Droplet model for autocorrelation functions in an Ising ferromagnet

Chao Tang, Hiizu Nakanishi, and J. S. Langer
Institute for Theoretical Physics, University of California, Santa Barbara, California 93106
(Received 8 March 1989)

The autocorrelation function, $C(t)=\langle S_i(0)S_i(t)\rangle-\langle S_i^2(0)\rangle$, of Ising spins in an ordered phase ($T<T_c$) is studied via a droplet model. Only noninteracting spherical droplets are considered. The Langevin equation for droplet fluctuations is studied in detail. The relaxation-rate spectra for the corresponding Fokker-Planck equation are found to be (1) continuous from zero for dimension $d=2$, (2) continuous with a finite gap for $d=3$, and (3) discrete for $d\geq 4$. These spectra are different from the gapless form assumed by Takano, Nakanishi, and Miyashita for the kinetic Ising model. The asymptotic form of $C(t)$ is found to be exponential for $d\geq 3$ and stretched exponential with the exponent $\beta=\frac{1}{2}$ for $d=2$. Our results for $C(t)$ are consistent with the scaling argument of Huse and Fisher, but not with Ogielski's Monte Carlo simulations.

I. INTRODUCTION

The statistical fluctuations in a system that has come to thermodynamic equilibrium in a state of broken symmetry may be qualitatively different from those that occur in more symmetric systems. A deep relation between symmetry breaking and fluctuations, if it exists, would obviously be important to understand because symmetries are broken in continuous phase transitions. An even more intriguing possibility is that investigations along these lines might lead to theories of the very slow relaxation mechanisms that are characteristic of glassy systems.

The simplest example of the situation of interest here is an Ising ferromagnet below its transition temperature in zero external field. In the thermodynamic limit, "up-down" symmetry of this system is broken spontaneously; it takes an infinitely long time for, say, the "up" state to transform to the "down" state via thermal fluctuations. On the other hand, large but finite regions of "down" spins do occur in the "up" state with finite probability. Such fluctuations should be specially long lived because they are locally indistinguishable from equilibrium states and, thus, the thermodynamic driving force which causes them to dissipate is relatively weak. If these fluctuations are statistically significant, they may make observable contributions to the long-time behavior of the spin autocorrelation function. It is this possibility that we shall examine in what follows.

The bases for our discussion are three prior investigations[1-3] in which this problem has been examined from different points of view. In each of these analyses, the Ising system described above has been studied with Glauber (nonconserving) kinetics, and an attempt has been made to evaluate the autocorrelation function $C(t)$:

$$C(t)=\langle S_i(t)S_i(0)\rangle-\langle S_i\rangle^2\ , \tag{1.1}$$

for large values of the time t. Here, $S_i=\pm 1$ is the Ising spin at the ith site, and the angular brackets denote a thermodynamic average for one of the equilibrium states, say, the "up" state.

Our approach is most directly related to that of Huse and Fisher.[1] These authors make the intuitively appealing argument that the dominant contributions to $C(t)$ at large times t must come from "down" spin which happen to be at or near the centers of large, slowly decaying droplets of the "down" state. Because the deterministic lifetime of a droplet of radius R is of order $\sqrt{t}$ and the probability of appearance of such a droplet is proportional to $\exp(-\sigma R^{d-1})$, where d is the spatial dimensionality and σ is a factor that contains the surface energy; thus

$$C(t)\approx \exp(-ct^{\beta})\ , \tag{1.2}$$

where $\beta=(d-1)/2$ and c is a constant. For $d=2$, the right-hand side of (1.2) is a stretched exponential with $\beta=\frac{1}{2}$. Because (1.2) predicts $\beta>1$ for $d>3$, Huse and Fisher conclude that $d=3$ is an upper critical dimension for the droplet model and that correlations will relax exponentially ($\beta=1$) for $d>3$.

Note the strong statistical assumption that is implicit in the above argument—that droplets which have survived for a long time t must, with high probability, have at some time in their history been large of order $\sqrt{t}$. In what follows we shall recover a result of the form (1.2) in a detailed study of fluctuations in the droplet approximation, but shall show that the strong statistical assumption is not always justified (e.g., for $d>3$), even with this special picture of the kinetic Ising model. (The possibility of a failure of this assumption in higher dimensions was also suggested by Huse and Fisher.)

