Bounds on the decay of the autocorrelation in phase ordering dynamics

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We investigate the decay of temporal correlations in phase ordering dynamics by obtaining bounds on the decay exponent λ of the autocorrelation function [defined by $\lim_{t_2 \gg t_1} \langle \phi(\mathbf{r},t_1)\phi(\mathbf{r},t_2)\rangle \sim L(t_2)^{-\lambda}$]. For a nonconserved order parameter, we recover the Fisher and Huse inequality, $\lambda \geq d/2$. For a conserved order parameter, we find $\lambda \geq d/2$ only if $t_1 = 0$. If t_1 is in the scaling regime, then $\lambda \geq d/2+2$ for $d \geq 2$ and $\lambda \geq 3/2$ for $d=1$. For the one-dimensional scalar case, this, in conjunction with previous results, implies that the value of λ depends on whether $t_1 = 0$ or $t_1 \gg 1$. Our numerical simulations for the two-dimensional, conserved scalar order parameter show that $\lambda \approx 4$ for t_1 in the scaling regime, consistent with our bound. The asymptotic decay when $t_1 = 0$, while exhibiting an unexpected sensitivity to the amplitude of the initial correlations, is slower than when $t_1 \gg 1$ and obeys the bound $\lambda \geq d/2$.

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Phase separation dynamics proceeds when a system is quenched from its high temperature, homogeneous phase to a low temperature, inhomogeneous phase (where several phases coexist in equilibrium). Due to its simple description yet rich behavior, phase ordering dynamics has greatly enhanced our understanding of nonequilibrium processes [1]. At late times, the spatial distribution of domains can be described by a single time-dependent length, $L(t)$, which typically grows algebraically in time, $L(t) \sim t^{1/z}$. This is reflected in a scale invariant equal-time correlation function $C(r,t)$. More recently it has been realized that the temporal decay of the correlations and the response to past perturbations are scale covariant. In particular, the asymptotic decay of the two-time autocorrelation function, $C(r,t_1,t_2) = \langle \phi(\mathbf{r},t_1)\phi(\mathbf{0},t_2)\rangle$, defines an independent exponent λ via $\lim_{t_1 \ll t_2} C(0,t_1,t_2) \sim [L(t_1)/L(t_2)]^{\lambda}$. This exponent, which is dependent on the spatial dimension d, bears no relation to z and so its value provides a sensitive test for approximate theories of phase ordering kinetics [2–8]. Although the autocorrelation function has been studied extensively for nonconserved order parameter (NCOP) dynamics [2,4–10], there has been much less discussion of the conserved order parameter (COP) case [11–14]. In a recent publication, Majumdar *et al.* have shown both numerically and analytically that $\lambda = 1$ for the one-dimensional, scalar COP with $t_1 = 0$ [12].

In this paper, we obtain bounds on the decay of the autocorrelation function. For nonconserved order parameters, we find $\lambda \geq d/2$ independent of the first time t_1, consistent with a general argument of Fisher and Huse [2]. However, for conserved order parameters, we find that the bound depends on the choice of t_1. Specifically, $\lambda \geq d/2$ for $t_1 = 0$ (assuming the quench is from a high temperature phase), but, for t_1 in the scaling regime, $\lambda \geq d/2+2$ for $d \geq 2$ and $\lambda \geq 3/2$ for $d=1$. This difference arises from the small k behavior of the scattering intensity $S(k,t_1)$. In conjunction with the exact result for λ, for the one-dimensional scalar model, with $t_1 = 0$ [12], we conclude that for $d=1$, λ depends on whether $t_1 = 0$ or $t_1 \gg 1$. To carry out the investigation in higher dimensions, we perform an extensive numerical integration of the Cahn-Hilliard equation [see Eq. (4) below] in $d=2$. For $t_1 = 0$ we find quite surprisingly, that the asymptotic value of λ is extremely sensitive to initial conditions. We find that $\lambda = 2$, when the amplitude of the initial correlations is large, while $\lambda \approx 3$ (with a possibility of a slower decay at late times) when the amplitude is small. On the other hand, when t_1 is in the scaling regime, there is no dependence on the initial state, and $\lambda \approx 4$. We also derive bounds on λ for quenches to and from the critical point. Moreover, our results easily extend to vector order parameters.

