Possibilities and Limitations of Gaussian Closure Approximation for Phase Ordering Dynamics.

Chuck Yeung$^{a,c}$, Y. Oono$^b$ and A. Shinozaki$^{b,c}$

$^a$ Department of Physics, University of Toronto, Toronto, Ontario M5S-1A7 CANADA
$^b$ Department of Physics, Materials Research Laboratory, and Beckman Institute, University of Illinois at Urbana-Champaign, Urbana, IL 61801 USA
$^c$ Department of Physics and Astronomy, University of Pittsburgh, Pittsburgh, PA 15260 USA

March 23, 2022

Abstract

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 gives different results is a sign of the breakdown of the Gaussian assumption. We concentrate on the non-conserved order parameter case but also touch on the conserved order parameter case. We demonstrate that the Gaussian assumption is fundamentally flawed in the latter case.

PACS 64.60.Cn, 64.75.+g, 64.70.Kb
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 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, 2, 3]. Analytic progress has been confined to the case of $O(n)$ component order-parameter in the limit of large $n$ [4, 5, 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) [1, 11, 12, 13, 14]. For the scalar order parameter case the simplest closure approximation is to assume that the order parameter field $\phi(r, t)$ is a Gaussian stochastic field [9]. This method fails, since, on the scale of the characteristic domain size $L(t)$, $\phi(r, t)$ is effectively discontinuous. Instead progress is achieved by assuming that there exists an underlying Gaussian field which is nonlinearly related to $\psi(r, t)$ [12, 13, 14].

In this paper, we will explore the reliability of closure approximations based on the assumption of an underlying Gaussian stochastic field. We first discuss phase ordering dynamics without conservation of order parameter. In particular we will discuss the limitations of this assumption and the relation between different implementations of the Gaussian closure. One method of introducing the underlying field is based on the dynamics of random interfaces. We will denote this scheme 'I'. It was first proposed by Ohta, Jasnow and Kawasaki (OJK) [12] for systems with non-conserved order parameter (NCOP). This method was later extended by Ohta and Nozaki [15] to systems with conserved order parameter (COP).

A second manner of introducing the underlying field is by writing the order parameter as $\psi(r, t) = f(u(r, t))$, where $f$ is the interface profile. We will call this method scheme ‘B’ since it relies on the full bulk dynamics. Oono and Puri (OP) [13] introduced this scheme to remove inconsistencies in the original OJK argument. There are two manners in which the bulk closure scheme has been applied. The first (scheme Bu) obtains a closed equation for the correlation function of $u(r, t)$. Oono and Puri demonstrated that this provides reasonable results for the NCOP case [13]. The second (scheme B$\psi$) constructs a closed equation for the correlation function of $\psi(r, t)$ using the assumed Gaussian nature of the underlying field $u(r, t)$. Oono applied the B$\psi$ closure to the COP case [14] but found that it did not yield the experimentally observed scaling, $L(t) \sim t^{1/3}$. Mazenko has applied the B$\psi$ closure to both the NCOP [14] and COP cases [17, 18]. For the NCOP case, Mazenko found the B$\psi$ closure gives different results from Ohta-Jasnow-Kawasaki and Oono-Puri but is also in reasonable agreement with experiment [14]. For the COP case, Mazenko found that $L(t) \sim t^{1/4}$ [17] and included an ad-hoc term to enforce $L(t) \sim t^{1/3}$ behavior. The modified version still contains unphysical features [18] such as the violation of the Tomita sum rule [19, 20].
In this paper we will concentrate on phase ordering dynamics without conservation of
order parameter but we will also discuss the case in which the order parameter is conserved.
For the non-conserved order parameter case, the closure schemes reproduce many features
observed in domain growth, such as dynamical scaling and predictions for the growth of \( L(t) \),
the structure factor \([12, 13, 14]\) and the decay of two-time correlations \([21, 22, 23]\). Variations
of these models have been used to study finite-size effects \([24]\), finite temperature effects
\([13, 25]\), effects of long-range initial conditions \([26]\), systems with long-range interactions
\([27]\), and systems with a higher component order parameter \([28, 29, 30]\). However, the
predictions of the different closure schemes do not always coincide \([31]\).

In Section II, we derive a modified form of the OJK result starting with the interface
approach (scheme I) correcting for inconsistencies in the original argument \([12]\). In Section
III, we discuss the bulk \( u \)-closure (scheme \( B_u \)) for the NCOP case and demonstrate that
asymptotically it provides the same result as the interface approach (scheme I). This was
originally suggested by Oono and Puri using a heuristic argument \([13]\). We next discuss the
bulk \( \psi \)-closure (scheme \( B_\psi \)). As shown by Liu and Mazenko \([31]\), this closure scheme
leads to different predictions from the interface approach. We will discuss each approach in
a parallel manner to emphasize the relation between the approaches. We will show that the
\( Bu \) and \( B_\psi \) closures uses the same approximation so that their different predictions signify
the breakdown of that approximation. In Section IV we study the approximate Gaussian
nature of \( u(r,t) \) through numerical simulations. We find that the single point probability
distribution function \( P(u) \) decays as a Gaussian at the tails but decays slower than Gaussian
near \( u = 0 \). In Section V we present our findings for the conserved order parameter case.
We find that, in this case, the Gaussian closure is fundamentally flawed. In Section VI we
summarize our findings.

