Breakdown of linear dynamics in phase-ordering kinetics

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Simulations and experiments have shown that the linear theory in phase-ordering dynamics fails first on short length scales rather than long length scales as suggested previously. By following the example of a simple coupled nonlinear system, we show that the linear theory breaks down first at the largest wave numbers due to the nonlinear slaving of the most stable Fourier modes to the larger amplitude unstable modes. The range of wave numbers in which the dynamics is nonlinear expands toward smaller wave numbers with time. We present numerical results verifying the mode slaving hypothesis and determine $t_{\rm br}(k)$, the time at which the linear theory breaks down as a function of wave number k.

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Phase-ordering kinetics occurs when a system undergoes a temperature quench from a disordered state above the critical point to a final state inside the coexistence curve [1]. If the order parameter is conserved, such as in a binary alloy, this process is known as spinodal decomposition while if the order parameter is not conserved, as in an order-disorder transition, this is known as continuous ordering. Both these processes are of technological interest to the material science and metallurgic community. For example, alloys become embrittled when phase separation occurs [2]. On a fundamental level, the phase-ordering process is ubiquitous and serves as an acid test in the understanding of nonequilibrium dynamics.

The phase-ordering process can be divided into several time regimes. At early times when the composition fluctuations are small, the dynamics are believed to be linear and characterized by exponential growth of composition fluctuations [3,4]. At later times, the system effectively consists of domains of the different phases separated by sharp interfaces. In this late stage, the domain growth is determined by the dynamics of the interfaces and characterized by dynamical scaling; that is, the domain morphology is invariant and can be described by a characteristic length scale that grows in time [5–7]. This paper is concerned with the limits of the early-time regime at which the linear theory first fails to describe the evolution of the Fourier modes.

The linear theory for spinodal decomposition was introduced in 1959 by Cahn and Hilliard [3] and later extended by Cook to include thermal noise [4]. They predicted that at early times the composition fluctuations will grow exponentially at long wavelengths while the short-wavelength modes relax to "false" equilibrium values. Binder [8] argued that the time at which the linear theory fails depends on the rate of growth of the fastest growing mode. Although experiments [9] and simulations [10,11] lend some support to the linear Cahn-Hilliard-Cook theory, there is no support for the breakdown first occurring at the fastest growing mode. In fact, experimentally, the linear predictions fail first on short length scales [12]. Ising model simulations [13,14] also showed that the linear theory fails on short length scales even when the long-length-scale behavior is described by linear theory. The failure on small length scales was found to correspond to the formation of isolated domains of one phase or the other in a sea where the order parameter is zero [13,14]. More recently Corberi, Coniglio, and Zannetti [15] have argued that there are actually two early-time regimes; a regime in which the composition fluctuations first smooths out and decreases diffusively followed by a regime in which the local order parameter grows rapidly towards the equilibrium values.

In this paper, we clarify the above observations by considering the dynamics in Fourier space. In particular, we show that the time at which the linear theory fails depends on the wave number. Using the concept of mode slaving we argue that the most stable modes become adiabatically slaved to the larger-amplitude less-stable modes. Therefore the linear theory breaks down first at the largest wave numbers and the regime in which the dynamics are nonlinear expands toward smaller wave number with time. We extend the mode slaving arguments to systems for which the relaxation time scales of the different modes are not well separated. Finally, we present numerical results verifying the mode slaving hypothesis and determine the wave-number dependence of the time $t_{\rm br}(k)$ at which mode k deviates from the linear behavior. In the large-k limit this breakdown time $t_{\rm br}$ behaves as $t_{\rm br} \sim (\ln \Delta)/k^{-2}$, where Δ is the amplitude of the initial conditions. In this paper we will restrict ourselves to the nonconserved order parameter. However, much of the arguments discussed below are also applicable to the conserved order parameter.