We begin by obtaining the lower bounds on λ. The equal point autocorrelation $C(t_1,t_2) \equiv C(\mathbf{0},t_1,t_2)$ is related to the k space autocorrelation $S(k,t_1,t_2)$ by

$$C(t_1,t_2) = \int d\mathbf{k} \langle \delta\phi_{\mathbf{k}}(t_1)\delta\phi_{-\mathbf{k}}(t_2)\rangle = \int d\mathbf{k} S(k,t_1,t_2).$$

Here $\phi(\mathbf{r},t)$ is the order parameter at point $\mathbf{r}$ and time t and $\delta\phi(\mathbf{r},t) \equiv \phi(\mathbf{r},t) - m_0$ with $m_0 = V^{-1}\int d\mathbf{r}\phi(\mathbf{r},t)$ and the Fourier transform $\delta\phi_{\mathbf{k}}(t) \equiv V^{-1/2}\int d\mathbf{r} e^{-i\mathbf{k}\cdot\mathbf{r}}\delta\phi(\mathbf{r},t)$. The angular brackets indicate an average over initial conditions. Note that for a critical quench $\langle m_0\rangle = 0$ and $\langle m_0^2\rangle$ is $\mathcal{O}(V^{-1})$, whereas for an off-critical quench $\langle m_0\rangle$ is $\mathcal{O}(1)$.

Using the Cauchy-Schwartz inequality, we find

$$\begin{aligned} C(t_1,t_2) &\leq \int d\mathbf{k} \langle \delta\phi_{\mathbf{k}}(t_1)\delta\phi_{-\mathbf{k}}(t_1)\rangle^{1/2} \\ &\quad \times \langle \delta\phi_{\mathbf{k}}(t_2)\delta\phi_{-\mathbf{k}}(t_2)\rangle^{1/2} \\ &\sim \int d\mathbf{k} S(k,t_1)^{1/2} S(k,t_2)^{1/2}, \end{aligned} \tag{1}$$

where $S(k,t) = S(k,t,t)$.

Now assume t_2 to be in the scaling regime with $t_2 > t_1$. At late times, the scattering is due to the sharp interfaces or defects. The k modes $\delta\phi_{\mathbf{k}}$ at times t_1 and t_2 will be uncor-

related when the interfaces move a distance greater than $2\pi/k$, so that $S(k,t_1,t_2)$ decreases rapidly for $k(L(t_2)-L(t_1))\gg 1$. The upper limit of the integral over k in Eq. (1) can then be cut off at $2a\pi/L(t_2)$ where a is a constant of $\mathcal{O}(1)$. This corresponds to assuming that $S(k,t_1,t_2)$ decays faster than $[kL(t_2)]^{-d}$ for $t_2\gg t_1$. For $L(t_2)\gg L(t_1)$, only the small k behavior of $S(k,t_1)$ contributes to the integral. Assume that $\lim_{k\to 0}S(k,t_1)\sim k^{\beta}$ ($\beta\geq 0$). For quenches to zero temperature, $S(k,t_2)$ will have the scaling form $S(k,t_2)=L(t_2)^d f(kL(t_2))$. Substituting into Eq. (1) (with the appropriate limits of integration), gives

$$\lim_{t_2\gg t_1} C(t_1,t_2)\sim L(t_2)^{-\lambda}$$

$$\leq L(t_2)^{d/2}\int_0^{2a\pi/L(t_2)} dk k^{d-1}k^{\beta/2}f(kL(t_2)),$$

$$\sim L(t_2)^{-(d+\beta)/2}.$$

This immediately gives a lower bound on λ,

$$\lambda\geq\frac{\beta+d}{2}.$$

The argument just presented is very general and holds for conserved and nonconserved, scalar and vector order parameters.

We now consider specific dynamical scenarios. Let T_I and T_F be the temperatures of the initial and final states, respectively. We first focus on quenches from the high temperature phase ($T_I=\infty$) to zero temperature ($T_F=0$). Since the initial state is disordered, $\lim_{k\to 0}S(k,0)\sim k^0$. This is true irrespective of the dynamics. Let us now consider a dynamics where the order parameter is not conserved. In this case, $\lim_{k\to 0}S(k,t_1)\sim k^0$ for both $t_1=0$ and t_1 in the scaling regime, and therefore $\beta=0$. This implies that for nonconserved dynamics,

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

This inequality was also obtained by Fisher and Huse using general scaling arguments [2] and is consistent with all results to date [4,5,7–10].