II Interface Approach

In this section, we derive a modified form of the Ohta-Jasnow-Kawasaki and Oono-Puri
results demonstrating the features of the interface scheme. We assume the dynamics are
described by 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 \( \xi \) is the interfacial width, \( \psi(r,t) \) is the scalar order parameter and \( \mu_B(\psi) \) is the
portion of the local chemical potential which contains no gradient terms. We assume \( \mu_B \)
is an odd function of \( \psi \), the equilibrium values of \( \psi \) are \( \pm 1 \); and there exists a solution,
\( \psi = f(z) \), corresponding to a stationary planar interface at \( z = 0 \), i.e., \( f(z) \) obeys

\[
0 = -\mu_B(f(z)) + \frac{\xi^2}{2} \frac{d^2 f}{dz^2}.
\]

For example, if \( \mu_B = -\psi + \psi^3 \) then \( f(z) = \text{tanh}(z/\xi) \). However, other than the requirement
that \( f(z) \) increases monotonically from \(-1 \) to \( 1 \) over a length-scale \( \xi \), the exact form \( f(z) \) is
unimportant [13, 14]. This universality is closely related to the universality in the dispersion relation around the interface [13].

At late times, the width $\xi$ is small relative to the characteristic domain size $L(t)$, and the domain growth is determined by the motion of the sharp interfaces. The interface dynamics can be derived from the TDGL [7, 8, 34],

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

(2.2)

Here the normal velocity of the interface, $v_n$, is defined as positive when the ‘minus’ phase moves into the ‘plus’ phase, the normal $\hat{n}$ points into the ‘plus’ phase; and the local curvature, $\kappa = \nabla \cdot \hat{n}$, is positive for a bump of the ‘minus’ phase into the ‘plus’ phase.

The essence of the interface method is to rewrite the interfacial equation in terms of an indicator field $u(r, t)$. The indicator field is defined so that $u > 0$ ($u < 0$) in the plus (minus) phase, $u = 0$ at the interface, and, near the interface, $|u|$ is the distance to the interface, so that, $\nabla u(r, t) = \hat{n}(r, t)$ near $u = 0$ [12, 13]. The main motivation for introducing $u(r, t)$ is that it remain continuous on all length-scales, while $\psi(r, t)$ is effectively discontinuous upon rescaling distances by $L(t)$. Therefore simple decoupling approximations should be more trustworthy for the indicator field $u$ than for order parameter field $\psi$.

In terms of $u$ the interfacial dynamics (Eq. (2.2)) is

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

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

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

(2.4)

The function $\tilde{P}$ has the following properties: $\tilde{P}(0, \nabla u) = 0$, since Eq. (2.3) must be recovered at $u = 0$. Due to the symmetry of the TDGL, $\tilde{P}(u, \nabla u)$ 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, $\tilde{P}(u, \nabla u)$ is assumed to be local, i.e., $\tilde{P}(u, \nabla u)$ depends only on a finite number of derivatives of $u$. Using heuristic arguments, Oono and Puri replaced $P(u, \nabla u)$ by a function of time chosen to maintain the equilibrium interface thickness [13]. In the following section, we demonstrate that $P(u, \nabla u)$ can be obtained exactly from the TDGL using the bulk $u$-closure (Bu approach) and therefore schemes I and Bu are asymptotically equivalent for the non-conserved order parameter case.

From Eq. (2.4), the two-point correlation function, $\langle u_1 u_2 \rangle$, obeys

$$\frac{\partial \langle u_1 u_2 \rangle}{\partial t_1} = \frac{\xi^2}{2} \left( \nabla_1^2 \langle u_1 u_2 \rangle + \langle P(u_1, \nabla_1 u_1) u_2 \rangle \right),$$

(2.5)
where \( u_i = u(\mathbf{r}_i, t_i) \) and \( t_1 \neq t_2 \). Note that the local constraint \(|\nabla u| = 1\) at the interface forces \( \langle u_1^2 \rangle \) to grow as \( L_i^2 \), where \( L_i = L(t_i) \).

We now need to make an assumption concerning the statistics of \( \{ u(\mathbf{r}, t) \} \). The simplest assumption is that \( u \) is a Gaussian stochastic field. As noted previously, the rationale is that \( u \) is ‘equicontinuous’ for all length scales, so simple decoupling scheme may be more trustworthy for it than for the original order parameter field.

For any Euclidean symmetric Gaussian stochastic field, \( \langle P(u_1, \nabla u_1) u_1 u_2 \rangle \) has a simple form

\[
\langle P(u_1, \nabla u_1) u_1 u_2 \rangle = p(\langle u_1^2 \rangle, \nabla^2 \langle u_1 u_1 \rangle) \langle u_1 u_2 \rangle,
\]

(2.6)

where \( \nabla^2 \langle u_1 u_1 \rangle = \lim_{r \to r_1} \nabla^2 \langle u(\mathbf{r}_1, t_1) u(\mathbf{r}', t_1) \rangle \). Note that the function \( p \) does not depend on \( t_2 \) or \( \mathbf{r}_1 - \mathbf{r}_2 \) so we can replace \( p \) as a function of \( t_1 \):

\[
\langle P(u_1, \nabla u_1) u_1 u_2 \rangle = p(t_1) \langle u_1 u_2 \rangle.
\]

(2.7)

For \( t_1 \neq t_2 \), Eq. (2.3) becomes

\[
\frac{\partial \langle u_1 u_2 \rangle}{\partial t_1} = \frac{\xi^2}{2} \left( \nabla^2 \langle u_1 u_2 \rangle + p(t_1) \langle u_1 u_2 \rangle \right).
\]

(2.8)

For \( t_1 = t_2 = t \), we have

\[
\frac{\partial g(r,t)}{\partial t} = \xi^2 \left( \nabla^2 g(r,t) + p(t) g(r,t) \right),
\]

(2.9)

where \( g(r,t) \equiv \langle u(\mathbf{r}, t) u(0,t) \rangle \). The Gaussian approximation means that the detailed local constraint \(|\nabla u| = 1\) is no longer met but is replaced by a global constraint \( \langle u^2 \rangle \sim L^2 \). This requires that \( \xi^2 p(t) = (d + 2)/(2t) \), where \( d \) is the spatial dimension. This is exactly the form of \( p(t) \) chosen in [13] so that the interface thickness is time independent.

