(u)$, which can only come about from a nonanalytic free energy [15]. However, we do not expect this distinction to be important.] We can choose Q so that the Gaussian integrals are simplified

$$Q(u) = \frac{i}{\pi}\int_{-\infty}^{+\infty} d\omega \frac{1}{\omega} \exp\left(-\frac{\xi^2\omega^2}{2} - i\omega u\right). \tag{2.10}$$

That is, $Q(u)$ is the sign function mollified by a Gaussian function. Now we can explicitly perform the Gaussian average to obtain

$$p(t) = \frac{1 - \langle |\nabla u|^2 \rangle}{\sqrt{\pi(\xi^2 + \langle u^2 \rangle)/2}}, \tag{2.11}$$

and, in the limit of $\langle u^2 \rangle \gg \xi^2$,

$$\langle \psi_1 \psi_2 \rangle = \frac{2}{\pi} \arcsin\left(\frac{\langle u_1 u_2 \rangle}{(\langle u_1^2 \rangle + \xi^2)^{1/2} (\langle u_2^2 \rangle + \xi^2)^{1/2}}\right). \tag{2.12}$$

Thus we are left with a closed equation for $\langle u_1 u_2 \rangle$ and a closed ordinary differential equation for $p(t)$. The only meaningful solution $p(t)$ of the latter equation behaves asymptotically just as describe above. The scaling result can also be obtained giving the same asymptotic result as discussed in the interface approach [21].

In the bulk-ψ approach the auxiliary field $u(\mathbf{r}, t)$ is introduced in the same way as in the bulk-u approach. Using the TDGL one obtains an expression for $\langle \psi\psi \rangle$ using the assumption that u is a Gaussian stochastic field. This approach was applied by Mazenko [14] for the nonconserved-order-parameter approach.

Therefore the approximations made in the bulk-u, interface, and bulk-ψ approaches use exactly the same assumptions. However, these methods produce qualitatively different results, such as, for the asymptotic decay two-time correlation function [22–25]. These discrepancies are indications of the failure of this assumption of an underlying Gaussian field.

III. NUMERICAL RESULTS

In the previous discussion we have shown that $\{u(\mathbf{r}, t)\}$ is not a Gaussian random field. However, we have also argued that this assumption may be a reasonable first approximation. In this section we study the statistics of $\{u(\mathbf{r}, t)\}$ directly through a numerical updating of Eq. (2.8). As noted previously, no approximations are needed to proceed from Eq. (2.1) to Eq. (2.8) so this is equivalent to a simulation of the TDGL equation.

For numerical efficiency we choose $Q(u) = 2$ if $u > 1$, $Q(u) = 2u$ if $1 \geq u \geq -1$, and $Q(u) = -2$ if $u < -1$. This effectively approaches 2 $\mathrm{sgn}(u)$ in the limit of $L(t) \gg 1$. We discretize the system with mesh size $\delta x = 1.0$ and time steps $\delta t = 0.05$. To reduce lattice effects we used a sphericalized Laplacian as described in Ref. [16]. For these large time and space steps the update corresponds to a cell dynamical system (CDS) [16]. The simulation was performed on 800×800 lattices with periodic boundary conditions, and repeated on 400×400 lattices to check for finite size effects. The results for $n = 400$ and $n = 800$ begin to deviate at about $t = 400$ indicating that the data for $n = 800$ is not affected by size problems. An average was taken over 18 independent initial conditions for the larger lattice.

Figure 1 shows a plot of $L(t)^2$ vs $\langle u(r,t)^2 \rangle$ for $t = 25, 50, 100, 200$, and 400. For $L(t)$ we use the inverse interfacial density. The line has a slope of unity. We see that $L(t)^2 \sim \langle u(r,t)^2 \rangle$, in agreement with the arguments above. We next calculate the single point probability function $P(u,t)$ for each time. Figure 2 shows $\ln P(u^2/\langle u^2 \rangle)$ vs $u^2/\langle u^2 \rangle$ for $t = 50, 100, 200$, and 400. It is clear that the probability distribution scales during this time range. We also observe that the tail of the probability distribution decays in a Gaussian manner but the region near $u = 0$ is flatter than that for the Gaussian distribution. Figure 3 shows the same data plotted with $-\ln[-\ln P(u^2/\langle u^2 \rangle)]$ vs $\ln u^2/\langle u^2 \rangle$. The straight line has a slope of 2. Figure 4 shows the flatness $\langle u^4 \rangle/(\langle u^2 \rangle^2)$. The times are the same as that of Fig. 1 plus the point at $t = 0$. The flatness is 3 at $t = 0$ since the initial

