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Nonequilibrium autocorrelations in phase-ordering dynamics

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We investigate the ordering process of an unstable system governed by a nonconserved scalar order parameter using a theoretical approach developed previously. The two-time order-parameter correlation function is shown to obey asymptotic dynamical scaling. The temporal evolution of the autocorrelation function exhibits power-law decay with a nonequilibrium exponent. We find a relation between this exponent and a nonlinear eigenvalue controlling the equal-time structure factor. This relation, as well as the predicted values of the exponent, is compared with direct numerical simulations of a cell-dynamical-system (CDS) model of the ordering process.

I. INTRODUCTION

An understanding is beginning to emerge that the nonequilibrium kinetics of first-order phase transitions lead to additional qualitative phenomena when compared to the linear response kinetics valid near equilibrium.[1] A simple example is the phase-ordering process of a system following a rapid quench from a high temperature to a thermodynamically unstable state at a low temperature. Most work in this area has focused on the late stages of the phase separation process, where there is evidence of universality and scaling. This robust behavior is due to the existence of a single characteristic length, $L(t)$, which controls the ordering process. For pure systems, accumulating experimental,[2] numerical,[3] as well as analytical[4-8] studies demonstrates that the growth of $L(t)$, typical of the growing domain size, follows an algebraic growth law in time, $L(t)\sim t^n$, with n being a growth exponent. It is well established that n is independent of the spatial dimensionality and that $n=\frac{1}{2}$ in the universality class of the model A with nonconserved order parameter, while $n=\frac{1}{3}$ for model B where the order parameter is conserved. Moreover, the scattering structure factor $C(\mathbf{r},t)$ of the evolving system exhibits dynamics scaling

$$C(\mathbf{r},t)=\psi_0^2 F_0(\mathbf{r}/L(t)) , \tag{1.1}$$

where $C(0,t)=\psi_0^2$, ψ_0 is the equilibrium value of the order parameter, and F_0 is a universal function dependent on spatial dimensionality and possibly other parameters characteristic of the particular universality class of the system. While the scaling (1.1) is reasonably well established, theoretical determination of F_0 remains formidable.

A widely known, quantitative theory for the scaling function for a non-conserved order parameter was proposed by Ohta, Jasnow, and Kawasaki (OJK).[6] They used an approach based on an equation of motion for random interfaces. The structure factor given by OJK, when compared with two-dimensional numerical simulations available at that time, showed reasonable agreement. However, a particular feature of the OJK coordinate-space structure factor is its rather weak dependence upon the spatial dimensionality of the system, which appears only through a dimensionality dependent kinetic coefficient. More recently, a functional-integral approach to the domain growth problem was developed by one of us,[7] which accounts explicitly for the power-law growth of $L(t)$, as well as the dynamical scaling of the structure factor. One central result of the theory is that the asymptotic universal function F_0 is the solution of a nonlinear eigenvalue problem with an eigenvalue μ which is uniquely determined by proper boundary conditions. In contrast to the OJK result, our scaling function displays explicit dependence upon spatial dimensionality d. In a recent work,[9] we showed that our theory interpolated sensibly between $d=1$ and $d=\infty$. In one dimension, our scaling function recovers the exact result of the kinetic Ising model of Glauber;[10] in $d=\infty$, it coincides with the form factor of OJK. We therefore believe that the OJK form is only asymptotically valid in $d=\infty$. The structure factor we obtained in two dimensions has been compared with accurate numerical simulations of the kinetic Ising model, yielding very good agreement which is superior to that of the OJK form.

In this paper we investigate the dynamical scaling of the two-time correlation function and, in particular, the decay of the autocorrelation function in a time-dependent Ginzburg-Landau (TDGL) model for a nonconserved order parameter. The method we employ is rooted in the framework of the previously developed functional-integral approach.[7] The usefulness of multiple-time correlation functions in phase-ordering dynamics was put forward by Furukawa,[11] who pointed out the possibility of dynamical scaling in the late stages. Although there exist, though preliminary, numerical simulations of the kinetic Ising and cell dynamical models, the multiple-time correlation function has seldomly been explored theoretically. Zannetti and Mazenko[12] examined the case of an n-vector model in the large-n limit. This model was recently studied by Newman and co-workers[13] using a $1/n$ expansion. In the case of a scalar order parameter, Yeung and Jasnow[14] studied the multiple-time correlation function and the non-self-averaging properties of the OJK model. In this work, we shall derive (in Sec. II) and solve the equation of motion for the two-time correlation

function in the scaling regime. As in the case for the equal-time structure factor, we find that the two-time correlation function is also exactly solvable in one dimension, leading to the known result for the kinetic Ising model. The OJK correlation function obtained by Yeung and Jasnow is also recovered in the limit $d \to \infty$.