We model the phase-ordering dynamics via the time-dependent Ginzburg-Landau (TDGL) equation

$$\frac{\partial \phi(\mathbf{r},t)}{\partial t} = -\frac{\delta F\{\phi\}}{\delta \phi(\mathbf{r},t)} = \phi(\mathbf{r},t) - \phi^3(\mathbf{r},t) + \nabla^2 \phi(\mathbf{r},t). \quad (1)$$

Here $\phi(\mathbf{r},t)$ is the local order parameter, which describes, for example, the local species concentration and $F\{\phi\}$ is the coarse-grained free energy, which we take to be the ϕ^4 free energy. For simplicity we assume that the mobility is a constant that we set to unity and we take the

low-temperature limit by neglecting a thermal noise term in Eq. (1). Here we assume that the coarse-grained description is valid down to length scales smaller than the interfacial thickness between the two coexisting phases. This will be the case for Ising models with long-range interactions and for polymer blends if the correlation length is much larger than the radius of gyration.

In Fourier space the time-dependent Ginzburg-Landau equation becomes

$$\frac{\partial \phi_{\mathbf{k}}}{\partial t} = (\omega_k \phi_{\mathbf{k}} - N_{\mathbf{k}}\{\phi_q\}), \tag{2}$$

where the Fourier transform is $\phi_{\mathbf{k}} \equiv (2\pi)^{-d/2} \int d\mathbf{r}\, \phi(\mathbf{r}) e^{i\mathbf{k}\cdot\mathbf{r}}$ with d being the spatial dimension. The linear growth rate is $\omega_k = (1 - k^2)$ and the nonlinear term $N_{\mathbf{k}}$ is given by

$$N_{\mathbf{k}}\{\phi_{\mathbf{q}}\} = \frac{1}{(2\pi)^d} \int d\mathbf{q}_1 d\mathbf{q}_2 \phi_{\mathbf{k}-\mathbf{q}_1-\mathbf{q}_2} \phi_{\mathbf{q}_1} \phi_{\mathbf{q}_2}. \tag{3}$$

The linear dynamics is obtained by neglecting the nonlinear term $N_{\mathbf{k}}$, then each Fourier mode simply behaves exponentially with time

$$\phi_{\mathbf{k}}(t) = \phi_{\mathbf{k}}(0) e^{\omega_k t}. \tag{4}$$

The linear dynamics can be divided into two regimes; for k values below the critical value, $k_c = 1$, $\omega_k > 0$ and the Fourier modes grow exponentially, while for $k > k_c$, $\omega_k < 0$ and the modes decay exponentially to zero.

Binder estimated the time at which the linear theory failed by using the linear solutions for ϕ and finding the time when $\langle [\phi(\mathbf{r})]^2 \rangle$ was comparable to $\langle [\phi(\mathbf{r})]^4 \rangle$ [8]. He argued that the time when the nonlinear terms will become important is determined by the fastest growing mode. For the nonconserved order parameter the fastest growing mode is at $k = 0$. Hence the breakdown of the linear dynamics should be observable first at the largest length scales.

We argue that the linear theory will fail first for the most stable, high-wave-number, modes. To motivate this, we use an analogy with a standard multitime-scale analysis for low-dimensional dynamical systems [16]. Consider a system consisting of several modes nonlinearly coupled to each other. If there is a wide separation of time scales between the modes, the most stable, fastest modes will quickly relax to a value adiabatically determined by the slower, less stable modes; i.e., the most stable modes are slaved to the slower modes. Consider, for example, the following simple two-mode example:

$$\frac{d\phi_1}{dt} = \alpha\omega\phi_1 + \phi_2,$$

$$\frac{d\phi_2}{dt} = -\omega\phi_2 - \phi_1^3. \tag{5}$$

Here α is the ratio between the linear time scales of ϕ_1 and ϕ_2. We assume $|\alpha| \ll 1$ so that ϕ_2 relaxes on a time scale much faster than that of ϕ_1. Therefore ϕ_2 will quickly (on a time scale ω^{-1}) relax to a value determined by ϕ_1,

$$\phi_2 = -\frac{1}{\omega}\phi_1^3. \tag{6}$$

The dynamics of the slower mode on much larger time scales $(\alpha\omega)^{-1}$ becomes

$$\frac{d\phi_1}{dt} = \alpha\omega\phi_1 - \frac{1}{\alpha}\phi_1^3. \tag{7}$$

To relate this to the linear dynamics, assume that the values of ϕ_1 and ϕ_2 are both small at $t = 0$. The faster relaxing mode will stay in the linear regime for a time of order ω^{-1} while the slower mode will remain in the linear regime for times of order $(\alpha\omega)^{-1}$. Hence, in this case, the linear approximation fails first for the fastest relaxing mode.