The final step is to obtain the correlation function of \( \psi(\mathbf{r}, t) \) from \( u(\mathbf{r}, t) \). Near the interface \( \psi \) will be close to the planar interfacial profile so that \( \psi(\mathbf{r}, t) = f(u(\mathbf{r}, t)) \). At long times, the exact form of \( f \) should be irrelevant, (as long as \( f \) approaches \( \text{sgn}(u) \) as \( \xi \to 0 \)) [20]. Oono and Puri chose a form that simplifies the Gaussian integrals,

\[
f(u) = \text{sgn}(u) \ast \frac{1}{(2\pi \xi^2)^{1/2}} \exp \left( -\frac{u^2}{2\xi^2} \right)
\]

(2.10)

where \( \ast \) is the convolution. They find

\[
C_{12}(r) = \frac{2}{\pi} \arcsin \left( \frac{\langle u_1 u_2 \rangle}{(\langle u_1^2 \rangle + \xi^2)^{1/2}(\langle u_2^2 \rangle + \xi^2)^{1/2}} \right),
\]

(2.11)

where \( C_{12}(r) \equiv \langle \psi_1 \psi_2 \rangle \) and \( r = |\mathbf{r}_1 - \mathbf{r}_2| \). For any reasonable form of \( f(u) \), a WKB-type argument can be used to show that, \( C_{12}(r) \) approaches Eq. (2.11) when \( \langle u_1^2 \rangle \gg \xi^2 \).

In the original OJK analysis (as well as OP) there is a factor \( (d-1)/d \) in front of the diffusive term in the evolution equation for \( u \). This difference is due to the lack of
local constraint, $|\nabla u| = 1$, in OJK. (For OP, this is due to both its eclectic and logically opaque nature and the lack of this exact constraint.) With this exception, Eq. (2.4) with $p(t) = (d + 2)/(2\xi^2 t)$ together with Eq. (2.11) is the main result in Ref. [13]. The result of OJK can be obtained from this approach asymptotically without the difficulties in the original argument. In the original OJK derivation $u$ is not self-consistently defined, since $f$ was chosen to be $\text{sgn}(u)$ and $|\nabla u| = 1$ was not even approximately enforced [12, 13].

The interface approach predicts the following [12, 13]: (1) dynamical scaling with $L \sim t^{1/2}$, (2) the equal-time correlation function, $C(r,t)$ decays as a Gaussian at large $r$, (3) the scattering function, $S_k$, displays Porod’s law, $S_k \sim k^{-(d+1)}$ [33, 18], for the wavevectors $k$ in the range $L^{-1} \ll k \ll \xi^{-1}$ and (4) Tomita’s sum rule is obeyed [15], i.e., in the scaling limit, $\partial^m C/\partial x^m \big|_{x=r/L \to 0^+} = 0$ for any even $m$. For the two-time behavior, the interface approach predicts $C_{12}(0) \sim (t_1/t_2)^{-d/2}$ for $t_1/t_2 \ll 1$ and the autocorrelation function of the Fourier transforms of $\psi$ decays as a stretched exponential, $\ln\langle \psi_k(t_1)\psi_{-k}(t_2) \rangle \sim -(t_2/t_1)^{1/2}$, for $L_2 \gg L_1$ [22].

An alternative closure method is the one due to Kawasaki, Yalabik and Gunton [11]. This method tries to sum a diverging series whose $n$-th term is of order $\exp(n\gamma t)$ with $\gamma t \gg 1$. Although the result is finite and the scaling results are the same as that of Eq. (2.9), this diverging behavior causes the ratio $\xi/L(t)$ to vanish exponentially fast, an absurdity caused perhaps by the strongly diverging nature of the series.

III Bulk Approach

III.A Bulk $u$-closure (Scheme Bu)

In this subsection, we discuss the bulk $u$-closure (Bu). We demonstrate that the results of the interface approach can be obtained without the intermediate step of the interface description.

We introduce an auxiliary field $u(r,t)$ by

$$\psi(r,t) = f(u(r,t)).$$

(3.1)

The planar interface solution, $f$, obeys

$$\mu_B(f) = \frac{\xi^2 d^2 f}{2 du^2}.$$ 

At late stages, the interfacial profiles will be very close to the planar profile. Therefore this nonlinear mapping is exactly of the form required to enforce $u = 0$ at the interface and $\nabla u = \hat{n}$ near the interface.

In terms of $f(u)$, the TDGL is

$$\frac{\partial f}{\partial t} = -\frac{\xi^2}{2} \left[\frac{d^2 f}{du^2} - \nabla^2 f \right].$$

(3.2)
Applying the chain rule for \( \frac{df}{dt} \) and \( \nabla^2 f \), we obtain an expression for \( \partial u / \partial t \),

\[
\frac{\partial u}{\partial t} = \frac{\xi^2}{2} \left[ \nabla^2 u + \left( 1 - |\nabla u|^2 \right) Q(u) \right], \tag{3.3}
\]

where \( Q(u) \equiv -(df/du)^{-1} \frac{d^2 f}{du^2} \). For \( \mu_B = -\psi + \psi^3 \) we find \( f(u) = \tanh(u/\xi) \) and \( Q(u) = 2f(u) \). In general, \( Q(0) = 0 \), \( Q(u) \) is an odd function of \( u \) and \( Q(u) \) approaches \( d\mu_B/d\psi \) (a finite constant) as \( u \to \infty \). The result is that \( Q(u) \) must be proportional to \( \text{sgn}(u) \) in the limit \( \xi \to 0 \).