An interesting quantity in the phase-ordering process is the autocorrelation function $A(t,t') \equiv \langle \psi(0,t)\psi(0,t') \rangle$. It can be regarded intuitively as the "overlap" between order-parameter profiles at different times. Therefore we expect its time evolution to contain interesting information about the ordering process. For example, the remnant magnetization in spin-glass models can be thought of as a special form of the autocorrelation function, as will be discussed later. It was recognized by Fisher and Huse[15] that the decay of the autocorrelation function for $t \gg t'$ is governed by a power law with the characteristic length scale of the system, namely, $A(t,t') \sim L(t)^{-\lambda}$ where λ is a nonequilibrium exponent, having no apparent relation to the dynamical exponent n characterizing the growth of $L(t)$ itself. Furthermore, it was suggested, based on a scaling argument, that the exponent satisfy the bounds $d \geq \lambda \geq d/2$. The exponent λ has been calculated for the n-component Ginzburg-Landau model perturbatively in $1/n$.[13] Recently, λ was estimated in two dimensions via numerical simulations of the kinetic Ising model by Humayun and Bray.[16]

We will demonstrate, utilizing the general equation of motion derived for the two-time correlation function, that the autocorrelation function follows a power-law decay. Surprisingly, we discover a relation between the nonequilibrium exponent λ and the nonlinear eigenvalue μ determining the equal-time structure factor: $\lambda = d - \pi/4\mu$. When combined with the previously determined values of μ, we conclude that the bounds mentioned above are strictly satisfied. The OJK model predicts that $\lambda \equiv d/2$ in all dimensions, while our analysis suggests that the lower bound is approached only asymptotically as $d \to \infty$. To test our prediction for the exponent λ, numerical simulations of a cell-dynamical model is performed. The results of these simulations show unambiguously that λ is larger than the lower bound $d/2$ for $d \leq 3$. Results of our numerical simulations are also compared with currently existing results.

II. EQUATION OF MOTION FOR THE CORRELATION FUNCTION

Consider the TDGL equation for a nonconserved scalar order parameter $\psi(\mathbf{r},t)$:

$$\frac{\partial \psi(\mathbf{r},t)}{\partial t} = -\frac{\partial F(\psi(\mathbf{r},t))}{\partial \psi(\mathbf{r},t)} + \eta(\mathbf{r},t) , \tag{2.1}$$

where η is a Langevin noise which is taken as Gaussian distributed:
$\langle \eta(\mathbf{r},t) \rangle = 0, \langle \eta(\mathbf{r},t)\eta(\mathbf{r}',t') \rangle = 2T\delta(\mathbf{r}-\mathbf{r}')\delta(t-t')$, T is the temperature after the quench, and time scale is chosen by setting the bare kinetic coefficient equal to unity. The coarse-grained free energy F is assumed to have the form

$$F = \int d\mathbf{r} [\tfrac{1}{2}(\nabla\psi)^2 + V(\psi)] , \tag{2.2}$$

where $V(\psi)$ is a symmetric degenerate double-well potential. We define the two-time correlation function

$$C(\mathbf{r},t,t') = \langle \psi(\mathbf{r},t)\psi(0,t') \rangle , \tag{2.3}$$

where $\langle \cdots \rangle$ denotes ensemble averages over random initial conditions and the noise. In turn, the autocorrelation function is simply $A(t,t') \equiv C(0,t,t') = \langle \psi(0,t)\psi(0,t') \rangle$ and the equal-time correlation function or structure factor is $C(\mathbf{r},t) \equiv C(\mathbf{r},t,t) = \langle \psi(\mathbf{r},t)\psi(0,t) \rangle$. We have assumed a spatially homogeneous system.

To derive the equation of motion for the correlation function we employ a formalism developed by one of us. We will not get into details of the method which were thoroughly described in Ref. 7. However, we will illustrate the main steps of the theory as we proceed. The first key step involves the decomposition of order parameter into two separate pieces: an *ordering* part σ and a *fluctuating* part ζ:

$$\psi(\mathbf{r},t) = \sigma(\mathbf{r},t) + \zeta(\mathbf{r},t) , \tag{2.4}$$

where σ reflects the bulk ordering on large, growing length scales, and ζ is dominated by local fluctuations on short-length scales ($\sim$ the equilibrium correlation length ξ). The separation (2.4) is motivated by the recognition that $L(t) \gg \xi$ for sufficiently long times and the fact that noise does not seem to affect dynamical scaling for deep quenches. The decoupling of these two fields σ and ζ occurs in a relatively short time after the quench. Consequently, the ensemble averages over ψ can be replaced by that over σ, e.g., $\langle \psi\psi \rangle \to \langle \sigma\sigma \rangle$.