When the dynamics is such that the order parameter is conserved, then the bounds on λ depend on whether t_1 is zero or is in the scaling regime. Clearly, as before, $\lim_{k\to 0}S_k\sim k^0$ when $t_1=0$ and $\beta=0$. However, when t_1 is in the scaling regime, $\lim_{k\to 0}S(k,t)\sim k^4$ for $d\geq 2$ [15] and so $\beta=4$. For $d=1$, the dynamics is dominated by thermal noise and Majumdar *et al.* find that $\lim_{k\to 0}S(k,t)\sim k^2$ [12], so that $\beta=2$ for $d=1$. Therefore, for a conserved order parameter and t_1 in the scaling regime, we have

$$\lambda\geq\frac{d}{2}+2 \text{ if } d\geq 2 \quad\text{and}\quad \lambda\geq\frac{3}{2} \text{ if } d=1. \tag{3}$$

These bounds suggest that the exact value of the asymptotic exponent *may* in fact depend on whether t_1 is in the scaling regime or not, but of course do not rule out the possibility that the exponent is independent of t_1. However, for $d=1$, Majumdar *et al.* find analytically and numerically that $\lambda=1$ for $t_1=0$, while our bound suggests that $\lambda\geq 3/2$ for t_1 in the scaling regime [16]. Thus at least in $d=1$, it is clear that the value of λ depends on whether t_1 is in the scaling regime or not.

For vector fields (with m, the number of components of the order parameter, >2), an argument analogous to Ref. [15], gives the same $\lim_{k\to 0}S(k,t)\sim k^4$. This is supported by an extensive numerical integration of the Cahn-Hilliard equation [17]. Therefore the lower bounds on λ derived above are valid even for vector order parameters with $m>2$.

Quenches from the critical point ($T_I=T_c, T_F=0$) lead to long-range correlations of the initial configurations. In this case, $\lambda\geq d/2$ no longer holds. More generally if $S(k,0)\sim k^{-\sigma}$ we obtain $\lambda\geq(d-\sigma)/2$. (For critical dynamics $\sigma=2-\eta$, where η is the static critical exponent.) This is consistent with the result of Bray *et al.*, who found that, for nonconserved order parameter, $\lambda=(d-\sigma)/2$ for σ greater than a critical value σ_c [6].

Analysis of the bounds on the autocorrelation exponent for quenches *to* the critical point ($T_I=\infty, T_F=T_c$) has to start afresh from Eq. (1). Since t_2 is in the critical point scaling regime, the correlation function has the following scaling form: $S(k,t_2)\sim k^{-2+\eta}f_c(kL(t_2))$. Substituting this form into Eq. (1) gives $\lambda\geq(2d-2+\eta+\beta)/2$. Therefore, when $t_1=0$ we get $\lambda\geq(2d-2+\eta)/2$. When t_1 is also in the scaling regime, the bound on λ depends on the behavior of the scaling function $f_c(kL(t_1))$ as $kL(t_1)\to 0$. For nonconserved systems $\lim_{x\to 0}f_c(x)\to$const, or $\beta=-2+\eta$, leading to $\lambda\geq d-2+\eta$.

We test the above ideas and fix the value of λ using numerical simulations. As previously mentioned, exact analytical results and numerical computations on the one-dimensional scalar model have been carried out only for the case when $t_1=0$. In higher dimensions, however, the numerical results are not very conclusive [11]. We therefore compute the asymptotic value of λ by numerically integrating the Cahn-Hilliard equation in two dimensions,

$$\frac{\partial\phi(\mathbf{r},t)}{\partial t}=\nabla^2\mu(\mathbf{r},t), \tag{4}$$

where $\mu=-\phi+\phi^3-\nabla^2\phi$. We have used an Euler discretization with $\delta t=0.1$ and $\delta x=1.09$ and periodic boundary conditions. We discretize the Laplacian as

$$\nabla^2\phi_{i,j}=\frac{1}{\delta x^2}\frac{\sqrt{2}}{1+\sqrt{2}}\left[\frac{1}{2}\sum_{\text{NNN}}+\sum_{\text{NN}}-6\right]\phi_{i,j}.$$

This choice decreases lattice anisotropy effects and allows a larger δt before the onset of the checkerboard instability [18]. Decreasing δt has no effect on the numerical results, while increasing δx to 1.32 results in pinning effects that lead to a slower decay of the autocorrelation function at late times (even though the effect on the single-time behavior is less apparent). We solve this dynamical equation subject to $\phi_{i,j}(t=0)$ being uncorrelated random variables with variance Δ. We work on a finite lattice of size $n_x n_y$ (the number of lattice points in the x and y directions respectively) and calculate physical quantities (such as the energy density and correlation functions) averaged over several realizations of