A similar idea should hold for phase-ordering dynamics with the distinction that, due to the continuum of time scales, the separation between fast and slow modes is less clear. However, the most stable modes will still become slaved to the slower modes at lower wave number. Direct application of the slaving arguments gives

$$\langle \phi_{-\mathbf{k}}\, \phi_{\mathbf{k}} \rangle = \frac{1}{\omega_k^2} \langle N_{-\mathbf{k}}\, N_{\mathbf{k}} \rangle \tag{8}$$

for the slaved modes. Here the brackets indicate an average over initial conditions. A mode will become slaved on a time scale $\tau_k = (-\omega_k)^{-1}$. Since τ increases with decreasing k the boundary between the slaved and linear modes will move toward small wave number with time. When this boundary reaches k such that $|\omega_k|$ is approximately the maximum growth rate there is no longer a separation of time scales and the linear theory will break down for all wave numbers. Note that this minimum k is still a stable mode.

We test these ideas by observing the evolution of the structure factor for the TDGL equation [Eq. (1)] in one dimensions for the nonconserved order parameter,

$$\frac{\partial \phi(x,t)}{\partial t} = \phi(x,t) - [\phi(x,t)]^3 + \partial_x^2 \phi(x,t), \tag{9}$$

We take the initial conditions to be uncorrelated Gaussian variables so that $\langle \phi(x,0)\phi(x',0) \rangle = \Delta\delta(x - x')$. Two initial variances were examined with the data averaged over 52 initial configurations for initial amplitude $\Delta = (0.01)^2$ and 78 initial configurations for initial amplitude $\Delta = (0.1)^2$. We discretized space using a mesh size of $\delta x = 0.25$ on a lattice of $nx = 32768$ points. The update was performed using the Adam-Bashford method with $\delta t = 0.005$.

For each time step we calculated the structure factor, $S_k(t) = \langle \phi_k(t)\phi_{-k}(t) \rangle$ and compare it to the structure factor predicted by linear theory, $S_{k,\mathrm{lin}}(t)$. To eliminate any discrepancy due to discretization, the linear results were calculated using the same discretization and update schemes as used in the simulation. For this reason we plot our results with respect to the effective wave number k_{eff} defined by $k_{\mathrm{eff}}^2 \equiv \{2[1 - \cos(k\delta x)]\}/(\delta x)^2$. k takes on the values of $k = 2\pi m/L$ where $L = \delta x nx$ is the system size and m varies from $-nx/2$ to $nx/2$.

Figure 1 shows the ratio of $S_{k,\mathrm{lin}}(t)/S_k(t)$ as a function

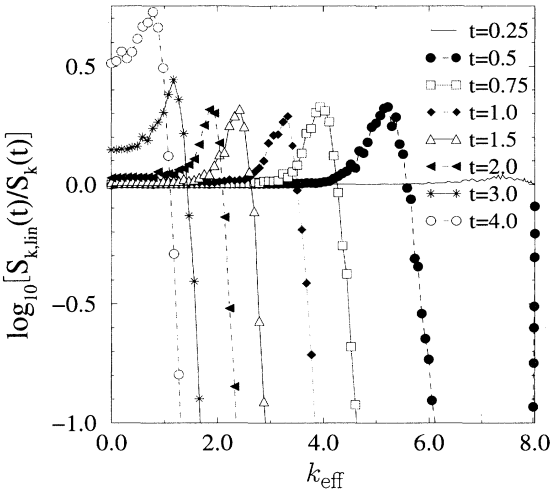

FIG. 1. The ratio $S_{k,\rm lin}(t)/S_k(t)$ where $S_{k,\rm lin}(t)$ is the linear prediction and $S_k(t)$ is the scattering intensity from the simulation. The data are for $\Delta = \langle[\phi_i(0)]^2\rangle = (0.1)^2$. At small k, $S_k(t)$ is just slightly less than $S_{k,\rm lin}(t)$. However, there is a sharp transition at $k = k_1(t)$ to a range where $S_{k,\rm lin}(t) > S_k(t)$ and finally a second crossover at $k = k_2(t)$ to a regime where $S_k(t) > S_{k,\rm lin}(t)$. The low-$k$ linear region shrinks with time.