Equation (3.3) is exactly the form required to extend the interface equation into the bulk using the interface approach (Eq. (2.4)) of the previous section. We can now identify the previously unknown ‘Lagrange multiplier’ function, \( \tilde{P}(u, \nabla u) \) in Eq. (2.4), with \( (1 - |\nabla u|^2) Q(u) \). We repeat the steps of the interface approach with the difference that we now have an explicit expression for \( \tilde{P}(u, \nabla u) \).

From Eq. (2.4) we obtain the expression for the correlation function \( \langle u_1 u_2 \rangle \),

\[
\frac{\partial \langle u_1 u_2 \rangle}{\partial t} = \xi^2 \left[ \nabla^2 \langle u_1 u_2 \rangle + \langle (1 - |\nabla u_1|^2)Q(u_1)u_2 \rangle \right], \tag{3.4}
\]

where we restrict the discussion to \( t_1 = t_2 = t \). The more general case is easily obtained.

To this point there are no approximations. To proceed further we again assume that \( \{u(\mathbf{r}, t)\} \) is a Gaussian stochastic field. Just as in scheme I, this assumption is totally uncontrolled. In the same spirit as before, we assume that explicit forms of \( f \) and \( Q \) are unimportant as long as they approach \( \text{sgn}(u) \) and \( 2\text{sgn}(u) \) in the limit of \( \xi \to 0 \). Therefore we choose a form of \( Q \) which simplifies the Gaussian integrals,

\[
Q(u) = \frac{i}{\pi} \int_{-\infty}^{+\infty} d\omega \frac{1}{\omega} \exp \left( -\frac{\xi^2 \omega^2}{2} - i\omega u \right). \tag{3.5}
\]

That is, \( Q(u) \) is the sign function mollified by a Gaussian function. Equation (3.4) can now be computed explicitly as

\[
\frac{\partial g(\mathbf{r}, t)}{\partial t} = \xi^2 \left[ \nabla^2 g(\mathbf{r}, t) + \frac{1 + \nabla^2 g(0, t)}{\sqrt{\pi(\xi^2 + g(0, t))}/2} g(\mathbf{r}, t) \right], \tag{3.6}
\]

where, as in the previous section, \( g(\mathbf{r}, t) = \langle u_1 u_2 \rangle \) for \( t_1 = t_2 = t \). That is, we have an equation exactly the same form as Eq. (2.4)

\[
\frac{\partial g(\mathbf{r}, t)}{\partial t} = \xi^2 \left[ \nabla^2 g(\mathbf{r}, t) + p(t) g(\mathbf{r}, t) \right], \tag{3.7}
\]

where

\[
p(t) = \frac{1 + \nabla^2 g(0, t)}{\sqrt{\pi(\xi^2 + g(0, t))}/2} \tag{3.8}
\]

is now an explicitly known function of \( t \).
We are left with a closed equation for \( g(r, t) \) which can be solved as an initial value problem. However, it is more instructive to consider the scaling limit. We define the characteristic length-scale by \( L^2 \equiv \frac{g(0, t)}{\sqrt{-\partial^2 g(r, t)/\partial r^2}|_{r \rightarrow 0}} \) so that \( \nabla^2 g(0, t) = -\frac{dg(0, t)}{L^2} \).

With this definition we can show that \( L^2 \approx 2 \xi^2 t \). Substituting this in Eq. (3.7) we find

\[
\frac{dg(0, t)}{dt} = -\frac{d}{2t} g(0, t) + \xi^2 p(t) g(0, t),
\]

with \( p \) now given by

\[
\xi^2 p(t) = \frac{\xi^2 - \frac{dg(0, t)}{2t}}{\sqrt{\pi(\xi^2 + g(0, t))}/2}.
\]

The only meaningful asymptotic solution of this equation is \( g(0, t) \approx 2 \xi^2 t/d + O(1) \). This implies that asymptotically \( \xi^2 p(t) = (d + 2)/(2t) \), i.e., the same \( p(t) \) as that given by the interface approach. We also find that this is the asymptotic form if we solve the initial value problem using the explicit expression (Eq. (3.8)) for \( p(t) \).

Therefore we have demonstrated that the results of the interface approach can be obtained directly from the time-dependent Ginzburg-Landau equation without explicitly using the interface description. In particular we have shown that the previously unknown functions, whose properties were obtained by scaling arguments and intuitive physical requirements, can be obtained more directly and, furthermore, is exactly the form required by the scaling arguments.

### III.B Bulk \( \psi \)-closure (Scheme B\( \psi \))

In the bulk \( \psi \)-closure the auxiliary field \( u(r, t) \) is introduced in the same way as in the bulk \( u \)-closure, i.e., \( \psi(r, t) = f(u(r, t)) \), where \( f \) is the planar interfacial profile. A dynamical equation is then obtained directly for \( \langle \psi(r_1, t_1) \psi(r_2, t_2) \rangle \) under the assumption that \( \{u(r, t)\} \) is a Gaussian random field. Mazenko applied this scheme to the non-conserved order parameter case [14]. We summarize the argument below.