Imagine the landscape of σ at long times, when σ has taken its equilibrium value $\pm\psi_0$ almost everywhere except at *sharp* interfaces between the bulk ψ_0 and $-\psi_0$ regions. It is advantageous, in representing this order-parameter landscape, to introduce an auxiliary field $m(\mathbf{r},t)$ serving as a "coordinate" measuring the distance from point $\mathbf{r}$ to a nearby sharp interface:

$$\sigma(\mathbf{r},t) = \sigma(m(\mathbf{r},t)) . \tag{2.5}$$

The relation between σ and m is chosen to satisfy the classical equation of motion for an interface in equilibrium:[17]

$$\frac{1}{2}\frac{\partial^2 \sigma}{\partial m^2} - \frac{dV(\sigma)}{d\sigma} = 0 . \tag{2.6}$$

Now the statistical properties of the ordering field σ can be completely specified by the probability distribution of the auxiliary field m. The virtue of introducing m is apparent: though $\sigma(\mathbf{r},t)$ shows rapid spatial variation due to the presence of sharp interfaces, the variation of $m(\mathbf{r},t)$ in space is rather smooth. This smoothness is important as to allow us to make the key assumption that the probability distribution $P[m]$ is Gaussian, whose moments have to be determined self-consistently, as will be shown later. Of course non-Gaussian distributions can be tried if the Gaussian distribution is found unsatisfactory.

So far this has not been the case.

Having introduced the auxiliary field, ensemble averages of functions of ψ over random initial conditions are replaced by the averages involving σ with respect to the probability distribution $P[m]$, which can be readily evaluated. Suppose $p(m(\mathbf{r}_1,t))$ and $q(m(\mathbf{r}_2,t'))$ are two arbitrary functions, denoted by $p(1)$ and $q(2)$, respectively. To calculate the average $\langle p(1)q(2)\rangle$, we have

$$\langle p(1)q(2)\rangle = \int_{-\infty}^{\infty} dx_1 dx_2 p(x_1)q(x_2)\langle \delta(x_1-m(1))\delta(x_2-m(2))\rangle$$
$$= \int_{-\infty}^{\infty} dx_1 dx_2 p(x_1)q(x_2)\Phi(x_1,x_2) , \tag{2.7}$$

where

$$\Phi(x_1,x_2)=\langle \delta(x_1-m(1))\delta(x_2-m(2))\rangle = \int \frac{dk_1 dk_2}{4\pi^2} e^{-ik_1x_1-ik_2x_2}\langle e^{ik_1m(1)+ik_2m(2)}\rangle$$
$$= \int \frac{dk_1 dk_2}{4\pi^2} e^{-ik_1x_1-ik_2x_2} e^{-k_1^2S_0(1)/2-k_2^2S_0(2)/2-k_1k_2C_0(12)} , \tag{2.8}$$

where $S_0(1)=\langle m^2(1)\rangle$, $S_0(2)=\langle m^2(2)\rangle$, and $C_0(12)=\langle m(1)m(2)\rangle$. Equation (2.8) is a Gaussian integral in k_1 and k_2, which equals

$$\Phi(x_1,x_2)=\frac{\gamma}{2\pi\sqrt{S_0(1)S_0(2)}}\exp\left[-\frac{\gamma^2}{2}\left[\frac{x_1^2}{S_0(1)}+\frac{x_2^2}{S_0(2)}-\frac{2fx_1x_2}{\sqrt{S_0(1)S_0(2)}}\right]\right] , \tag{2.9}$$

where

$$f(12)=\frac{C_0(12)}{\sqrt{S_0(1)S_0(2)}} \tag{2.10}$$

and

$$\gamma=1/\sqrt{1-f^2} . \tag{2.11}$$

We are now in the position to derive the equation of motion satisfied by $C(12)\equiv C(\mathbf{r},t,t')$ in the scaling limit when both $S_0(1)$ and $S_0(2)$ are large. First neglect the noise term in (2.1) and replace $\psi(\mathbf{r},t)$ by $\sigma(1)$, multiply by $\sigma(2)$ on both sides and average over $P[m]$, to obtain

$$\frac{\partial C(12)}{\partial t}=\nabla_{\mathbf{r}}^2 C(12)-\tfrac{1}{2}C_{20}(12) , \tag{2.12}$$

where the notation

$$\sigma_n(1)=\frac{\partial^n\sigma(1)}{\partial m^n(1)} , \tag{2.13}$$

$$C_{nl}(12)=\langle \sigma_n(1)\sigma_l(2)\rangle \tag{2.14}$$

is introduced and (2.6) is used to express the derivative of the potential in terms of $\sigma(1)$. The convention $C_{00}(12)=C(12)$ is also implied. In the long-time limit, one can express $C_{20}(12)$ back in terms of $C(12)$ and thus close Eq. (2.12). This is accomplished conveniently by making use of two general identities[7]

$$\frac{\partial}{\partial S_0(1)}C_{nm}(12)=\tfrac{1}{2}C_{n+2,m}(12) \tag{2.15}$$

and

$$\frac{\partial}{\partial C_0(12)}C_{nm}(12)=\tfrac{1}{2}C_{n+1,m+1}(12) . \tag{2.16}$$