A second recent analytic approach to this problem is that of Takano, Nakanishi, and Miyashita.[2] These authors start with the Fourier decomposition of the complete time-evolution operator Λ for the kinetic Ising (Glauber) model, and write $C(t)$ in the form

$$C(t)=\sum_{\kappa,j}[\mu_j(\kappa)]^2\exp[-\lambda_j(\kappa)t]\ , \tag{1.3}$$

where the $\lambda_j(\kappa)$ are the eigenvalues of Λ, κ is the wave vector, j denotes all other indices needed to specify the eigenstate, and the $\mu_j(\kappa)$ are matrix elements of the spin S_i. Takano *et al.*, argue that the lowest branch $\lambda_0(\kappa)$ of the spectrum of relaxation rates must make the dominant contribution to $C(t)$, and they further assume that this branch is gapless and has the form $\lambda_0(\kappa)\approx\kappa^2$. Their crucial and, in our opinion, most problematic assumption is that the factor $[\mu_j(\kappa)]^2$ is a measure of the probability of finding a droplet of size κ^{-1}, and thus is proportional to $\exp(-c'\kappa^{1-d})$, where c' is another constant. Substituting these functional forms for λ and μ into (1.3) and evaluating the integral over κ by a saddle-point method, they find a stretched exponential of the form (1.2) with $\beta=(d-1)/(d+1)$ for all d. They also have performed Monte Carlo simulations for two-dimensional systems with results that are consistent with their prediction, $\beta=\frac{1}{3}$.

In an attempt to resolve these two conflicting results, Ogielski has performed extensive Monte Carlo simulations for the two-, three-, and four-dimensional kinetic Ising models.[3] His data for $C(t)$ fit the stretched exponential form (1.2) quite well for all three cases, and his values of β are more nearly consistent with the predictions of Takano *et al.* than with those of Huse and Fisher. On the other hand, he interprets his computations to indicate that there is a gap in the spectrum of relaxation rates $\lambda_0(\kappa)$ for $d=3$. Because a gap necessarily implies an ordinary exponential decay at large times, the latter result seems to be inconsistent with the stretched exponential seen in the autocorrelation function. Perhaps, as suggested by Ogielski, the truly asymptotic exponential decay appears only at times much longer than those which can be achieved in this simulations.[4]

In this paper we study in detail the Langevin equation which describes fluctuations in the radius of a single droplet of the kind considered by Huse and Fisher. Section II contains a general description of the transformation to a Fokker-Planck equation and the way in which a spectral analysis of that equation can be used to compute the autocorrelation function $C(t)$. In Sec. III we show that the eigenvalues of the Fokker-Planck operator (not to be confused with the time-evolution operator Λ mentioned previously) form (1) a continuous spectrum of relaxation rates starting from zero for $d=2$; (2) a continuous spectrum with a finite gap for $d=3$; and (3) a discrete spectrum for $d>4$.

Detailed solutions for the various cases are presented in Sec. IV. For $d=3$, the Fokker-Planck equation can be solved exactly and we find, as expected from the spectral analysis, a simple exponential decay law. For $d=2$, we use a Wenzel-Kramers-Brillouin (WKB) method to obtain an approximation that is asymptotically accurate at large times. We find that $C(t)$ is a stretched exponential with $\beta=\frac{1}{2}$, consistent with Huse and Fisher but inconsistent with the numerical simulations of the full Ising model. Interestingly, we find that the dominant contribution to $C(t)$ for $d=2$ comes from droplets which are initially small and grow to sizes of order $\sqrt{t}$ at times of order $t/2$ before decaying almost to zero at time t. The analogous statement is incorrect for dimensions $d\geq 3$.

II. GENERAL FORMALISM

We consider the equilibrium dynamical fluctuations in one of the two symmetry-breaking states of an Ising system with nonconserving kinetics. Specifically, we consider the spin autocorrelation function $C(t)$ defined in Eq. (1.1) and, following Huse and Fisher, assume that the behavior of $C(t)$ at large times t is determined by droplet-like fluctuations as described in the Introduction.

We now make the following more specific assumptions regarding these droplets: (1) Only a single droplet centered at the given spin site is relevant. (2) Only a spherical droplet needs to be considered. (3) Translational motion of the droplet is negligible. Given these assumptions, we write the autocorrelation function in the form

$$C(t)\approx\int B(R_0)P(R_0,t)dR_0\ , \tag{2.1}$$

where $B(R_0)$ is the probability density that a droplet of initial radius R_0 is present at time $t=0$, and $P(R_0,t)$ is the probability that this droplet is still present at time t.