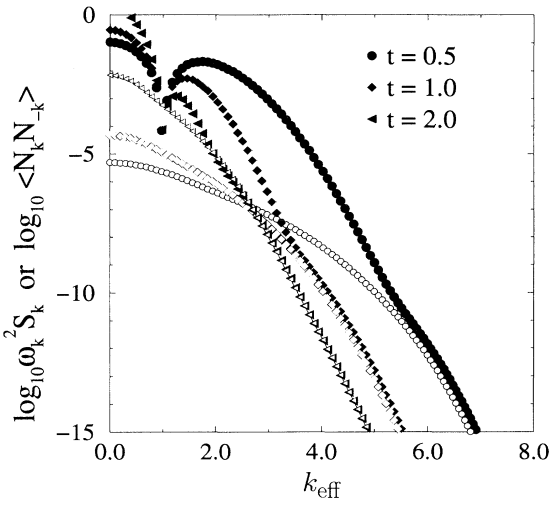

FIG. 2. Comparison of $\omega_k^2 S_k(t)$ (solid symbols) vs $\langle N_k(t)N_{-k}(t)\rangle$ (open symbols) for three different times. $\omega_k^2 S_k(t)$ and $\langle N_k(t)N_{-k}(t)\rangle$ differ at small k but coincide for large k. The range of k in which they coincide expands to smaller wave numbers with time. These quantities are equal for complete slaving.

of k for various times. For each time, we observe a range of wave numbers at small k in which $S_{k,\rm lin}(t)$ is slightly larger than $S_k(t)$ but the ratio is constant with k. For short times, the linear dynamics remains valid in this range of k up to a crossover $k = k_1(t)$. Here begins a second regime in which $S_{k,\rm lin}(t)$ is much larger then the measured scattering intensity, i.e., $S_k(t) < S_{k,\rm lin}(t)$. The ratio peaks and then decreases, crossing one at $k = k_2(t)$. For k values beyond $k_2(t)$, $S_{k,\rm lin}(t)$ is less then $S_k(t)$ and the ratio continues to decrease. Both $k_1(t)$ and $k_2(t)$ decreases with time so that the range of k for which linear theory is valid decreases with time. When $k_1(t) \approx k_c = 1$ we find that there is significant deviation from the linear prediction at all wave numbers. This occurs in this case at about $t = 3$. Figure 1 shows the results for $\Delta = \langle\phi_i^2\rangle = (0.1)^2$. Similar behavior is found for $\Delta = (0.01)^2$ except that the breakdown of the linear dynamics is delayed.

Figure 1 confirms that the linear theory breaks down at largest k first. To verify that this is due to adiabatic slaving of these modes Fig. 2 shows $\omega_k^2 S_k(t)$ and $\langle N_k(t)N_{-k}(t)\rangle$, where $N_k(t)$ is obtained directly from the simulation. The slaving hypothesis predicts that these quantities are equal for a slaved mode. Indeed we find that two quantities differ at low k but approximately coincide at high k. As time increases the region where these quantities coincide extends toward lower wave numbers.

Although Fig. 2 supports the mode-slaving hypothesis, a more careful analysis shows that the ratio $\omega_k^2\langle\phi_{\mathbf{k}}\phi_{-\mathbf{k}}\rangle/\langle N_k N_{-k}\rangle$ at higher $k_{\rm eff}$ is not exactly unity but the exact value of the ratio depends on the wave number and time. This is because the time scales of the evolution of the nonlinear term N_k are not sufficiently separated from the linear time scale, ω_k^{-1} of the relaxing modes. To correct for this effect, we note that the nonlinear term N_k in the evolution for mode k should be dominated by the higher-amplitude low-wave-number mode. From Fig. 1 there should be a regime in which mode k is no longer linear but the nonlinear coupling to that mode, N_k, is still governed by the linear behavior of the lower-wave-number modes. To see how this affects our slaving analysis we rewrite our two-mode model [Eq. (5)] assuming that the time scales of the two modes are not necessarily widely separated and that the slower mode is linear,