The procedure is to first compute $C_{11}(12)$. From (2.7),

$$C_{11}(12)=\int_{-\infty}^{\infty} dx_1 dx_2 \sigma'(x_1)\sigma'(x_2)\Phi(x_1,x_2) , \tag{2.17}$$

which in long-time limit when $S_0(1)$, $S_0(2)\gg 1$ gives to leading order

$$C_{11}(12)=\frac{\gamma(12)}{\sqrt{S_0(1)S_0(2)}}\frac{(2\psi_0)^2}{2\pi} \tag{2.18}$$

plus terms of higher order in powers of $S_0^{-1/2}(1)$ and $S_0^{-1/2}(2)$. Note that the leading term, which will dominate the asymptotics, is *independent* of the particular form of the potential V. We have only used the trivial identity

$$\int_{-\infty}^{+\infty} dx_1\sigma'(x_1)=2\psi_0 . \tag{2.19}$$

Now using (2.16) with $n=m=0$ and integrating over C_0, we obtain to leading order

$$C(12)=\frac{2\psi_0^2}{\pi}\arcsin[f(12)] . \tag{2.20}$$

Differentiation with respect to $S_0(1)$ using (2.15) gives

$$C_{20}(12)=-\frac{2\psi_0^2}{\pi S_0(1)}f(12)\gamma(12) . \tag{2.21}$$

Eliminating $f(12)$ between (2.20) and (2.21) leads to the main result of interest here,

$$C_{20}(12)=-\frac{2\psi_0^2}{\pi S_0(1)}\tan\left[\frac{\pi C(12)}{2\psi_0^2}\right] . \tag{2.22}$$

Inserting this result back into (2.12) and choosing $\psi_0=1$ without loss of generality, we arrive at the final equation of motion for the correlation function:

$$\frac{\partial C(\mathbf{r},t,t')}{\partial t}=\nabla_{\mathbf{r}}^2 C(\mathbf{r},t,t')+\frac{1}{\pi}\frac{\tan[\pi C(\mathbf{r},t,t')/2]}{S_0(t)}+\left[\frac{1}{S_0(t)^2}\right]+O\left[\frac{1}{S_0(t)S_0(t')}\right], \tag{2.23}$$

where we have used arguments t,t' instead of (1,2) in the equation. We shall assume $t\geq t'$ later on.

III. ASYMPTOTIC STRUCTURE FACTOR

First consider the equal-time correlation function or the structure factor $C(\mathbf{r},t)\equiv C(\mathbf{r},t,t)$. Noticing that

$$\frac{\partial C(\mathbf{r},t)}{\partial t}=\left.\frac{\partial C(\mathbf{r},t,t')}{\partial t}\right|_{t'=t}+\left.\frac{\partial C(\mathbf{r},t,t')}{\partial t'}\right|_{t'=t}, \tag{3.1}$$

it follows at once from (2.23) that

$$\frac{1}{2}\frac{\partial C(\mathbf{r},t)}{\partial t}=\nabla_{\mathbf{r}}^2 C(\mathbf{r},t)+\frac{1}{\pi}\frac{\tan[\pi C(\mathbf{r},t)/2]}{S_0(t)}+O\left[\frac{1}{S_0(t)^2}\right]. \tag{3.2}$$

For the late stages of the phase separation, we look for self-similar scaling solutions. Inserting the scaling form (1.1) into (3.2), and identifying $L^2(t)\equiv\pi S_0(t)\equiv 4\mu t$, gives for the asymptotic scaling function

$$\nabla^2 F_0+\mu\mathbf{x}\cdot\nabla F_0+\tan\left[\frac{\pi}{2}F_0\right]=0, \tag{3.3}$$

where $\mathbf{x}=\mathbf{r}/L(t)$ and μ is an eigenvalue to be determined under proper boundary conditions. In deriving (3.3), the late-stage condition $t\gg 1$ and $L(t),S_0(t)\gg 1$ has been used.