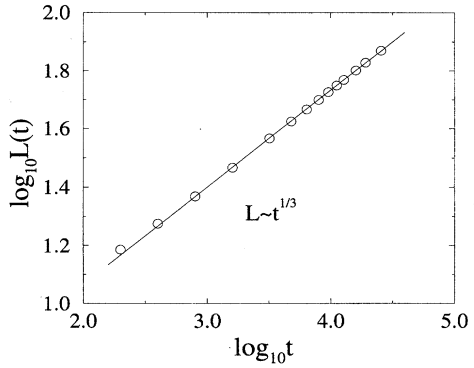

FIG. 1. Characteristic domain size (defined as the inverse of the interfacial density) vs time for $\Delta=0.25$. A very good $L(t)\sim t^{1/3}$ behavior is found for times $t>400$.

initial conditions. For $\Delta=0.03$ our lattice sizes range from $n_x=n_y=64$ (averaged over 3084 initial configurations), 256 (1120 configurations), and 1024 (42 initial configurations). For $\Delta=0.25$ we used $n_x=n_y=256$ (650 initial configurations) and 1024 (35 configurations).

We have used the interfacial area density as a measure of the characteristic length scale $L(t)$. Operationally, this is defined as $(2\delta x n_x n_y)/n_{opp}$, where $n_x n_y$ is the total number of lattice sites and n_{opp} is the number of sites whose nearest neighbor has a ϕ of opposite sign. Figure 1 shows that we recover the standard result that $L(t)$ grows as $t^{1/3}$ for all $t>200$ for $\Delta=0.25$. Other measures of the characteristic length scale, such as the first zero of the real space correlation function, also behave in the same manner (for $t>200$). To check for the single time scaling $S(k,t)=L(t)^d f(kL(t))$ we plot, in Fig. 2, the scaled scattering intensity, $L(t)^{-2}S(k,t)$ vs $kL(t)$ for times from $t=200$ to $t=51\,200$. Scaling is evident at later times and there is a clear regime at

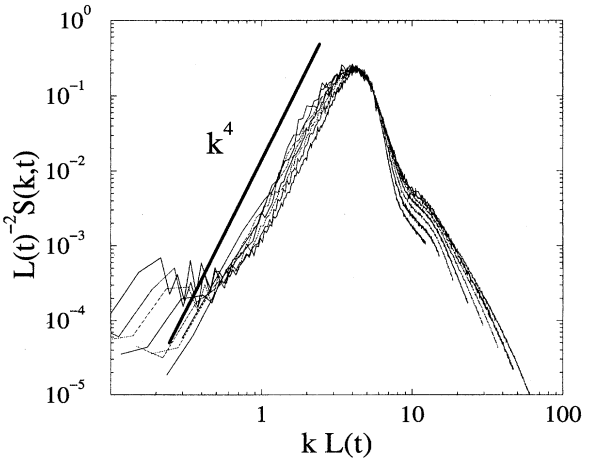

FIG. 2. Scaled scattering intensity for $\Delta=0.25$. Times $t=200$, 400, 800, 1600, 3200, 6400, 12 800, and 25 600 are shown. Although the earliest times do not yet show good scaling, even for $t=200$ there is a clear regime where $S(k)\sim k^4$ for small k.

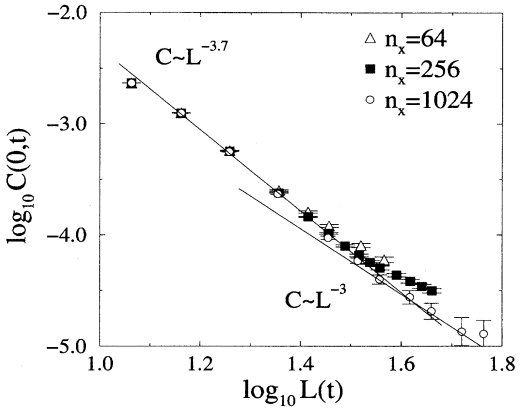

FIG. 3. Autocorrelation function $C(0,t)$ for $n_x=n_y=64$, 256, and 1024 for $\Delta=0.03$. The asymptotic decay is very difficult to extract in this case. There is a regime of about 1/2 a decade in L or 3/2 decades in time, where the correlation function decays as $L^{-3.7}$. However, there are indications of a crossover to a slower rate decay at later times but it is difficult to differentiate this from finite size effects which also produce a crossover to a slower decay.

low k where $S(k)\sim k^4$ at these times. Thus our bound on λ with $\beta=4$ should be operative when $t_1\geq 200$.