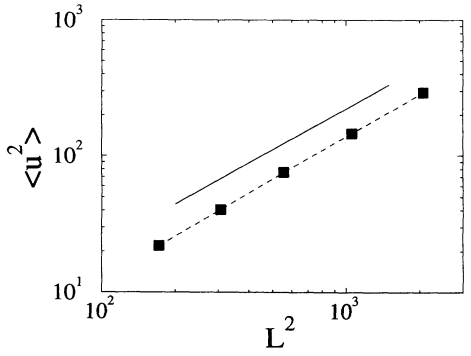

FIG. 1. A log-log plot of $\langle u^2 \rangle$ vs $L(t)^2$ from the 800×800 simulations. The solid line is a slope of 1. The statistical uncertainties are smaller than the symbol sizes. $L(t)$ is the inverse interfacial density. As predicted by the interface approach, $\langle u^2 \rangle$ grows as L^2. A fit to the form $\langle u^2 \rangle = aL^{2b} + c$ gives $b = 1.03$.

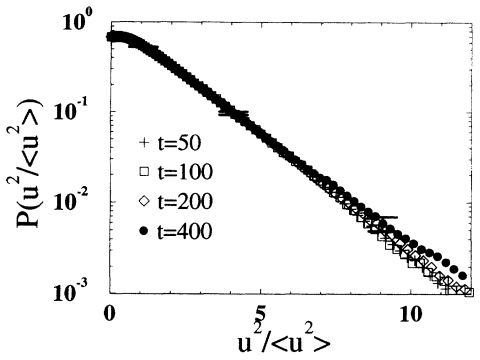

FIG. 2. The single point probability distribution $\ln P(u^2/\langle u^2\rangle, t)$ versus $u^2/\langle u^2\rangle$ for $t = 50, 100, 200$, and 400. Representative error bars are shown. This plot indicates that the tail of the distribution function decays as a Gaussian function, but there is a regime for $u^2/\langle u^2\rangle < 0.5$ which decays slower than predicted by the tails.

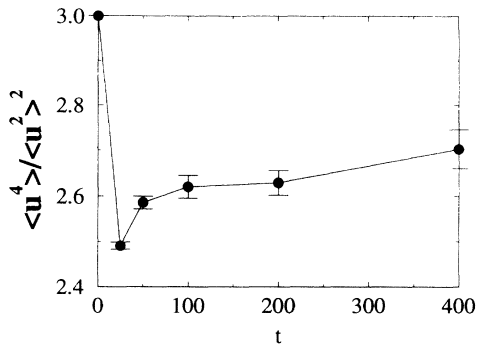

FIG. 4. The flatness $\langle u^4\rangle/\langle u^2\rangle^2$ versus time. The distribution for the initial condition is Gaussian so that at $t = 0$ the flatness is 3. For larger times the flatness is somewhat smaller than that of a Gaussian distribution.

distribution is Gaussian. For larger times the flatness is somewhat less than that expected for a Gaussian distribution.

From our numerical result the single point probability distribution of u is approximately Gaussian at the tails. However, the deviation from Gaussian behavior near $u = 0$ is very important since the location of the interface is at $u = 0$, which we have assumed controls the dynamics. We also note that we have only looked at the single point distribution function. To test the full Gaussianness of $\{u(\mathbf{r}, t)\}$ we must check also the two-point and two-time distributions, etc.

IV. CONSERVED ORDER PARAMETER CASE

In this section we show that the interface approach discussed in the preceding sections can be readily extended to phase ordering dynamics with conserved order parameter (spinodal decomposition) [26,27]. This approach leads to a very good fit of the real-space correlation function up to the second zero of the correlation function. That is, the Gaussian assumption can give good agreement with experiment for small to intermediate distances. However, we also show that the Gaussian approximation is fundamentally flawed in that it cannot correctly describe the long distance, small-wave-number behavior. Using a fitting of the empirical data, we find that Gaussian assumption requires that the spectral density of the Gaussian field be negative at small wave numbers.