The eigenvalue problem (3.3) is solved under the boundary condition $F_0(0)=1$ and the requirement that the Fourier transform of F_0 is well behaved. This fixes uniquely the eigenvalue $\mu^*(d)$ as a function of dimensionality. We showed earlier[9] that (3.3) is analytically soluble in $d=1$ and $d=\infty$. In one dimension, the scaling function is the same as that of the Glauber-Ising model

$$F_0(x)=\mathrm{erfc}(\sqrt{\mu/2}x), \tag{3.4}$$

where erfc is the complementary error function; in the other extreme $d\to\infty$, the structure factor agrees with that of the OJK model. In between these two limits, the theory displays explicit dependence of the structure factor on spatial dimensionality.[9] The calculated structure factor in two dimension was shown to be in good agreement with recent numerical simulations of a two-dimensional kinetic Ising model.[16]

IV. TWO-TIME CORRELATION FUNCTION

We now turn to the two-time correlation function in the late stages when scaling behavior is expected. To this end, suppose $t\geq t'\gg 1$ and both $S_0(t)$ and $S_0(t')$ are large. Neglecting terms higher order in $1/S_0(t)$ in (2.23), we obtain

$$\frac{\partial C(\mathbf{r},t,t')}{\partial t}=\nabla_{\mathbf{r}}^2 C(\mathbf{r},t,t')+\frac{1}{L^2(t)}\tan\left[\frac{\pi}{2}C(\mathbf{r},t,t')\right], \tag{4.1}$$

where $L^2(t)=4\mu^* t$ and μ^* is the eigenvalue that satisfies (3.3) and the proper boundary conditions. Equation (4.1) has to be supplemented by the condition $C(\mathbf{r},t',t')=C(\mathbf{r},t')$.

It is reasonable to look for scaling solutions of the correlation function using the ansatz

$$C(\mathbf{r},t,t')=C\left[\frac{\mathbf{r}}{L(t)},\frac{t}{t'}\right]=F(\mathbf{x},\tau), \tag{4.2}$$

where for convenience two independent variables, $\mathbf{x}=\mathbf{r}/2\sqrt{t}$, and $\tau=\ln(t/t')/4$, are used. Inserting the ansatz (4.2) into (4.1), we obtain the equation of motion

$$\frac{\partial F(\mathbf{x},\tau)}{\partial\tau}=\nabla^2\mathrm{F}+2\mathbf{x}\cdot\nabla\mathrm{F}+\frac{1}{\mu^*}\tan\left[\frac{\pi}{2}\mathrm{F}\right] \tag{4.3}$$

with "initial" condition $\mathbf{F}(\mathbf{x},\tau=0)=\mathrm{F}_0(\mathbf{x})$ where F_0 satisfies

$$\nabla^2 F_0+\mathbf{x}\cdot\nabla F_0+\frac{1}{\mu^*}\tan\left[\frac{\pi}{2}F_0\right]=0. \tag{4.4}$$

Note that (4.4) is the same as (3.3) after a simple scaling of lengths by a factor of $\sqrt{\mu^*}$.

Equation (4.1) or (4.3) can be rigorously solved in one dimension. It is somewhat easier to work with (4.1). Fourier transformation of the equal-time structure factor (3.4) gives

$$C(\mathbf{k},t')=\sqrt{32t'/\pi}\int_0^{\pi/2}d\theta\cos\theta\exp(-2k^2t'\cos^2\theta). \tag{4.5}$$

Keeping in mind that as $d\to 1$, $\mu^*\to\infty$, it follows easily from (4.1), after a Fourier transformation, that

$$C(\mathbf{k},t,t')=C(\mathbf{k},t')\exp[-k^2(t-t')]. \tag{4.6}$$

Inserting (4.5) into (4.6) and Fourier transforming back to coordinate space, we obtain the one-dimensional two-time correlation function

$$C(\mathbf{r},t,t')=\frac{2}{\pi}\int_0^{\arcsin\sqrt{2t'/(t+t')}}du\exp\left[-\frac{\mathbf{r}^2(1+\tan^2 u)}{4(t+t')}\right] \tag{4.7}$$

and in particular the autocorrelation function

$$A(t,t')=C(0,t,t')=\frac{2}{\pi}\arcsin\left[\frac{2t'}{t+t'}\right]^{1/2}. \tag{4.8}$$

These results are exactly what one obtains for the one-dimensional Glauber-Ising model.[18] For $t\gg t'$, the autocorrelation function exhibits a power-law decay

$$A(t,t')\sim\frac{2\sqrt{2}}{\pi}\left[\frac{t'}{t}\right]^{1/2}. \tag{4.9}$$

This power-law decay of the autocorrelation function is believed to be a quite generic feature of the domain growth problem. In general, one expects