The TDGL equation in terms of \( \psi = f(u) \) is given by Eq. (3.2),

\[
\frac{\partial f}{\partial t} = -\frac{\xi^2}{2} \left( \frac{d^2 f}{du^2} - \nabla^2 f \right).
\]

The expression for the correlation function \( \langle \psi_1 \psi_2 \rangle = \langle f_1 f_2 \rangle \) becomes

\[
\frac{\partial \langle f_1 f_2 \rangle}{\partial t} = \xi^2 \left( \langle \frac{d^2 f_1}{du_1^2} f_2 \rangle - \nabla^2 \langle f_1 f_2 \rangle \right),
\]

where \( f_i = f(u(r_i, t_i)) \). Here, we again let \( t_1 = t_2 = t \) for simplicity. An analogous equation can be written for \( t_1 \neq t_2 \).

To this point there has been no approximations. To proceed further, \( u \) is again assume to be a Gaussian stochastic field. Using the properties of Gaussian variables, one finds
\[(d^2 f_1/du_1^2)u_2 = \partial(f_1f_2)/\partial(u^2)\]. This gives a closed equation for \(C(r, t) = \langle \psi(r, t)\psi(0, t) \rangle\). The equation can then be solved numerically as an initial value problem. For later times, the exact form of \(f\) is irrelevant and we can use the \(f\) given in Eq. (2.10). Performing the Gaussian integrals, we find, \(\partial C/\partial\langle u^2 \rangle = -2\tan(\pi C/2)/\pi(\langle u^2 \rangle + \xi^2)\), and
\[
\frac{1}{\xi^2} \frac{\partial C(r, t)}{\partial t} = \frac{2}{\pi(\langle u^2 \rangle + \xi^2)} \tan \left( \frac{\pi C(r, t)}{2} \right) + \nabla^2 C(r, t). \tag{3.13}
\]

For \(\langle u^2 \rangle \gg \xi^2\) a scaling form can be assumed. A nonlinear eigenvalue problem is then found which can be solved numerically to obtain the correlation function \(C(r, t)\) [4].

This closure yields [4]: (1) dynamical scaling with \(L \sim t^{1/2}\), (2) that the quasi-elastic scattering intensity obeys Porod’s law at large \(k\), (3) that Tomita’s sum rule is obeyed [19], and (4) that the real space correlation function decays slightly slower than a Gaussian with \(C(x) \sim x^a \exp(-bx^2)\) where \(a > 0\) depends on \(\lambda\) and, hence, \(d\). For the two-time behavior, Liu and Mazenko predicts that \(C_{12}(0) \sim (L_1/L_2)^b\) for \(L_1 \ll L_2\) where \(b \approx 1.28\) for \(d = 2\) and \(b = 1.63\) for \(d = 3\) [23]. This closure also predicts that the auto-correlation function for the Fourier transforms will decay exponentially in \(t_2/t_1\) for \(t_2 \gg t_1\) [23].

### III.C The relation between the different approaches

From the above discussion the bulk \(u\)-closure and interface approaches are equivalent. We also see that the \(Bu\)-closure and the \(B\psi\)-closure uses exactly the same assumption: \(\psi = f(u)\) with \(u\) assumed to be Gaussian. However, we also see that there are discrepancies in the predictions of the two approaches. These discrepancies can be regarded as an indicator of the limit of the reliability of the Gaussian assumption. Experimentally [30] there is general consensus with regards to the features in which both approaches agree, i.e., \(L \sim t^{1/2}\), Porod’s law [35], and Tomita’s sum rule [19], although these are kinematic consequences.

To investigate the discrepancies between the different models, one must rely on numerical studies, either kinetic Ising model simulations with Glauber dynamics or cell dynamical schemes (CDS) corresponding to the time-dependent Ginzburg-Landau equation [32, 37]. Humayun and Bray [26], and Shinozaki [38] find that the equal time correlation function \(C(x)\) obtained by a CDS is better fitted by the OJK result, although, \(C(x)\) decays faster than both predictions. Liu and Mazenko studied the autocorrelation function \(C_{12}(0)\) and found that the decay exponent \(b\) is approximately 1.25 for \(d = 2\) and for \(d = 3\), preliminary results indicate \(b \approx 1.8\), i.e., the decay is faster than the OJK prediction of \(d/2\) and more in agreement with the prediction of the \(B\psi\) closure. On the other hand for smaller ratios of times \(t_2/t_1 \leq 10\), the OJK result seems to give a very good zero parameter fit to \(C_{12}(0)\) [21]. The numerical data for the autocorrelation of the Fourier transform \(\psi_k\) [29] has not yet been sufficient to compare the results of the two closures. In this case the bulk \(u\)-closure predicts that \(\langle \psi_k(t_1)\psi_{-k}(t_2) \rangle\) decays as a stretched exponential for \(t_2 \gg t_1\) [24] while the bulk \(\psi\)-closure predicts an exponential decay [23]. Nevertheless for many features, the Gaussian closures give reasonable agreement with experiment. A broad statement would be that, while \(B\psi\) gives better fits to the two-time behavior, schemes \(Bu\) and, hence, the scheme I, gives better fits to the equal time behavior.
The relation between the closures is further clarified in the \( d \to \infty \) limit. It was shown by Liu and Mazenko [31] that the scaling functions obtained from OJK and the \( B\psi \) closure coincide in the limit of \( d \to \infty \). Here we show more explicitly that the two closures are equivalent in this limit. In the limit of \( t \to \infty \), \( \langle \psi_1 \psi_2 \rangle \) depends only on the ratio \( \alpha(r,t) = g(r,t)/g(0,t) \) for \( t = t_1 = t_2 \). The evolution equation for \( \alpha(r,t) \) in the \( B\psi \) closure can be obtained from Eq. (3.6). In the limit of large \( t \) this becomes
\[
\frac{\partial \alpha(r,t)}{\partial t} = \xi^2 \left( \frac{\alpha(r,t)}{\langle u^2 \rangle} + \nabla^2 \alpha(r,t) \right) \tag{3.14}
\]
For the \( B\psi \) closure we can obtain the dynamical expression for \( \alpha(r,t) \) from Eq. (3.13),
\[
\frac{\partial \alpha(r,t)}{\partial t} = \xi^2 \left( \frac{\alpha(r,t)}{\langle u^2 \rangle} + \nabla^2 \alpha(r,t) + \alpha(r,t) |\nabla \alpha(r,t)|^2 \right). \tag{3.15}
\]
We can examine the \( d \to \infty \) limit by rescaling lengths by \( x = r/\sqrt{d} \). Eq. (3.14) becomes
\[
\frac{\partial \alpha(x,t)}{\partial t} = \xi^2 \left( \frac{\alpha(x,t)}{\langle u^2 \rangle} + \frac{1}{d} \frac{\partial^2 \alpha(x,t)}{\partial x^2} + \frac{d-1}{d} \frac{\partial \alpha(x,t)}{\partial x} \right). \tag{3.16}
\]
While Eq. (3.13) becomes
\[
\frac{\partial \alpha(x,t)}{\partial t} = \xi^2 \left( \frac{\alpha(x,t)}{\langle u^2 \rangle} + \frac{1}{d} \frac{\partial^2 \alpha(x,t)}{\partial x^2} + \frac{d-1}{d} \frac{\partial \alpha(x,t)}{\partial x} \right)^2. \tag{3.17}
\]
Taking the limit of \( d \to \infty \), we find that the equations for \( \alpha(r,t) \) become identical. Although this is not a proof, it does indicate that the Gaussian approximation may be valid in the limit of \( d \to \infty \).