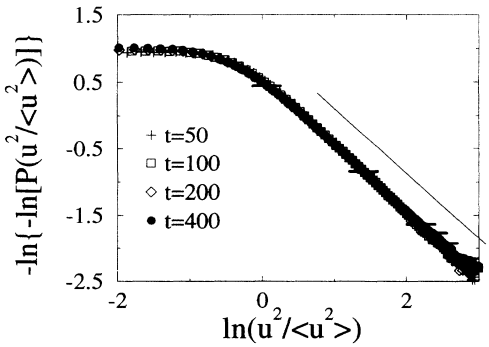

FIG. 3. The same data as in Fig. 2 plotted in the form $-\ln[-\ln P(u^2/\langle u^2\rangle, t)]$ vs $\ln(u^2/\langle u^2\rangle)$. The line has a slope of -1 indicating the Gaussian nature of the tail.

The bulk ψ closure was the first closure method used to study system with conserved order parameter [26,27]. An important difficulty with the bulk ψ approach is that one cannot simply use the nonlinear mapping $\psi(\mathbf{r}, t) = f(u(\mathbf{r}, t))$ to define the indicator field. This is because the condition that $|\psi(\mathbf{r}, t)| < \psi_{\text{eq}}$ is not met and therefore the mapping is not invertible. This is easily seen since, for the conserved case, local equilibrium near the interface requires that the deviation of ψ from its planar interface value is proportional to the local curvature. However, an indicator field can still be introduced using

$$\psi(\mathbf{r}, t) = f(u(\mathbf{r}, t)) + \phi(\mathbf{r}, t),$$

where the ϕ field accounts for the deviation from the planar interfacial profile. This method has been discussed by Mazenko [28].

The interface approach has the important advantage that no additional deviation field ϕ is needed. Using the methods and motivation discussed in Sec. II we now show that the interface approach is easily extended to the conserved-order-parameter case. We note that a Gaussian approach based on the linear dispersion relation for an almost planar interface have been discussed by Ohta and Nozaki [29]. They found a reasonable fit to the scattering intensity (except at low k). However, their approach fixes the growth exponent to be 1/3 by using the linear dispersion relation. In contrast, we will show that that the interface approach, along with the requirement of scaling, automatically gives $L(t) \sim t^{1/3}$.

Our starting point is the interface dynamic equation in terms of the u field

$$G * \delta(u)\partial_t u = \nabla^2 u, \tag{4.1}$$

where G is the Green's function for the Laplacian, i.e.,

$G = -\nabla^{-2}$ [30] and $*$ indicates the convolution. As in the nonconserved case, this equation is correct only at the interface. We assume the following bulk extension of the interface equation:

$$G * \delta(u)\partial_t u = J\nabla^2 u + q, \tag{4.2}$$

where J is a functional of u such that $J = 1$ at the interface and q is a functional of u with $q = 0$ at the interface. The functional q must be nonlocal, since the left-hand side of Eq. (4.2) is nonlocal, but $\nabla^2 q$ must be a local function of u. This extension into the bulk is motivated by an analogous idea behind Eq. (2.4). Applying ∇^2 to Eq. (4.2), and assuming that u is a Gaussian stochastic field, we find (after some straightforward algebra)

$$\frac{1}{2}\frac{1}{\sqrt{2\pi g(0,t)}}\partial_t g(r,t) = -R(t)\nabla^4 g(r,t) - \nabla^2 Q(t) g(r,t) + P(t) g(r,t), \tag{4.3}$$

where $g(r,t) = \langle u(r,t)u(0,t)\rangle$ and P, Q and R are yet unspecified functions of time only. The last term, $P(t)g(r,t)$, is due to the nonlocality of q. We now assume that there is a scaling regime and determine the forms of $R(t)$, $Q(t)$, and $P(t)$ necessary for a scaling solution to exist. The definition of u requires that $\langle u^2\rangle \sim L^2$ for large times, so that, in the scaling limit, $g(r,t)$ must be of the form