The dynamical fluctuations of a spherical droplet may be described by the Langevin equation[5]

$$\frac{dR}{dt}=-\frac{\Gamma\sigma}{R}+\frac{\eta(t)}{R^{(d-1)/2}}\ , \tag{2.2}$$

where σ is (apart from a constant factor) the surface tension at the droplet boundary, Γ is a transport coefficient, and $\eta(t)$ is a Gaussian noise with $\langle\eta(t)\rangle=0$ and

$$\langle\eta(t)\eta(t')\rangle=2\theta\Gamma\delta(t-t'),\quad \theta\equiv k_BT\ . \tag{2.3}$$

The reduction of noise by a factor of $R^{(d-1)/2}$ is due to averaging over the surface of the droplet. Equation (2.2) is valid for $R>\xi$, where ξ is the correlation length. For T well below T_c, ξ is of order of the lattice constant.

Now let

$$x=\frac{2}{d+1}R^{(d+1)/2},\quad \alpha=\sigma\left[\frac{d+1}{2}\right]^{(d-3)/(d+1)}\ . \tag{2.4}$$

Equation (2.2) becomes

$$\frac{dx}{dt}=-\Gamma\alpha x^{(d-3)/(d+1)}+\eta(t)$$
$$=-\Gamma\frac{dU}{dx}+\eta(t)\ , \tag{2.5}$$

where

$$U(x)=\left[\frac{d+1}{2d-2}\right]\alpha x^{(2d-2)/(d+1)}\ . \tag{2.6}$$

Equations (2.5) and (2.6) describe a one-dimensional random walk in a potential $U(x)$. The random walkers are subject to a drift force $-dU(x)/dx$ towards the origin. The probability density $\rho(x,t)$ of the random walkers satisfies the Fokker-Planck equation

$$\frac{1}{\Gamma}\frac{\partial\rho(x,t)}{\partial t}=\frac{\partial}{\partial x}\left[\rho\frac{dU}{dx}+\theta\frac{\partial\rho}{\partial x}\right]\ . \tag{2.7}$$

To complete the statement of the problem, we must impose a boundary condition at $x=0$, in addition to requiring $\rho(x,t)\rightarrow 0$ $(x\rightarrow\infty)$. We assume that if a droplet shrinks to size zero at some time t', it will not contribute to $C(t)$ for $t>t'$. Similarly, new droplets which appear after $t=0$ will not contribute to $C(t)$. Thus for purposes of computing the autocorrelation function, we place an absorbing wall at $x=0$; that is, $\rho(x=0,t)=0$.

Now our original problem of computing the autocorrelation function $C(t)$ becomes the following problem: Starting with an ensemble of random walkers distributed according to a thermal equilibrium distribution at $t=0$, compute how many of them are still present (not having been absorbed by the wall at $x=0$) at time t.

For an initial distribution, we choose the Boltzmann function

$$B(x)=A\exp[-U(x)/\theta]\ , \tag{2.8}$$

where

$$A=1\Big/\int_0^\infty dx\exp[-U(x)/\theta]$$

is a normalization factor. Note that this distribution has the correct R dependence, $B\sim\exp(-cR^{d-1})$, if x is expressed in terms of R using Eq. (2.4).

One sees from Eq. (2.6) that for $d=3$, the drift force on the random walkers is a constant, independent of the walker's position. For $d>3$, the force is larger for larger x; for $d<2$ the force is smaller for larger x. As we will see later, $d=3$ is the borderline separating two different long-time behaviors.

We now proceed to express $C(t)$ in terms of eigenfunctions of the operator on the right-hand side of Eq. (2.7). Let

$$\rho(x,t)=\psi(x,t)e^{-U(x)/2\theta}\ . \tag{2.9}$$

Substituting (2.9) into (2.7), we obtain

$$-\frac{1}{\Gamma}\frac{\partial\psi}{\partial t}=\mathcal{L}\psi\ , \tag{2.10}$$

where

$$\mathcal{L}=-\theta\frac{\partial^2}{\partial x^2}+V(x)\ , \tag{2.11}$$

and

$$V(x)=\frac{1}{4\theta}\left[\frac{dU}{dx}\right]^2-\frac{1}{2}\frac{d^2U}{dx^2}\quad(x>0)\ . \tag{2.12}$$