$$A(t,t')\sim\frac{1}{L^{\lambda}(t)}\quad\text{for } t\gg t', \tag{4.10}$$

where λ is a dynamical nonequilibrium exponent, which has no apparent relation to the usual growth exponent n. The exponent λ was introduced by Fisher and Huse in their study of the remnant magnetization in spin-glass models.[15] They also proposed a bound for λ based on scaling arguments. The lower bound is easy to understand through the following physical picture. Consider a large domain of size l. At an early time t', the domain contains many sharp interfaces. Therefore we can imagine that the order parameter oscillates randomly between the two equilibrium values $\pm\psi_0$ within the domain. Then the mean value of the order parameter per site within the domain is, by the central-limit theorem, of the order $\sim l^{d/2}/l^{d}\sim 1^{-d/2}$. By taking l equal to the typical domain size at t, we obtain essentially the maximum possible value for the overlap between order parameters at t and t'. Thus we have a lower bound $\lambda\geq d/2$. Interestingly, this picture of randomly distributed interfaces is the key assumption underlying the OJK model. Therefore it is not surprising that, as we shall see later, the OJK model predicts a λ strictly equal to this lower bound. In addition, Fisher and Huse also conjectured an upper bound $\lambda\leq d$. This upper bound is in essence the statement that biases in the initial order-parameter distribution do not diminish in time. Combining both, they suggested

$$d\geq\lambda\geq d/2 \tag{4.11}$$

to be valid in the pure Ising system quenched from infinite temperature to zero temperature. We immediately observe that our result in one dimension, $\lambda=1$, takes the upper bound value in (4.11).

Recently Yeung and Jasnow[14] investigated multiple-time correlation functions within the OJK model. In particular, they obtained the two-time correlation function

$$C_{\mathrm{OJK}}(\mathbf{r},t,t')=\frac{2}{\pi}\arcsin\left[\left[\frac{2\sqrt{tt'}}{t+t'}\right]^{d/2}e^{-r^2/4(t+t')}\right]. \tag{4.12}$$

It follows automatically that $A(t,t')\sim(t'/t)^{d/4}$ as $t\gg t'$, and $\lambda=d/2$. Going back to our theory, we point out that (4.1) can also be solved perturbatively in the large-d limit, in the same spirit as in Ref. 9. In fact, it is readily verified by inserting (4.12) into (4.1) that the OJK result solves Eq. (4.1) to first order in $O(1/d)$. This reinforces our assertion that the OJK theory is only asymptotically correct as $d\to\infty$. Note that the outcome $\lambda=d/2$ in *all* dimensions in the OJK model is an inevitable consequence of the assumption in their theory that domain walls are randomly distributed. As will be discussed in the next section, numerical evidence in both $d=2$ and $d=3$, as well as the exact result in $d=1$ leads to the conclusion that the lower bound $d/2$ is not achieved in these dimensions. Below, we shall determine the exponent λ in our theory for arbitrary dimension d.

We propose, and derive below, that

$$\lambda=d-\pi/4\mu^{*}. \tag{4.13}$$

Recalling from Ref. 9 that $\infty\geq\mu^{*}(d)\geq\pi/2d$, we see that λ as given by (4.13) automatically satisfy the bounds (4.11) set up by Fisher and Huse. The upper and lower bounds are attained at $d=1$ and $d=\infty$, respectively. We now derive the exponent relation (4.13). Consider the long-time limit of (4.1) when $t\gg t'$ and $C(\mathbf{r},t,t')$ is small. We can then make the substitution $\tan(\pi C/2)\sim\pi C/2$ and obtain

$$\frac{\partial C}{\partial t}=\nabla^2 C+\frac{\pi}{8\mu^{*}t}C. \tag{4.14}$$

Defining $\tilde{C}(\mathbf{r},t,t')=t^{-\pi/8\mu^{*}}C(\mathbf{r},t,t')$, we find that $\tilde{C}(\mathbf{r},t,t')$ satisfies the diffusion equation

$$\frac{\partial\tilde{C}}{\partial t}=\nabla^2\tilde{C}. \tag{4.15}$$

The asymptotic behavior of the diffusion equation in d dimensions is well known:

$$\tilde{C}(\mathbf{r},t,t')\sim t^{-d/2}\exp(-r^2/4t). \tag{4.16}$$

Therefore it follows as $t\gg t'$ that

$$A(t,t')=t^{\pi/8\mu^{*}}\tilde{C}(0,t,t')\sim t^{-d/2+\pi/8\mu^{*}}\sim L(t)^{-(d-\pi/4\mu^{*})}, \tag{4.17}$$

which, in turn, leads to (4.13).