$$\frac{\partial\phi_1}{\partial t} = \alpha\omega\phi_1,$$
$$\frac{\partial\phi_2}{\partial t} = -\omega\phi_2 - \phi_1^3, \tag{10}$$

where α is no longer assumed to be small. This is easily solved to give

$$\phi_2(t) = \phi_2^{\rm lin}(t) + \frac{1}{\omega(3\alpha+1)}[\phi_1(t)]^3,$$

where $\phi_1(t) = \phi_1^{\rm lin}(t) = \phi_1(0)e^{-\alpha\omega t}$ and $\phi_2^{\rm lin}(t) = \phi_2(0)e^{-\omega t}$. At long times the first term is negligible so that $\phi_2(t)$ is still proportional to the nonlinear term but the constant of proportionality is $1/[\omega(3\alpha+1)]$ instead of $1/\omega$. We call this partial slaving in contrast to complete slaving when $|\alpha| \ll 1$.

Using this analogy, we assume that there is a regime in which ϕ_k is proportional to the nonlinear N_k,

$$\phi_k = \frac{\beta_k}{\omega_k} N_k\ , \tag{11}$$

where β_k does not depend strongly on time. Note that $\beta_k = 1$ for complete slaving. Substituting into the equation of motion for the structure factor gives

$$\frac{\beta_k}{2\omega_k}\frac{\partial}{\partial t}\langle N_k N_{-k}\rangle = \beta_k\langle N_k N_{-k}\rangle - \langle N_k N_{-k}\rangle, \tag{12}$$

or

$$\beta_k = \omega_k\left(\omega_k - \frac{1}{2\langle N_k N_{-k}\rangle}\frac{\partial}{\partial t}\langle N_k N_{-k}\rangle\right)^{-1}. \tag{13}$$

Since the nonlinear term N_k is dominated by the linear large-amplitude lower-k modes, we can substitute the linear forms for ϕ_q in N_k,

$$\begin{aligned}\langle N_{-k,\mathrm{lin}}(t)N_{k,\mathrm{lin}}(t)\rangle &= \left(\frac{1}{2\pi}\right)^2\int dk'dk''\int dq'dq''\; e^{(\omega_{k-k'-k''}+\omega_{k'}+\omega_{k''}+\omega_{k-q'-q''}+\omega_{q'}+\omega_{q''})t}\\ &\quad\times\langle\phi_{k-k'-k''}(0)\phi_{k'}(0)\phi_{k''}(0)\phi_{-k+q'+q''}(0)\phi_{-q'}(0)\phi_{-q''}(0)\rangle,\\ &= 6\Delta^3\left(\frac{1}{2\pi}\right)^2\left[\left(\int dk' e^{2\omega_{k'}t}\right)^2 e^{2\omega_k t} + \int dk'\int dk''\; e^{2(\omega_{k-k'-k''}+\omega_{k'}+\omega_{k''})t}\right],\end{aligned} \tag{14}$$

where the subscript lin denotes that we have used the linear forms of ϕ_k to estimate the nonlinear terms. This replacement corresponds to a second-order expansion in the nonlinearity. Since $\omega_k = 1 - k^2$ the integrals over k are Gaussian integrals and

$$\langle N_{k,\mathrm{lin}}(t)N_{-k,\mathrm{lin}}(t)\rangle = \left(\frac{3\Delta^3}{8\pi t}\right)\left[3e^{2(2\omega_0+\omega_k)t} + 2\sqrt{3}e^{6\omega_{k/3}t}\right]. \tag{15}$$

For the stable modes the first term is negligible relative to the second if $k^2 t > 1$ and we get

$$\langle N_{k,\mathrm{lin}}(t)N_{-k,\mathrm{lin}}(t)\rangle \approx \left(\frac{3\sqrt{3}\Delta^3}{4\pi t}\right)e^{(6\omega_{k/3})t}. \tag{16}$$

Hence the nonlinear term $N_{k,\mathrm{lin}}$ is dominated by the $q = k/3$ modes. These modes remain linear at the time the k mode first becomes nonlinear, thus justifying the use of the linear forms to estimate N_k. Substituting into Eq. (13) gives

$$\beta_k = \frac{\omega_k}{\omega_k - 3\omega_{k/3} + (2t)^{-1}}. \tag{17}$$

Note that the assumption that that β_k is independent of time is not correct, however, this dependence is negligible for large, k. Note also that in the limit of large k $\beta_k \to 3/2$ [17].