Let $\varphi_k(x)$ denote the eigenfunctions of the operator $\mathcal{L}$,

$$\mathcal{L}\varphi_k(x)=\varepsilon(k)\varphi_k(x)\ , \tag{2.13}$$

with $\varphi_k(x=0)=0$ and

$$\int_0^\infty\varphi_k^*(x)\varphi_{k'}(x)dx=\delta_{k,k'}\ . \tag{2.14}$$

Then

$$\psi(x,t)=\sum_k a_k\varphi_k(x)e^{-\varepsilon(k)\Gamma t}\ , \tag{2.15}$$

where

$$a_k=\int_0^\infty\psi(x,t=0)\varphi_k^*(x)dx\ . \tag{2.16}$$

If we start with a random walker at $x=x_0$ at $t=0$, then $\rho(x,t=0)=\delta(x-x_0)$, and, from Eq. (2.9), $\psi(x,t=0)=e^{U(x)/2\theta}\delta(x-x_0)$. Equation (2.16) then gives $a_k=\exp[U(x_0)/2\theta]\varphi_k^*(x_0)$. The Green's function, i.e., the probability density that this random walker will be at position x at time t, is

$$\begin{aligned}\rho(x,t|x_0,t=0)&=e^{-U(x)/2\theta}\psi(x,t|x_0,t=0)\\&=\sum_k e^{-[U(x)-U(x_0)]/2\theta}\varphi_k^*(x_0)\varphi_k(x)e^{-\varepsilon(k)\Gamma t}\ .\end{aligned} \tag{2.17}$$

The survival probability for this random walker is

$$P(x_0,t)=\int_0^\infty\rho(x,t|x_0,t=0)dx\ . \tag{2.18}$$

We now define the weighted Green's function $G(x,x_0;t)$

$$\begin{aligned}G(x,x_0;t)&\equiv Ae^{-U(x_0)/\theta}\rho(x,t|x_0,t=0)\\&=Ae^{-[U(x)+U(x_0)]/2\theta}\sum_k\varphi_k^*(x_0)\varphi_k(x)e^{-\varepsilon(k)\Gamma t}\ ,\end{aligned} \tag{2.19}$$

where $A\exp[-U(x_0)/\theta]$ is the equilibrium distribution, Eq. (2.8). Note that $G(x,x_0;t)$ is invariant under the interchange of x and x_0. If we start with the equilibrium distribution at $t=0$, the distribution function at time t, $\rho(x,t)$, is simply

$$\rho(x,t)=\int_0^\infty G(x,x_0;t)dx_0\ . \tag{2.20}$$

The weighted survival probability is

$$\bar{P}(x_0,t)\equiv Ae^{-U(x_0)/\theta}P(x_0,t)=\int_0^\infty G(x,x_0;t)dx\ , \tag{2.21}$$

where $P(x_0,t)$ is the unweighted survival probability given in Eq. (2.18). The right-hand sides of Eqs. (2.20) and (2.21) are equal to each other, thus $\bar{P}(x_0,t)=\rho(x=x_0,t)$, because of the symmetry in $G(x,x_0;t)$. From Eqs. (2.1) and (2.19)–(2.21), we have

$$\begin{aligned}C(t)&=\int_0^\infty\bar{P}(x_0,t)dx_0\\&=\int_0^\infty\rho(x,t)dx\\&=A\sum_k e^{-\varepsilon(k)\Gamma t}\left|\int_0^\infty e^{-U(x)/2\theta}\varphi_k(x)dx\right|^2\ .\end{aligned} \tag{2.22}$$

As we will see later in the paper, another quantity of interest is the half-time history. Namely, if a random walker is at position x_0 at $t=0$ *and* at t, what is its distribution at $t/2$? This quantity is simply the product of two Green's functions

$$\begin{aligned}H(x,t/2|x_0t;x_00)&\equiv\rho(x_0,t|x,t/2)\rho(x,t/2|x_0,t=0)\\&=\left|\sum_k\varphi_k^*(x_0)\varphi_k(x)e^{-\varepsilon(k)\Gamma t/2}\right|^2\ ,\end{aligned} \tag{2.23}$$

where Eq. (2.17) is used for the last equality.