IV Numerical Results

In the previous discussion we have shown that the \( \{u(r,t)\} \) 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 \( \{u(r,t)\} \) directly through a numerical updating of Eq. (3.6). As noted previously, no approximations are needed to proceed from the TDGL (Eq. (2.1)) to Eq. (3.3) 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 \( \text{sgn}(u) \) in the limit of \( L(t) \gg 1 \). We discretize the system with mesh size \( \delta x = 1.0 \) and time steps \( \delta t = .05 \). To reduce lattice effects we used a sphericalized Laplacian as described in Ref. [32]. For these large time and space steps the update corresponds to a CDS [34]. The simulation was performed on \( 800 \times 800 \) lattices with periodic boundary conditions. and repeated on \( 400 \times 400 \) lattices to check for finite size effects. The results for \( n = 400 \) and \( n = 800 \) begins 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 $u = 0$ 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. $u^2/\langle u^2 \rangle$. 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 Figure 1 plus the point at $t = 0$. The flatness is 3 at $t = 0$ since the initial distribution is Gaussian. For larger times we find that flatness is somewhat less than that expected for a Gaussian distribution.

From our numerical result we find that 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 $\{u(r,t)\}$ we must look also at the two-point and two-time distributions.

V Conserved order parameter case

The bulk $\psi$ closure (B$\psi$-scheme) approach was originally introduced to study systems with conserved order parameter (spinodal decomposition). Here we summarize our study of the conserved order parameter case emphasizing the difficulties of this approach.

The first important difficulty is that one cannot simply use the nonlinear mapping $\psi(r,t) = f(u(r,t))$ to define the indicator field. This is because the condition that $|\psi(r,t)| < \psi_{eq}$ is not met so that the mapping is not invertible. This is easily seen since, for the conserved case, local equilibrium near the interface requires that the deviation of $\psi$ from its planar interface value is proportional to the local curvature. However an indicator field can still be introduced using

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

where the $\phi$ field accounts for the deviation from the planar interfacial profile and the $u$ field has the same properties as in the nonconserved case. One can then make the Gaussian assumption for the $u$ field with the $\phi$ field coupled in such a way as to enforce the $L(t) \sim t^{1/3}$ growth

However, even with this extension we find that the Gaussian assumption is incompatible with conserved order parameter dynamics. Since the extra field $\phi$ is of order $1/L$ we can neglect its direct effects in the correlation function in the scaling limit. We note that Eq.
the relation between $\langle \psi \psi \rangle$ and $\langle uu \rangle$,

$$C(r, t) = \frac{2}{\pi} \arcsin \left( \frac{\langle u_1 u_2 \rangle}{\langle u^2 \rangle} \right),$$

is independent of the dynamics and is true as long as $u(r, t)$ is a Gaussian stochastic field. 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 an empirically obtained $C(r, t)$. Figure 5 shows the spectral density $\langle u_k(t)u_{-k}(t) \rangle$ obtained in this manner using $C(r, t)$ from a very accurate three dimensional CDS simulation of spinodal decomposition [40]. We find that the spectral density $\langle u_k(t)u_{-k}(t) \rangle$ becomes significantly negative at small wavenumbers $q = kL(t) < 0.5$. (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 can say that the Gaussian assumption is not a reasonable description of the large length-scale behavior. In addition we find that (inset of Figure 5) 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 wavenumbers just above that of the peak of the scattering intensity. We can conclude that the bulk closure approaches are inherently flawed for the conserved order parameter case.

Since interface approaches also use the Gaussian assumption and the same relation Eq. (2.11), they are also flawed. However, Ohta and Nozaki’s modest success justifies a more systematic study of the interface approach. This is especially the case since the Ohta-Nozaki approach fixes the growth exponent to be 1/3 by an intuitive but rather ad hoc manner [15]. Here we briefly summarize our interface attempt and its limitation.