We have found that it is extremely difficult to extract an unambiguous value for the decay exponent when $t_1=0$, since the two-time correlation function is surprisingly sensitive to the magnitude of the initial correlations (the variance Δ) and to finite size effects. Figure 3 shows the two-time autocorrelation function for the smaller variance $\Delta=0.03$ with $n_x=64$, 256, and 1024, while Fig. 4 shows the same for $\Delta=0.25$ and $n_x=256$ and 1024. The behavior for the smaller variance $\Delta=0.03$ is especially complicated. We find that $C(0,t)\sim L^{-3.7}$ for approximately a half decade in L, which corresponds to about one-and-a-half decades in time. There is a definite crossover to a slower decay at later times with

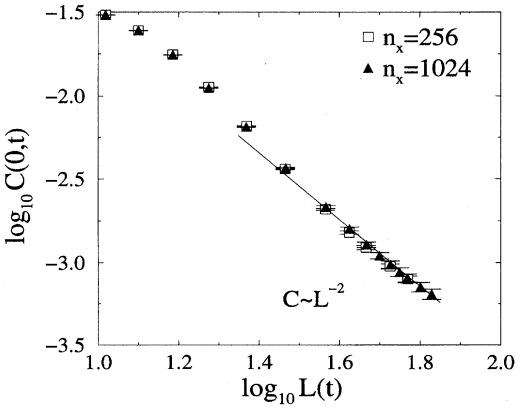

FIG. 4. Autocorrelation function $C(0,t)$ for $n_x=n_y=256$ and 1024 for $\Delta=0.25$. In this case we do not find strong finite size effects. The autocorrelation function decays at a slower rate in this case than for $\Delta=0.03$. We find that $C(0,t)$ behaves approximately as L^{-2}.

$C(0,t) \sim L^{-3.0}$ for approximately half a decade in time. There are indications of a further slowing down in the decay at even later times. However, this slower decay seems to be a finite size effect as the crossover to slower decay occurs earlier for smaller system sizes. Thus for $\Delta = 0.03$, we would hesitate to provide an unambiguous value for the asymptotic decay exponent.

As is clear from Fig. 4 however, an increase in the variance to a value $\Delta = 0.25$ yields much less ambiguous results. The correlation function in this case decays much slower than for $\Delta = 0.03$ and the slope shows lesser variation. We find that at late times, $C(0,t) \sim L^{-2}$ obtains a good fit for a little more than a 1/3 decade in $L(t)$ or for more than one decade in time. Moreover, we do not see strong finite size effects; the result for $n_x = n_y = 256$ shows no systematic deviation from the result for $n_x = n_y = 1024$. Note that in spite of this strong sensitivity to Δ, the decay exponent is consistent with the bound $\lambda \geq d/2$ *for either value* of Δ.

This sensitivity to initial conditions is, at least to us, unexpected. We do not have a detailed understanding of this phenomenon, and so we offer a conjecture as to how this could arise. It is clear that the linear dynamics regime of the Langevin equation (when there is an exponential growth of the structure function), holds for a longer time when the variance of the initial correlations is smaller. In this linear regime the scattering intensity will be strongly peaked at the fastest growing Fourier mode $k = k_{max}$. The domains emerging from this linear regime, will almost entirely have a curvature of k_{max}^{-1} and so the autocorrelation function will be determined for a very long time by the autocorrelation at $k = k_{max}$. Only when the amplitude at k_{max} has decreased sufficiently, will the two-time correlation be determined by the behavior at low wave numbers. On the other hand, when the dynamics proceeds from initial conditions with a high variance, e.g., $\Delta = 0.25$, the linear dynamics regime is almost nonexistent. Domains form and grow almost immediately and hence the correlations will not be dominated by one wave number.

The strong effects of finite size seen when the initial variance is small are also surprising since finite size effects on single-time quantities become important when $L(t)$ is of the order of the lattice dimension, L_0. Thus the usual length scales extracted from single-time quantities are identical for $n = 256$ and $n = 1024$ at these late times. However, since $C(0,t)$ decays rapidly with t, any small systematic effect becomes increasingly relevant as t increases. Clearly, finite size effects on $C(0,t)$ may be important (though not necessarily so) when the spread in $C(0,t)$ is of the same order as $C(0,t)$. The spread in $C(0,t)$ decreases as $L_0^{-d/2}$ and, based on our simulations, depends only weakly on $L(t)$. Hence finite size effects can become important when $C(0,t) \sim L(t)^{-\lambda} \sim L_0^{-d/2}$, i.e., much earlier than for single-time quantities. This may account for why finite size effects are observed when the variance Δ is small. For the larger variance this effect is not evident: this is because the autocorrelation function decays much slower for large initial variance, and then the mean value is always found to be larger than the spread during our simulations.