Figure 3 shows a plot of the ratio $\beta_k^2\langle N_k N_{-k}\rangle/[\omega_K^2 S_k(t)]$ for $\Delta = \langle[\phi_i(0)]^2\rangle = (0.1)^2$. Here $\langle N_k N_{-k}\rangle$ is obtained from the simulation but we use Eq. (17) for β_k. From the plot, there is a range of k in which this ratio is unity. This range becomes smaller with time since the ratio is unity when the $k/3$ mode is linear. Comparison with Fig. 1 shows that the ratio $\beta_k^2\langle N_k N_{-k}\rangle/S_k(t)$ first becomes significant at the value of k when $S_k(t)$ first deviates from $S_{k,\mathrm{lin}}(t)$ and the plateau region begins at the value of k at which $S_{k,\mathrm{lin}}(t)/S_k(t)$ passes through one. For $S_k(t)$ much larger than the plateau region $S_k(t) \approx \langle N_k N_{-k}\rangle/\omega_k^2$. This is because, in this regime, the dynamics of the $k/3$ mode will not be linear but will be adiabatically slaved to the even slower $k/9$ modes (for $k > 9$). Hence, there is a larger separation of time scales and a more complete slaving holds.

We have presented qualitative evidence that the breakdown of the linear regime is determined by partial slaving; however, a more quantitative measure is desirable. We define a breakdown time at which a particular k mode no longer obeys linear theory using three criteria from the ratio of $S_{k,\mathrm{lin}}(t)/S_k(t)$ (see Fig. 1): $t_{\mathrm{dev}}(k)$, the time at which $S_{k,\mathrm{lin}}(t)/S_k(t)$ first varies by more than 2.5% from the ratio at $k = 0$, t_{peak}, the time at which $S_{k,\mathrm{lin}}(t)/S_k(t)$ is largest and t_{unity}, the time at which $S_{k,\mathrm{lin}}(t)/S_k(t)$ again passes unity. These values were obtained from Fig. 1 using a cubic fit around the peak. To compare with our partial slaving analysis we define t_δ as the time at which the partial slaving prediction for $S_k(t)$ is a factor δ of the $S_{k,\mathrm{lin}}(t)$,

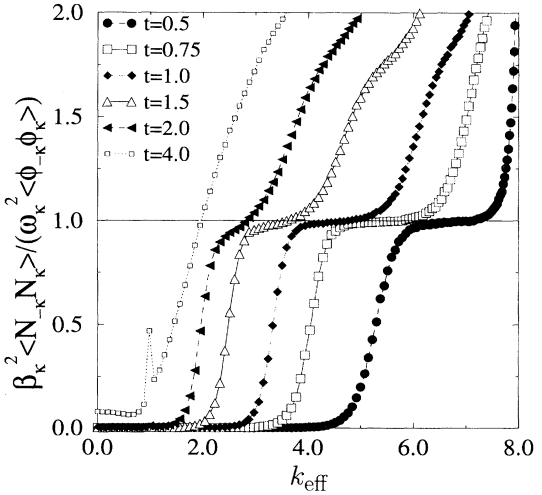

FIG. 3. Plot of $\beta_k^2\langle N_k N_{-k}\rangle/[\omega_k^2 S_k(t)]$ for various times. There is a plateau region where this ratio is unity. This plateau shrinks and moves toward lower k with time. The lower k limit of the plateau corresponds to k mode where the linear theory first breaks down. The upper k limit corresponds to where the $k/3$ modes starts to deviate from the linear dynamics.

$$\frac{\beta_k^2}{\omega_k^2}\,\langle N_{k,\mathrm{lin}}(t_\delta)N_{-k,\mathrm{lin}}(t_\delta)\rangle = \delta S_{k,\mathrm{lin}}(t_\delta). \tag{18}$$

We used $\delta = 1$ and $\delta = 0.01$ in our comparison.