$$g(r,t) = L(t)^2 g(r/L(t)), \tag{4.4}$$

where $g(0) = 1$. Rescaling $x = r/L(t)$, we rewrite Eq. (4.3) as

$$\lambda\left(2g(x) - x\frac{dg}{dx}\right) = -R(t)\nabla^4 g(x) - L(t)^2\nabla^2 Q(t) g(x) + L(t)^4 P(t) g(x), \tag{4.5}$$

where $\lambda = L'(t)L(t)^2/2\sqrt{2\pi}$, which must be time-independent asymptotically. The coefficients R, L^2Q, and L^4P must converge to nonzero constants in the $t \to \infty$ limit; if they diverge or vanish, we obtain physically absurd results. Hence, asymptotically Eq. (4.5) becomes

$$\lambda\left(2g(x) - x\frac{dg(x)}{dx}\right) + \nabla^4 g(x) + A\nabla^2 g(x) - Bg(x) = 0, \tag{4.6}$$

where we have rescaled λ to get rid of the numerical coefficient in front of the ∇^4 term, and A and B are constant. The requirement that $g(0)$ is finite fixes B to be $B = (2+d)\lambda$. Hence, in the k space, we arrive at

$$\lambda\frac{dg}{dk} = (-k^3 + k)g, \tag{4.7}$$

where A, which must be positive, has been scaled out and λ remains an unspecified constant. The resultant equation is similar to the Ohta-Nozaki equation [29], although in our case $L \sim t^{1/3}$ follows from our starting point of the interface equation. We note that the derivation of Eq. (4.7) is straightforward if we assume the existence of a scaling form [31].

Using Eq. (4.7) we obtain $C(r/L(t)) = \langle\psi(\mathbf{r},t)\psi(\mathbf{0},t)\rangle$ and fit it to an empirically obtained $C(r/L(t))$ from a very accurate three dimensional CDS simulation of spinodal decomposition [32]. The best fit is obtained for $\lambda = 0.013$. Figure 5 shows that the fit is very good up to approximately the second zero of $C(x)$. This is further shown in the inset in which $x^2C(x)$ is plotted to show that goodness of the fit is not simply because $C(x)$ is small at larger x. However, for larger r/L the deviation from the Gaussian closure result becomes significant. This is particularly apparent in the scattering intensity. In this case there is a very good fit for $q > 0.5$, but the conservation law is violated and $S_{k=0} \neq 0$.

We now ask whether the fact that the interface approach does not describe the low k behavior correctly is due to details of the interface approach or is due to a more fundamental problem with the assumption of an underlying Gaussian field. (With the $B\psi$ approach $S_{k=0} = 0$, but $S_k \sim k^2$ [28] for small k rather than $S_k \sim k^4$, as is observed [32,33].) Note that Eq. (2.12), the relation between $\langle\psi\psi\rangle$ and $\langle uu\rangle$, is independent of the dynamics and is true as long as $u(\mathbf{r},t)$ is a Gaussian stochastic field. Therefore, ignoring terms of order ξ/L, we have

$$C(r,t) \equiv \langle\psi_1\psi_2\rangle = \frac{2}{\pi}\arcsin\left(\frac{\langle u_1 u_2\rangle}{\langle u^2\rangle}\right). \tag{4.8}$$

(This result also holds for the $B\psi$ approach since the additional deviation field ϕ is of order $1/L$. Its direct effects in the correlation function can be neglected in the scaling limit.) Therefore, assuming u is a Gaussian field, we can invert this relation to obtain $\langle u_1 u_2\rangle/\langle u^2\rangle$ from the empirically obtained $C(r,t)$ [32]. Figure 6 shows the spectral density $\langle u_k(t)u_{-k}(t)\rangle$ obtained in this manner using $C(r,t)$ from the three dimensional CDS simulation [32]. The spectral density $\langle u_k(t)u_{-k}(t)\rangle$ becomes significantly negative at small wave numbers $q = kL(t) < 0.5$. [The peak of $\langle\psi_k(t)\psi_{-k}(t)\rangle$ occurs at approximately $q = 1$.] Since the spectral density must be positive definite, we conclude that the Gaussian closure approaches are inherently flawed in describing the large length-scale behavior