On the other hand, the autocorrelation function is very robust with respect to initial conditions, when t_1 is in the scaling regime. Figure 5 shows $C(t_1,t_2)$ vs $L(t_2)/L(t_1)$ for $t_1 = 100$, 200, and 400 with $\Delta = 0.25$ and for $t_1 = 200$ with $\Delta = 0.03$. The figure shows results for the largest employed lattice size, since we did not find strong finite size effects for either value of Δ. A fairly good collapse of the data for these times demonstrates a good fit to the scale covariant form for these values of t_1. For $\Delta = 0.25$ and $t_1 = 200$ we find that the autocorrelation function decays as $(L_2/L_1)^{-4}$ for over half a decade in L_2 or 1.5 decades in time. Hence, we find a late-time regime in which $\lambda \approx 4$ [16] consistent with our bound that $\lambda \geq 3$ when $\beta = 4$ and $d = 2$.

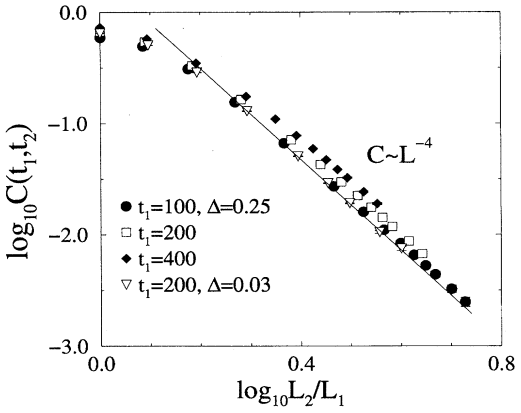

FIG. 5. $C(t_1,t_2)$ vs $L(t_2)/L(t_1)$ for $t_1 = 200$, 400, and 800 for $\Delta = 0.25$ and for $t_1 = 200$ for $\Delta = 0.03$. We find a very good L^{-4} decay in this case.

Recently Marko and Barkema [14] performed a Monte Carlo simulation on a kinetic Ising model using an algorithm that accelerated the diffusion of spins in the bulk relative to that along the interface, so that they could access the late-time regimes. They found that the correlation function does decay with a larger power when t_1 is in the scaling regime compared to when $t_1 = 0$. However, they found that $\lambda \approx 3$ in two dimensions, which, though consistent with our bound, is not in agreement with our simulations for the two-dimensional Cahn-Hilliard equation. For $d = 3$ they found $\lambda \approx 2.5$, which would violate our bound of $\lambda \geq 3.5$ (using $\beta = 4$ and $d = 3$) . However, it was found from their simulations that the small k behavior of the scattering intensity seems to behave as $k^{2.2}$ rather than as k^4 seen in the Cahn-Hilliard simulations and polymer experiments. If we use a $\beta = 2.2$, then our bound in three-dimensions reads $\lambda \geq 2.6$, which is consistent, to within numerical errors, with their results. Further work will be necessary to understand the difference between the kinetic Ising model and the coarse-grained description.

Having provided useful lower bounds on λ, we now ask whether is it possible to bound λ from above. Unfortunately, we have not been able to provide useful upper bounds. However, we note that the bound [Eq. (3)] as well as our numerical results violate the upper bound conjectured by Fisher and Huse, $\lambda \leq d$ [2]. As they originally noted, their conjecture contains many assumptions. Moreover, inasmuch as their ar-

gument is aimed at the decay of the magnetization, their conjecture has validity only when the order parameter is *not* conserved and when $t_1=0$ [so that $S(k,t_1)\sim k^0$].

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[16] $S(k,t)\sim k^4$ holds exactly only as $t\to\infty$. Therefore the second bound $\lambda\geq(d+4)/2$ holds for $\lim_{t_2/t_1\to\infty}\lim_{t_1\to\infty}$. As shown in Fig. 2 at any finite time there will be a regime at small k in which $S(k,t)\sim k^0$. Hence, for large but finite t_1 there is a large range of t_2/t_1 for which the bound holds with the range increasing with t_1.
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