III. RELAXATION-RATE SPECTRA FOR THE FOKKER-PLANCK EQUATION

Before calculating $C(t)$, let us derive some features of the relaxation rates. The relaxation-rate spectra for the

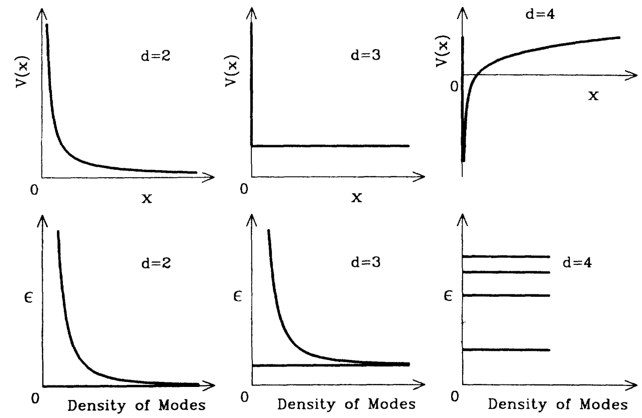

FIG. 1. Schematic graphs for the potentials $V(x)$ (upper row) and the density of modes (lower row) for $d=2$, $d=3$, and $d=4$.

Fokker-Planck equation (2.7) are the same as the eigenvalue spectra $\varepsilon(k)$ of (2.13). Without the absorbing boundary at $x=0$, the smallest relaxation rate of (2.7) would be zero, the equilibrium distribution (2.8) would be a stationary solution, and all other solutions will decay in time. With the boundary condition $\rho(0,t)=0$ or $\psi(0,t)=0$, the eigenvalues of (2.11) will be pushed up somewhat because, in quantum mechanical language, more "kinetic energy" is needed to suppress the wave function at the boundary; thus, all solutions now decay.

For $d=2$, the potential is given by

$$V(x)=\frac{\alpha^2}{4\theta x^{2/3}}+\frac{\alpha}{6x^{4/3}} . \tag{3.1}$$

All the eigenstates are scattering states and the spectrum is continuous from zero without a gap. The density of eigenvalues has an $\varepsilon^{-1/2}$ singularity at $\varepsilon=0$. For $d=3$, the potential is constant,

$$V(x)=\frac{\sigma^2}{4\theta} , \tag{3.2}$$

and the spectrum is again continuous but with a finite gap, $\sigma^2/4\theta$. The density of eigenvalues has the same singularity, as in the case of $d=2$ above the gap. In the case $d\geq 4$, the potential is monotonically increasing with $V(x)\rightarrow-\infty$ $(x\rightarrow 0)$, and $V(x)\rightarrow+\infty$ $(x\rightarrow\infty)$. All the eigenfunctions are bound states and the spectrum is discrete with a lowest eigenvalue ε_0 greater than zero. These results are illustrated in Fig. 1.

IV. CALCULATION OF AUTOCORRELATION FUNCTIONS

Now we consider the autocorrelation functions in this model.

A. The case of $d\geq 4$

Because the spectrum $\varepsilon(k)$ is discrete, the slowest mode in the sum in Eq. (2.22) will make the largest contribution to $C(t)$ starting from relatively early time t; that is, $C(t)\sim\exp(-\varepsilon_0\Gamma t)$ except for very small t. There is no way to get a stretched exponential within this model for $d\geq 4$.

B. Exact results for $d=3$

Due to the extremely simple form of the potential (3.2), we can calculate $C(t)$ exactly in this case. Because there is a gap in the relaxation rate spectrum, we expect that the asymptotic form of $C(t)$ will be exponential. However, the lowest mode has zero weight in Eq. (2.22), and thus there turns out to be a power-law correction.

The equation we must solve is (2.13) with the operator,

$$\mathcal{L}=-\theta\frac{\partial^2}{\partial x^2}+\frac{\sigma^2}{4\theta} . \tag{4.1}$$