Figure 4 shows that $t_{0.01}$ coincides very well with t_{dev}. The goodness of the fit depends on the value of δ. However, varying δ by as much as a factor of two has a much smaller effect on the goodness of the fit then, for example, using a criteria based on $S_{k,\mathrm{lin}}(t)$ and $\langle N_k N_{-k}\rangle$ without the β_k^2 factor. On the other hand, t_1 corresponds exactly with t_{peak}. Figure 5 shows the same analysis for the smaller $\Delta = \langle[\phi_i(0)]^2\rangle = (0.01)^2$. We also find excellent agreement between t_1 and t_{peak} and $t_{0.01}$ and t_{dev}. The difference is that, in this case, the linear dynamics hold for a longer time for all Fourier modes.

Defining the breakdown time as $t_{\mathrm{br}}(k) = t_1(k)$ we find, in the limit of small Δ,

$$t_{\mathrm{br}}(k) \approx -\frac{3\ln\Delta}{2(3+k^2)}\ , \tag{19}$$

which at large k becomes

$$t_{\mathrm{br}}(k) \sim -\frac{3\ln\Delta}{2k^2}. \tag{20}$$

So that $t_{\mathrm{br}}(k)$ increases with decreasing Δ and decreasing k. We emphasize that this holds for very small Δ and large k. Care should be taken in using this asymptotic result. For our simulations, for example, the finite k and Δ corrections are important and the asymptotic form would not lead to a very good fit to $t_{\mathrm{br}}^{\mathrm{expt}}$.

Finally we discuss the relation between our analysis and earlier results. Our analysis is consistent with the simulations of the long-range Ising model by Gross and co-workers [13,14] who found that the linear theory first breaks down at wave numbers $k \approx k_c$ and larger. Similar behavior was also observed in polymer and alloy experiments [12,18]. On the other hand, earlier experiments and simulations have concentrated on the low-wave-number behavior and therefore have not focused on the k-dependent breakdown discussed here [9,11,10]. When the linear theory was observed to fail first at short length scales [9] this was attributed to thermal noise or to the invalidity of the coarse-grained description on these scales. Our analysis indicates that there is a third possibility for this failure due to slaving of the high-wave-number modes.

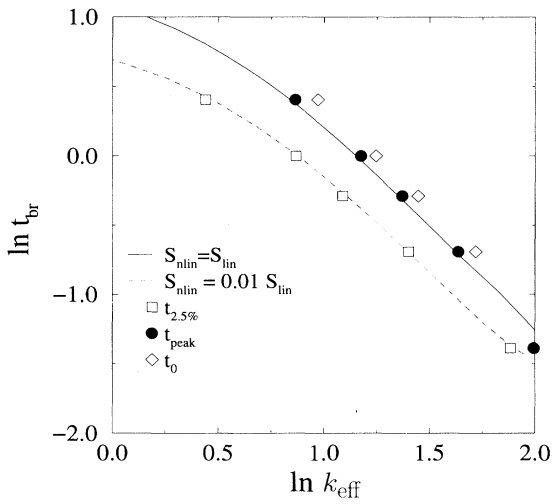

FIG. 4. The breakdown time $t_{\mathrm{br}}(k)$ for $\Delta = \langle[\phi_i(0)]^2\rangle = (0.1)^2$. The empirical breakdown times are defined as follows: t_{dev} (□): the k modes at which $S_k(t)/S_{k,\mathrm{lin}}(t)$ deviates 2.5% from the value at $k = 0$. t_{peak} (•) the k mode at which $S_{k,\mathrm{lin}}(t)/S_k(t)$ reaches a maxima and t_{unity} (◇) the k modes at which the $S_{k,\mathrm{lin}}(t)/S_{k,t}$ passes through one. This is compared to the theoretical values t_δ for $\delta = 0.01$ (dashed line) and $\delta = 1.0$ (solid line). Very good agreement is found between t_{dev} and $t_{0.01}$ and between t_{peak} and t_1.