We have so far discussed the solution of (4.1) in two limiting cases of one and infinite dimension. In principle,

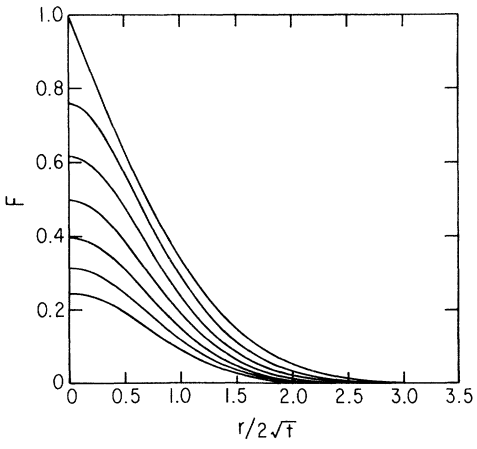

FIG. 1. Two-time correlation function $F(r/2\sqrt{t},\tau)$ vs $r/2\sqrt{t}$, where $\tau\equiv\ln(t/t')/4$. Curves from top down correspond to $\tau=0.0$, 0.1, 0.2, 0.3, 0.4, 0.5, and 0.6. The curve for $\tau=0$ is the equal-time scaling function.

the two-time correlation function in any dimension is given by the solution of (4.3) and (4.4). Unfortunately, (4.3) appears analytically intractable in general. We can, however, numerically integrate the initial value problem using an implicit finite-difference scheme. As a by product, the validity of (4.13) was further checked to be correct. In Fig. 1, we plot the two-dimensional two-time correlation function at sequential times $t \geq t'$. Note the absence of a cusp in the correlation function near $\mathbf{r}=0$ for any $t > t'$. On the other hand, the cusp for the equal-time structure factor reflects the well-known Porod's law. Although a direct experimental determination of the two-time correlation function seems difficult, it may be possible to compare our result with accurate numerical simulations.

V. NUMERICAL SIMULATION

We showed in the previous sections the existence of dynamical scaling for two-time correlation functions. We have also related the nonequilibrium exponent λ to the nonlinear eigenvalue $\mu^*(d)$, which was previously accurately determined.[7,9] In particular, $\mu^*(2)=1.1042$ and $\mu^*(3)=0.5917$. These values imply for the nonequilibrium exponent $\lambda(d=2)=1.2887$ and $\lambda(d=3)=1.6726$. In this section, we perform numerical simulations of a cell-dynamical system (CDS) model to test these predictions.

Before presenting our own result, we first review the existing numerical works. An early simulation was carried out by Fisher and Huse,[15] who studied a nearest-neighbor kinetic Ising model quenched to zero temperature using a Metropolis algorithm. They obtained in two dimensions $\lambda_2 \approx 1.25$, derived from averages over 80 random initial conditions on a 400^2 square lattice. The same system was later studied using a slightly larger system (500^2) by Furukawa,[11] who obtained a value consistent with $\lambda^2 \approx 1.30$. Most recently, a more extensive simulation was done by Humayun and Bray[16] using an even larger (1000^2) system averaged over 160 histories. The exponent they extracted was $\lambda_2 \approx 1.24$. All these values of λ_2 are considerably larger than the lower bound $d/2=1$.

In three dimensions, simulation results are scarce. We are only aware of the simulation by Fisher and Huse[15] on a 80^3 simple cubic lattice with Ising dynamics. Unfortunately their result is not conclusive. While the simulation suggested a value close to the lower bound $\lambda_3 \sim 1.50$, the authors cautioned, however, due to the quality of the convergence, that any value in the range $1.50 \leq \lambda \leq 1.65$ would be equally consistent with their data.

We have performed simulations in both two and three dimensions, using a CDS model. The (deterministic) CDS model, proposed by Oono and Puri,[19] has proved valuable in studying the late stages of phase-ordering processes. The explicit CDS update we used is

$$\psi(\mathbf{r},t+1)=F(\psi(\mathbf{r},t)+D(\langle\langle\psi(\mathbf{r},t)\rangle\rangle-\psi(\mathbf{r},t)) , \quad (5.1)$$

where $\langle\langle \cdots \rangle\rangle$ denotes averaging over nearest-, next-nearest, and (in three-dimensionals only) next-next-nearest-neighbor cells. The weighing ratio are 2:1 in two dimensions and 6:3:1 in three dimensions. We choose the map function $F(\psi)=A\tanh\psi$ with $A=1.3$, and diffusion constant $D=0.5$. The two-dimensional simulations were performed on 600^2 square lattices. We fix $t'=30$, and monitor the evolution of our system up to times $t=3000$. At the latest time, the typical domain size is about $\frac{1}{15}$ of the system size. Smaller systems are also comparatively studied to prevent finite-size effects. We averaged over 30 random initial conditions to obtain the autocorrelation function

$$A(t,t')=\sum_{\mathbf{r}}\psi(\mathbf{r},t)\psi(\mathbf{r},t')/N ,$$

where N is the total number of sites on the lattice. $A(t,t')$ is then fitted to the algebraic growth law $A(t,t')\sim t^{-\lambda_{\rm eff}/2}$ with an effective exponent $\lambda_{\rm eff}$ which slowly changes with time. The truly asymptotic value of the exponent λ can be extracted by interpolating to $t\to\infty$. In Fig. 2, we plot the measured effective exponent $\lambda_{\rm eff}$ as a function of $1/\sqrt{t}$ for both two and three dimensions. Our extrapolated two-dimensional exponent is $\lambda_2=1.246\pm0.02$. The error estimate is from the statistical analysis of data, and therefore does not include possible systematic deviations. Our result from the CDS model agrees with those obtained both by Humayun and Bray[16] and by Fisher and Huse[15] in a different model, the kinetic Ising model. On the other hand, our predicted value 1.2887 does appear larger, although only slightly, than the numerical estimates.