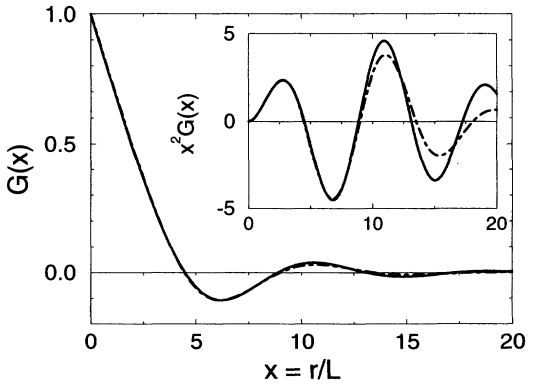

FIG. 5. Plot of the real-space correlation function $C(r,t)$ from the 3D CDS simulations (dash line) vs the result using the Gaussian closure (solid line). The inset shows $(r/L)^2C(r,t)$ for the same range of r/L. The fit is very good up to the second zero in the correlation function.

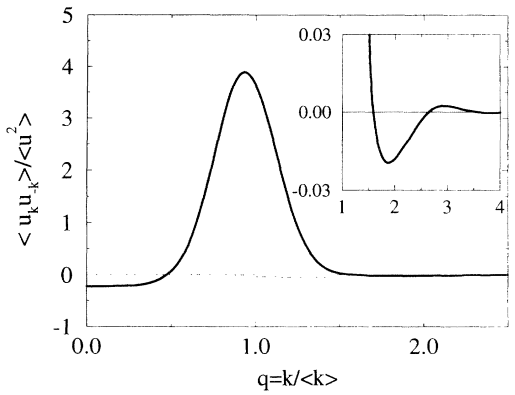

FIG. 6. Plot of the spectral density $\langle u_k(t)u_{-k}(t)\rangle$ from the $\langle \psi_k(t)\psi_{-k}(t)\rangle$ obtained under the Gaussian assumption from the 3D spinodal decomposition simulation (dashed line). The inset is a magnification of the spectral density for values of q just above the peak. There is a violation of positivity both for $q < 0.5$ and $1.5 < q < 4$, indicating that the Gaussian assumption is invalid for the conserved-order-parameter case.

of phase-ordering dynamics with conservation of order parameter. In addition, as shown in the inset of Fig. 6, there is also a violation at positivity in the very important range of q from approximately 1.5 to 4. This corresponds to the structure at wave numbers just above that of the peak of the scattering intensity.

To summarize, the interface approach is readily extendable to the conserved-order-parameter case. The interface dynamics, along with the assumption of scaling, leads to a growth exponent of 1/3. The resulting Gaussian closure leads to a very good fit of the correlation function for small and intermediates lengths. However, the Gaussian assumption cannot correctly describe the large length-scale behavior since, in order to reproduce the empirically observed results, the spectral density of the Gaussian field must be negative at small wave number. In this sense, a correct description of the large length-scale behavior of the correlation function and scattering intensity would require that the Gaussian field be unphysical.

V. DISCUSSION AND SUMMARY

We have discussed the reliability of the underlying Gaussian assumption in all the existing closure approximations used in phase ordering dynamics both with and without conservation laws. For the nonconserved case, we have clarified the relation between different approaches and demonstrated that the interface and bulk u approaches are exactly equivalent. We have demonstrated that the interface approach can be extended in an analogous manner to the conserved-order-parameter case.

For the nonconserved-order-parameter case one can explicitly obtain the equation for the underlying field u. We numerically integrate this equation and show that the single-point distribution function $P(u)$ is decays in a Gaussian manner at the tails but flatter near $u = 0$. Therefore although $P(u)$ is approximately Gaussian it has important corrections as the interface dynamics are controlled by the region near $u = 0$.

Using the arguments for the nonconserved-order-parameter case, we extend the interface approach to the conserved-order-parameter case. We show that this leads to an nontrivial fit of the correlation function $C(r,t)$ and scattering intensity $S_k(t)$ for small to moderate scaled distances $r/L(t)$. However, we find that the Gaussian approach is more fundamentally flawed in its description of the long distance correlations. Therefore the Gaussian approximation cannot be used to recover the entire empirically observed form factor.