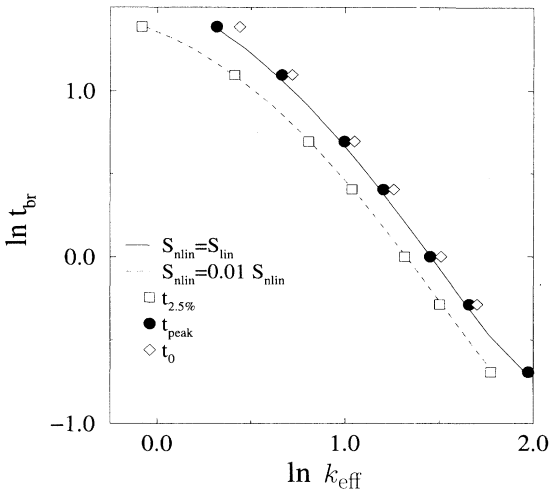

FIG. 5. Same as Fig. 4 but for a smaller amplitude initial condition, $\Delta = \langle[\phi_i(0)]^2\rangle = (0.01)^2$. The behavior is similar to $\Delta = (0.1)^2$ but the linear theory holds for a longer time for any particular k.

To compare with other theoretical results we note that the initial regime during which the modes of wave number $k > k_c$ becomes slaved corresponds to the diffusive relaxation regime discussed by Corberi, Coniglio, and Zannetti [15]. This leads to a reduction in $\langle[\phi(x,t)]^2\rangle$. Note also that since we replaced ϕ_q by their linear values in $N_k(t)$ we are effectively performing a second-order expansion in the nonlinearity. Most previous works have discussed a first-order or Gaussian expansion [11,19,20]. An analysis using this truncation would not lead to a strong k dependence in the breakdown time. To see this, we consider the Gaussian approximation [19]. The evolution equation for the scattering intensity is

$$\frac{\partial\langle\phi_k(t)\phi_{-k}(t)\rangle}{\partial t} = \omega_k\langle\phi_k(t)\phi_{-k}(t)\rangle + (2\pi)^{-1/2} \times \int dq_1 dq_2\ \langle\phi_{k-q_1-q_2}\phi_{q_1}\phi_{q_2}\phi_{-k}\rangle.$$

In the Gaussian approximation ϕ_k are assumed to be Gaussian so that the nonlinear term can be decoupled to give

$$\frac{\partial S_k(t)}{\partial t} = \left(\omega_k - 3(2\pi)^{1/2}\int dq S_q(t)\right) S_k(t)$$
$$= \left(\omega_k - 3\langle[\phi(x)]^2\rangle\right) S_k(t),$$

where $\langle[\phi(x)]^2\rangle$ must be obtained self-consistently. The correction to the linear coefficient does not depend on k and therefore does not give any k dependence in the breakdown time. Note also that the first-order expansion always leads to a reduction in the scattering intensity relative to the linear growth. We observed that at very small wave numbers that this is indeed the case but that this effect is small.

Finally, although we have discussed the results for the nonconserved order-parameter case only, many of our arguments, for example, slaving and partial slaving, also hold for conserved systems. However, the actual detailed integrations may be more difficult. A more important omission may be the neglect of thermal noise in our analysis. There is an intricate interplay between the initial fluctuation amplitude Δ and the thermal noise. In some regimes the thermal noise will be dominant and we expect it will have important effects on our mode-slaving analysis.

To summarize, we have demonstrated that the linear theory of phase-ordering dynamics breaks down first for the highest-wave-number modes. This is due to the fact that the stable high-wave-number modes become adiabatically slaved to the higher-amplitude less-stable modes. We showed that $t_{\rm br}(k)$, the breakdown time as a function of wave number k, can be understood in terms of partial slaving. In particular, we find that the breakdown time in the limit of small initial variance Δ and large k behaves as $t_{\rm br} \sim -3\ln\Delta/(2k^2)$. Our theoretical predictions are confirmed by numerical simulations.

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