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

$$G \ast \delta(u) \partial_t u = \nabla^2 u,$$  \hspace{1cm} (5.1)

where $G$ is the Laplacian Green’s function, i.e., $G = -\nabla^{-2}$ [11]. As in the non-conserved case, this equation is correct only at the interface. We assume the following bulk extension of the interface equation:

$$G \ast \delta(u) \partial_t u = J\nabla^2 u + Q,$$  \hspace{1cm} (5.2)

where $J$ is a function of $u$ such that $J = 1$ at the interface, and $Q$ is a functional of $u$ and its derivatives up to the second order with $Q = 0$ at the interface. The extension is motivated by an analogous idea behind Eq. (2.4). Applying the Laplacian to Eq. (5.2), and assuming that $u$ is a Gaussian stochastic field, we find (after some algebra)

$$\frac{1}{2} \frac{1}{\sqrt{2\pi g(0, t)}} \partial_t g(r, t) = -R(t)\nabla^4 g(r, t) - \nabla^2 Q(t)g(r, t) + P(t),$$  \hspace{1cm} (5.3)

where $g(r, t) = \langle u(r, t)u(0, t) \rangle$ and $P, Q$ and $R$ are yet unspecified functions of time only. 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. From the definition of $u$ we require $\langle u^2 \rangle \sim L^2$ for large times, so that, in the scaling limit, $g(r, t)$ must be of the form

$$g(r, t) = L(t)^2 g(r/L(t)),$$  \hspace{1cm} (5.4)
where \( g(0) = 1 \). Rescaling \( x = r/L(t) \), we get Eq. (5.3) with Eq. (5.4) as

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

where \( \lambda = L'(t) L(t)^2 / 2\sqrt{2\pi} \) which must be time-independent asymptotically. The coefficients \( R, L^2 Q \) and \( L^4 P \) must converge to a non-zero constant in the \( t \to \infty \) limit; if they diverge, we get physically absurd results, while the same is true if these coefficients vanish. Hence, asymptotically Eq. (5.5) becomes

\[
\lambda (2g(x) - x \frac{dg(x)}{dx}) + \nabla^4 g(x) + A \nabla^2 g(x) - B g(x) = 0,
\]

where we have rescaled \( \lambda \) to get rid of the numerical coefficient in front of the double Laplacian, and \( A \) and \( B \) are constant. For \( g(0) \) to be finite, \( B = (2 + d) \lambda \) is required. Hence, in the \( k \)-space, we arrive at

\[
\lambda \frac{dg}{dk} = (-k^3 + k)g,
\]

where \( A \) is scaled out, which must be positive and \( \lambda \) remains an unspecified constant. The resultant equation is very similar to the Ohta-Nozaki equation, and has the same defect, although in our case \( L \sim t^{1/3} \) follows from our starting point of the interface equation.

Given these caveats we fit the empirically obtained \( C(r, t) \), using the \( g(r, t) \) obeying Eq. (5.7). We find that \( \lambda = 0.013 \) gives the best fit to \( C(r) \). Figure 6 shows that the fit is very good up to approximately the second zero of \( C(r) \). This is further shown in the inset in which \( (r/L)^2 C(r) \) is plotted to show that goodness of the fit is not simply because \( C(r) \) is small. As noted previously, the Gaussian assumption is invalid for long length scales and the fit becomes increasingly worse for larger \( x \). The same holds for the scattering intensity. We find a very good fit for \( q > 0.5 \) but the conservation law is violated due to the invalidity of the Gaussian field assumption at small \( k \).

To conclude, we find that the Gaussian assumption can give a nontrivial fit to a limited range of length-scales for the correlation function and wavenumbers for the scattering intensity. However, given the fundamental flaw in the Gaussian assumption at larger distances, we feel that there is no point in making a more concerted effort.

VI Discussion and Summary

We have discussed the relation between different closure approximations for phase ordering without conservation of order parameter. These closure approximations are based on the assumption of an underlying Gaussian stochastic field \( u(r, t) \). We discuss two general methods, the interface approach in which a dynamical equation for \( u(r, t) \) is obtained from the interfacial dynamics and the bulk approaches in which the \( u(r, t) \) is defined by the nonlinear relation \( \psi(r, t) = f(u(r, t)) \), where \( f \) is the planar interfacial profile. The bulk approach is
further subdivided into the bulk $u$-closure and bulk $\psi$-closure in which dynamical equations are obtained for the correlation function of $u$ and $\psi$, respectively.[13]

We have derived a modified form of the original interface approach [12] and shown that it is completely equivalent to the bulk $u$-closure. We then have demonstrated that the only assumptions of the bulk $\psi$-closure and bulk $u$-closure is that $\{u(r,t)\}$ is a Gaussian stochastic field. Our conclusion is that the discrepancies in the predictions of the bulk $u$-closure and bulk $\psi$-closure is due to a breakdown of that approximation.

We have also discussed the Gaussian closure for the conserved case. We have shown that the Gaussian approximation is more fundamentally flawed in this case. However, the interface closure approach still leads to a nontrivial fit of the correlation function $C(r,t)$ and scattering intensities $S_k(t)$ except at large scaled distances $r/L(t)$ or at small scaled wavenumbers $kL(t)$.

Acknowledgements

We would like to thank Prof. David Jasnow and Dr. Timothy Rogers for helpful discussions. C.Y. is grateful to the National Science Foundation for support under grant DMR 89-14621 and Natural Sciences and Engineering Research Council of Canada. Y.O. and A.S. gratefully acknowledge the support of the National Science Foundation under grant DMR 90-15791.