The set of eigenfunctions satisfying the boundary condition $\varphi_k(x=0)=0$ is

$$\varphi_k(x)=\sqrt{2/\pi}\sin(kx) \quad (k>0) , \tag{4.2}$$

with the eigenvalue

$$\varepsilon(k)=\theta k^2+\frac{\sigma^2}{4\theta} . \tag{4.3}$$

Substituting Eqs. (2.6), (4.2), and (4.3) into Eq. (2.22), we have

$$\begin{aligned} C(t)&=\frac{\sigma}{2\pi\theta}\int_0^\infty e^{-(\theta k^2+\sigma^2/4\theta)\Gamma t}\left|\frac{2k}{(\sigma/2\theta)^2+k^2}\right|^2 dk\\ &=\frac{\tau+2}{2}\operatorname{erfc}\left[\frac{\sqrt{\tau}}{2}\right]-\sqrt{\tau/\pi}e^{-\tau/4} , \end{aligned} \tag{4.4}$$

where $\tau=\sigma^2\Gamma t/\theta$ is the dimensionless time and

$$\operatorname{erfc}(z)=\frac{2}{\sqrt{\pi}}\int_z^\infty e^{-u^2}du \tag{4.5}$$

is the complementary error function. Using the asymptotic expansion of erfc(z), we obtain the asymptotic expansion for $C(t)$

$$\begin{aligned} C(\tau)=\frac{8e^{-\tau/4}}{\sqrt{\pi}\tau^{3/2}}\Bigg[1-2\frac{1\times 3}{\tau/2}+3\frac{1\times 3\times 5}{(\tau/2)^2}\\ -4\frac{1\times 3\times 5\times 7}{(\tau/2)^3}+\cdots\Bigg] . \end{aligned} \tag{4.6}$$

In order to have a clearer picture, let us study the distribution function $\rho(x,t)$ and the weighted survival probability $\bar{P}(x_0,t)$, Eqs. (2.20) and (2.21). The weighted Green's function $G(x,x_0;t)$, Eq. (2.19), can be easily evaluated. We have

$$\begin{aligned} G(x,x_0;t)&=\frac{e^{-\sigma^2\Gamma t/4\theta}}{\sqrt{4\pi\theta\Gamma t}}e^{-(\sigma/2\theta)(x+x_0)}\\ &\quad\times(e^{-(x-x_0)^2/4\theta\Gamma t}-e^{-(x+x_0)^2/4\theta\Gamma t}) . \end{aligned} \tag{4.7}$$

And

$$\rho(x,t)=\bar{P}(x,t)=\int_0^\infty G(x,x_0;t)dx_0$$
$$=\frac{\sigma}{2\theta}\left[e^{-(\sigma/\theta)x}\text{erfc}\left[\frac{\Gamma\sigma t-x}{\sqrt{4\theta\Gamma t}}\right]-\text{erfc}\left[\frac{\Gamma\sigma t+x}{\sqrt{4\theta\Gamma t}}\right]\right]. \quad (4.8)$$

One sees from Eq. (4.8) that for any given initial size x the survival probability decays exponentially as $e^{-\tau/4}$ for $t \gg x/\Gamma\sigma$, or in a dimensionless form $\tau \gg r^2/2$, where $r=\sqrt{2\sigma x/\theta}$ is the dimensionless droplet radius. One can also show easily from Eq. (4.8) that the most probable droplet size $x^* \to 2\theta/\sigma$ as $t\to\infty$, although the term "most probable" makes less and less sense because the peak gets broader and broader. Accordingly, initially as well as presently, small droplets make the most contribution to the autocorrelation function $C(t)$ at all times. Recall that the drift force on the random walkers is constant for $d=3$ [Eq. (2.5)], so it does not distinguish between small or large droplets. Because small droplets have larger Boltzmann weights initially, they dominate $C(t)$ at all times. The half-time distribution function $H(x,t/2|x_0t;x_00)$, Eq. (2.23), for $x_0=x^*=2\theta/\sigma$ is easily calculated,

$$H(x,t/2|x_0t;x_00)$$
$$=\frac{e^{-\sigma^2\Gamma t/4\theta}}{2\pi\theta\Gamma t}\left(e^{-(x-x_0)^2/2\theta\Gamma t}-e^{-(x+x_0)^2/2\theta\Gamma t}\right)^2 . \quad (4.9)$$

This function remains peaked near x_0 for all t. Thus for a small droplet with initial and present size $\sim x^*=2\theta/\sigma$, the most probable history remains at all times in the neighborhood of x^*, staying small. From the above discussion, we see that in three dimensions the large droplet fluctuations do not contribute significantly to $C(t)$. This result is consistent with the asymptotic exponential decay of $C(t)$, but not with the picture proposed by Huse and Fisher.