Our three-dimensional simulations are performed on lattice of sizes 64^3, 72^3, 80^3, 88^3, and 128^3, each averaged over 200 random initial conditions, except for 128^3 where 50 initial conditions are used. We choose $t'=40$ and evolve the system up to $t=700$. Unlike the two dimen-

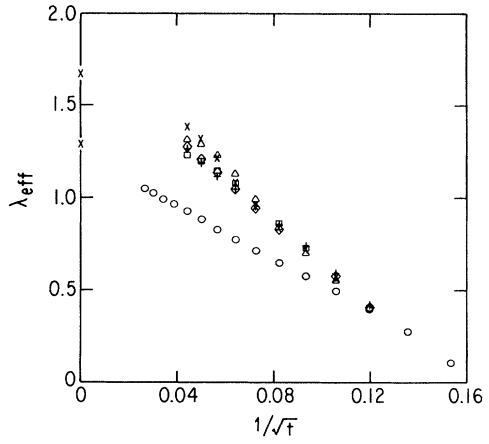

FIG. 2. Effective exponent $\lambda_{\rm eff}$ vs $1/\sqrt{t}$. The asymptotic value of λ can be obtained by interpolating $\lambda_{\rm eff}$ to $t\to\infty$. ○ corresponds to two-dimensional (2D) simulations on 600^2 lattices. Symbols +, □, △, ◇, and × correspond to 3D simulations on 64^3, 72^3, 80^3, 88^3, and 128^3 lattices, respectively. The two points on the vertical axis are predictions from (4.13).

sional case, we believe that the finite-size effects do influence our data to some extent at the latest times. This presumably accounts for the data scatter on Fig. 2 at large t. However, we are unable to extract definitively any systematic trend of the finite-size effects. The interpolation of Fig. 2 gives $\lambda_3 = 1.838 \pm 0.2$. This value is large compared with the only available study by Fisher and Huse.[15] We believe that our result rules out the possibility of λ_3 equaling the lower bound 1.5. The predicted value from our theory 1.6726 also agrees with the numerical value within the rather conservative error estimates. Caution is due, however, to the large statistical uncertainties in both our study and that of Fisher and Huse. An accurate estimate of λ_3 is still lacking at present. It would be important to determine accurately the value of λ_3. This will not only check the true universality of this nonequilibrium exponent, but will also provide an interesting test for any theory of the phase-ordering dynamics. We believe that these aspects deserve further study.

VI. CONCLUSIONS

To summarize, we have demonstrated that the two-time correlation function associated with the growth kinetics of a system with a nonconserved scalar order parameter satisfies dynamical scaling in the late stages. The equation of motion for the two-time correlation function is derived in a closed form. The theory presented here offers considerable improvement to the well-known approach of OJK, in several aspects. First, our theory is exactly solvable both in one dimension and in the limit of $d \to \infty$. In one dimension it recovers the exact result of the kinetic Ising model. And in the limit of high dimensionality, it includes the OJK result as a special case. Second, study of the power-law decay of the autocorrelation function reveals a relation between the nonequilibrium exponent λ and the nonlinear eigenvalue associated with the equal-time scaling function. A direct consequence of this relation is that exponent λ is always *greater* than $d/2$, the value obtained from the OJK model. This fact seems to imply that the OJK assumption of randomly distributed interfaces needs to be modified to account for correctly the ordering process.

Our numerical results in both two and three dimensions agrees qualitatively with our theoretical prediction. In two dimensions, the measured value of λ_2 also agrees with the previous simulations of the two-dimensional kinetic Ising model. These agreements suggest that the TDGL, CDS, and the kinetic Ising model are in the same universality class regarding this exponent. In view of the slight difference between the predicted and the measured value of λ_2, as well as the large statistical uncertainty in λ_3, we point out that more work is certainly needed to examine the validity of this universality, especially in three dimensions, where an accurate estimate of λ_3 is still unavailable.

The concepts of multiple-time correlation and the power-law decay of autocorrelations should be investigated in the case of phase separation kinetics of conserved order parameters. An interesting attempt has already been made by Furukawa.[11]

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