References

[1] J.D. Gunton, M. san Miguel and P.S. Sahni, in Phase Transitions and Critical Phenomena Vol. 8, edited by C. Domb and J.L. Lebowitz, pp. 267, (Academic Press, New York, 1983).

[2] H. Furukawa, Adv. Phys. 34, 703 (1985).

[3] K. Binder, in Phase Transformations of Materials (Materials Science and Technology) Vol. 5, edited by P. Haasen, pp. 405, (Springer-Verlag, Berlin, 1991).

[4] A. Coniglio and M. Zanetti, Europhys. Lett. 10, 575 (1989); A. Coniglio and M. Zanetti, 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, 161-185 (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] T. Ohta and H. Nozaki, in Space-Time Organization in Macromolecular Fluids, Springer Series in Chemical Physics, Vol. 51, edited by F. Tanaka, M. Doi, and T. Ohta (Springer, Berlin, 1989).

[16] Y. Oono, unpublished (1989).

[17] G.F. Mazenko, Phys. Rev. Lett. 63 1605 (1989).

[18] G.F. Mazenko, Phys. Rev. B, 43 5747 (1991).

[19] H. Tomita, Prog. Theor. Phys. 72, 6566 (1984). H. Tomita, in Formation, Dynamics and Statistics of Patterns, ed. K. Kawasaki et al., (World Scientific, 1989).

[20] A. Shinozaki and Y. Oono, Phys. Rev. Lett. 66, 173 (1991).

[21] C. Yeung and D. Jasnow, Phys. Rev. A, 42, 10523 (1990).

[22] C. Yeung and D. Jasnow, J. Phys. A 23, L1309, (1990).

[23] F. Liu and G.F. Mazenko, Phys. Rev. B 44, 9185 (1991).

[24] H. Guo, Q. Zheng and J.D. Gunton, Phys. Rev. B 38, 11547 (1988).

[25] M. Grant and J.D. Gunton, Phys. Rev. B 28, 5496 (1983).

[26] K. Humayun and A.J. Bray, Phys. Rev. B 46, 10594 (1992).

[27] HAYAKAWA H. Hayakawa, Z. Racz and T. Tsuzuki, Phys. Rev. E 47, 1499 (1993).

[28] H. Toyoki, Phys. Rev. B 45, 1965 (1992).

[29] A.J. Bray and S. Puri, Phys. Rev. Lett. 67, 2670 (1991).

[30] F. Liu and G.F. Mazenko, Phys. Rev. B 45, 6989 (1992).

[31] F. Liu and G.F. Mazenko, Phys. Rev. B 45, 4656 (1992).

[32] Y. Oono and S. Puri, Phys. Rev. Lett. 58, 836 (1988); S. Puri and Y. Oono, Phys. Rev. A 38, 1542 (1988).
[33] A. Shinozaki and Y. Oono, Phys. Rev. E 47, 804 (1993).

[34] L. Bronsard and R.V. Kohn, J. Differential Equations 90, 211 (1991).

[35] P. Debye, H.R. Anderson and H. Brumberger, J. Appl. Phys. 28, 679 (1957); G. Porod, in Small Angle X-Ray Scattering, edited by O. Glatter and L. Kratky, (Academic Press, N.Y., 1983).

[36] M.T. Collins and H.C. Teh, Phys. Rev. Lett. 30, 781 (1973); T. Hashimoto, T. Miyoshi and M. Ohtsuka, Phys. Rev. B 13, 1119 (1976).

[37] Y. Oono and A. Shinozaki, Forma 4, 75 (1989); Y. Oono, IEICE Trans. E74, 1379 (1991).

[38] A. Shinozaki, unpublished.

[39] H. Furukawa, J. Phys. Soc. Jpn. 58, 216 (1989); H. Furukawa, Phys. Rev. B 40, 2341 (1989); H. Furukawa, Phys. Rev. B 42, 6438 (1990).

[40] A. Shinozaki and Y. Oono, unpublished.

[41] 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. Lond. A 422, 261 (1989).

[42] During the completion of this paper, we have received a preprint by H. Tomita in which he obtains a result similar to Eq. (5.7).

[43] We have received a preprint by A.J. Bray and K. Humayun which discusses the Bu closure and its equivalence with the OJK interface approach using the extra assumption that $Q(u) \sim u$. 
Figure 1: A log-log plot of $\langle u^2 \rangle$ vs. $L(t)^2$ from the $800 \times 800$ 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. We find that $\langle u^2 \rangle$ grows as $L^2$ as predicted by the interface approach. A fit to the form $\langle u^2 \rangle = aL^{2b} + c$ gives $b = 1.03$.

Figure 2: A plot of the single point probability distribution $\ln P(u^2/\langle u^2 \rangle, t)$ versus $u^2/\langle u^2 \rangle$ for $t = 50, 100, 200, 400$. Representative error bars are shown. This plot indicates that the tail of the distribution function decays as a Gaussian but there is a regime for $u^2/\langle u^2 \rangle < .5$ which decays slower than predicted by the tails.

Figure 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.

Figure 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.

Figure 5: 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 3-d spinodal decomposition simulation (dashed line). The inset is a blow up of the spectral density for values of $q$ just above the peak. We find that there is a violation of positivity both $q < .5$ and $1.5 < q < 4$, indicating that the Gaussian assumption is clearly invalid for the conserved order parameter case.

Figure 6: Plot of the real space correlation function $C(r,t)$ from the 3-d CDS simulations (dash line) versus the result using the Gaussian assumption with the B$\psi$ scheme (solid line). The inset shows $(r/L)^2C(r,t)$ for the same range of $r/L$. We find very good fits up to the second zero in the correlation function.