C. The case of $d=2$

We now consider the two-dimensional system. In this case, we have a continuous spectrum from zero and a nonexponential decay can be expected. The potential $U(x)$ [Eq. (2.6)] on the random walkers is

$$U(x)=\tfrac{3}{2}\alpha x^{2/3} . \quad (4.10)$$

The larger the x, the smaller the drift force is; thus large droplets may play an important role in determining $C(t)$. The "Schrödinger equation" (2.13) is

$$\left[-\theta\frac{\partial^2}{\partial x^2}+V(x)\right]\varphi_\varepsilon(x)=\varepsilon\varphi_\varepsilon(x) , \quad (4.11)$$

where the potential $V(x)$ [Eq. (2.13)] is

$$V(x)=\frac{\alpha^2}{4\theta x^{2/3}}+\frac{\alpha}{6x^{4/3}} . \quad (4.12)$$

Although we cannot solve Eqs. (4.11) and (4.12) exactly, we can estimate the asymptotic behavior for large t using a WKB approximation (see the Appendix for mathematical details). To check these asymptotic estimates, and also to obtain results valid for all t, we have solved (2.5) numerically. Our results are shown in Figs. 2–5 and are referred to throughout the following paragraphs.

The asymptotic form of $C(t)$ turns out to be

$$C(\tau)\propto e^{-\sqrt{(2/3)\tau}} , \quad (4.13)$$

where $\tau=\sigma^3\Gamma t/\theta^2=3\alpha^3\Gamma t/2\theta^2$ is the dimensionless time. Equation (4.13) is a stretched exponential with exponent $\beta=\frac{1}{2}$. The time after which Eq. (4.13) is expected to be valid is

$$\tau_0\approx\tfrac{8}{3} . \quad (4.14)$$

In Fig. 2(a), $C(\tau)$ is plotted as obtained by a direct numerical solution of the Langevin equation (2.5) for $d=2$. It nicely fits a stretched exponential form $C(\tau)\sim e^{-0.82\sqrt{\tau}}$, in excellent agreement with Eq. (4.13) which gives $C(\tau)\sim e^{-0.8165\sqrt{\tau}}$. Also shown in Fig. 2(b) and 2(c), are numerical solutions of (2.5) for $d=3$ and $d=4$, which illustrate the simple exponential behavior deduced above.

It is also shown in the Appendix that the most probable droplet size x^* at time t is

$$x^*=\left[\frac{\theta^2\Gamma t}{9\alpha}\right]^{3/8} , \quad (4.15a)$$

or

$$r^*=(\tfrac{3}{8}\tau)^{1/4} , \quad (4.15b)$$

where $r=\sigma R/\theta=\sigma(3x/2)^{2/3}/\theta$ is the dimensionless droplet radius. Equation (4.15) also means that at time τ the droplets of *initial* radius r^* contribute most to $C(\tau)$. We compare Eq. (4.15) with our numerical solution of (2.5). Figure 3 shows the distribution function $\rho(x,t)$ as a function of x for various t. The maximum point for each curve x^* is plotted as a function of t in Fig. 4. The agreement with Eq. (4.15a) is very satisfactory. In order to see how the relatively small droplets of size $r\sim\tau^{1/4}$ that dominate the distribution at early and late times produce

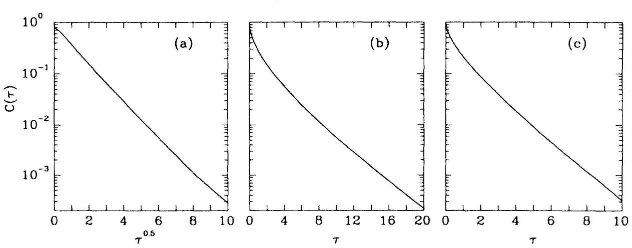

FIG. 2. Numerical results for the autocorrelation function $C(\tau)$ as a function of the dimensionless time τ which is defined as $\tau\equiv\sigma^{(d+1)/(d-1)}\theta^{-2/(d-1)}\Gamma t$ for (a) $d=2$, (b) $d=3$, and (c) $d=4$.