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[1] 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 Press, New York, 1983), Vol. 8, p. 267.
[2] H. Furukawa, Adv. Phys. **34**, 703 (1985).
[3] K. Binder, in *Phase Transformations of Materials (Materials Science and Technology)* edited by P. Haasen (Springer-Verlag, Berlin, 1991), Vol. 5, p. 405.
[4] A. Coniglio and M. Zannetti, Europhys. Lett. **10**, 575 (1989); A. Coniglio and M. Zannetti, Physica A **163**, 325 (1990).
[5] T.J. Newman and A.J. Bray, J. Phys. A **23**, L279 (1990); T.J. Newman, A.J. Bray, and M.A. Moore, Phys. Rev. B **42**, 4514 (1990); A.J. Bray and K. Humayun, J. Phys. A **23**, 5897 (1990).
[6] A.J. Bray and K. Humayun, Phys. Rev. Lett. **68**, 1559 (1992).
[7] S.M. Allen and J.W. Cahn, Acta Metall. **27**, 1085 (1979).
[8] K. Kawasaki and T. Ohta, Prog. Theor. Phys. **67**, 147 (1982).
[9] J.S. Langer, Acta Metall. **21**, 1649 (1973).
[10] J.S. Langer, M. Bar-On, and H.D. Miller, Phys. Rev. A **11**, 1417 (1975).
[11] K. Kawasaki, M.C. Yalabik, and J.D. Gunton, Phys. Rev. A **17**, 455 (1978); K. Kawasaki, Physica A **161**, 161 (1992).
[12] T. Ohta, D. Jasnow, and K. Kawasaki, Phys. Rev. Lett. **49**, 1223 (1982).
[13] Y. Oono and S. Puri, Mod. Phys. Lett. B **2**, 861 (1988).
[14] G.F. Mazenko, Phys. Rev. B **42**, 4487 (1990).
[15] A.J. Bray and K. Humayun, Phys. Rev. E **48**, 1609 (1993).

[16] Y. Oono and S. Puri, Phys. Rev. Lett. **58**, 836 (1988); S. Puri and Y. Oono, Phys. Rev. A **38**, 1542 (1988).
[17] A. Shinozaki and Y. Oono, Phys. Rev. E **47**, 804 (1993).
[18] L. Bronsard and R.V. Kohn, J. Differential Equations **90**, 211 (1991).
[19] Y.-G. Chen, Y. Giga, and S. Goto, J. Diff. Geom. **33**, 749 (1991).
[20] L.C. Evans and J. Spruck, J. Diff. Geom. **33**, 635 (1991).
[21] The interface method works only in $d(>1)$ space. In one-space the interface equation trivially gives $v_n = 0$, that is, patterns do not change. The bulk approximation is applicable in any space and seems to contradict this conclusion. However, as long as the interface thickness is finite, even codimension-one patterns evolve in time, albeit slowly. Thus there is no inconsistency in the bulk approach. Of course, this does not mean that the one-space version is as reliable as the higher space versions.
[22] C. Yeung and D. Jasnow, Phys. Rev. A **42**, 10 523 (1990).
[23] C. Yeung and D. Jasnow, J. Phys. A **23**, L1309 (1990).
[24] F. Liu and G.F. Mazenko, Phys. Rev. B **44**, 9185 (1991).
[25] K. Humayun and A.J. Bray, Phys. Rev. B **46**, 10 594 (1992).
[26] G.F. Mazenko, Phys. Rev. Lett. **63** 1605 (1989).
[27] Y. Oono (unpublished).
[28] G.F. Mazenko, Phys. Rev. B **43**, 5747 (1991).
[29] T. Ohta and H. Nozaki, in *Space-Time Organization in Macromolecular Fluids*, edited by F. Tanaka, M. Doi, and T. Ohta, Springer Series in Chemical Physics, Vol. 51 (Springer, Berlin, 1989).
[30] K. Kawasaki and T. Ohta, Prog. Theor. Phys. **68**, 129 (1982); G. Caginalp, Phys. Rev. A **39**, 5887 (1989); R.L. Pego, Proc. R. Soc. London Ser. A **422**, 261 (1989).
[31] During the completion of this paper, we have received a paper by H. Tomita [Prog. Theor. Phys. **90**, 521 (1993)] in which he obtains a result similar to Eq. (4.7).
[32] A. Shinozaki and Y. Oono, Phys. Rev. Lett. **66**, 173 (1991); Phys. Rev. E **48**, 2622 (1993).
[33] C. Yeung, Phys. Rev. Lett. **61**, 1135 (1988).