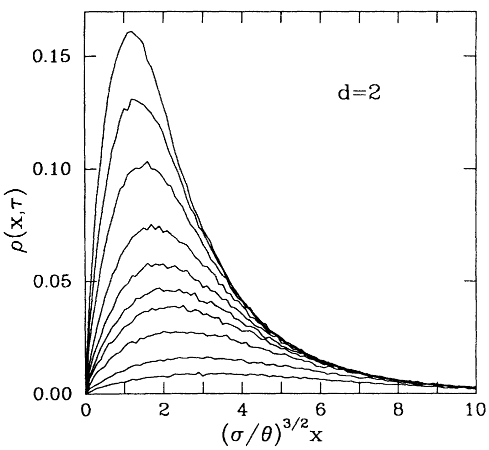

FIG. 3. Numerical results for the distribution $\rho(x,t)$ for $d=2$ as a function of the dimensionless droplet size $(\sigma/\theta)^{3/2}x$ for $\tau=0.5$, 0.7, 1.0, 1.5, 2.0, 2.5, 3.0, 4.0, 6.0, and 8.0 from upper to lower curves.

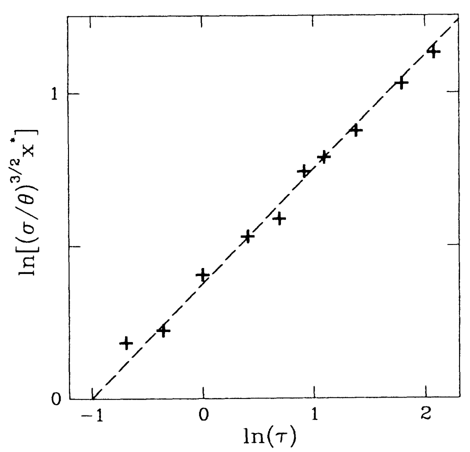

FIG. 4. Peak positions $(\sigma/\theta)^{3/2}x^*$ of $\rho(x,\tau)$ in Fig. 3 vs the dimensionless time τ. The dashed line is a straight line with slope $\frac{3}{8}$.

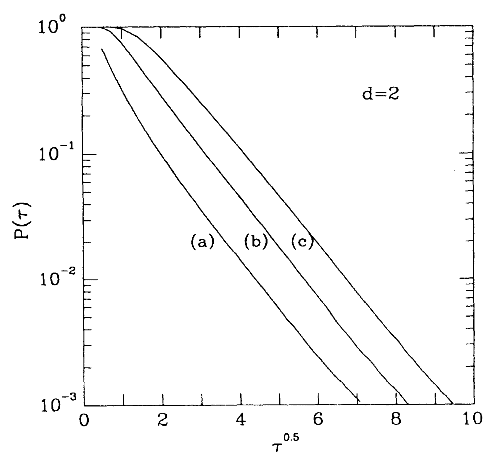

FIG. 5. Numerical results for the survival probability $P(r_0,\tau)$ for $d=2$: curve (a) $r_0=1$; curve (b) $r_0=2$; (c) $r_0=3$, where r_0 is the dimensionless droplet radius defined in the text.

$\beta=\frac{1}{2}$ in (1.2), we calculate the half-time history, Eq. (2.23), in the Appendix. It is shown there that the half-time distribution $H(r,\tau/2|r_0\tau;r_0 0)$ is peaked at $r_h\sim\sqrt{\tau}$ with a width $\Delta r\sim\tau^{1/4}$ for $r_0\ll r_h$. Because $\Delta r/r_h\sim\tau^{-1/4}$, the peak is relatively sharp. Thus the most probable way for droplets of size $r\sim\tau^{1/4}$ to survive until time τ is that they first grow large to sizes of order of $r_h\sim\sqrt{\tau}$, then shrink back to about the original size. Because the statistical weight of this process is $\sim\exp(-cr_h)\sim\exp(-c'\sqrt{\tau})$, the contribution of these droplets to $C(t)$ is $\exp(-c'\sqrt{\tau})$. For $d=2$, therefore, and probably for any $d<3$, the picture of droplet fluctuations proposed by Huse and Fisher seems to be self-consistent. We also derive the asymptotic form of weighted survival probability $\bar{P}(x_0,t)$ in the Appendix,

$$\bar{P}(r_0,\tau)\sim e^{-\sqrt{(2/3)\tau}}\quad(\tau\gg\tfrac{8}{3}r_0^2,\ r_0\gg1)\ . \tag{4.16}$$

So, the survival probability asymptotically decays as a stretched exponential at least for any initially large droplets. This result is not surprising, because the difference between the ensemble that starts only with droplets of a given size and one that starts as a Boltzmann distribution becomes smaller and smaller as time evolves. Our numerical solutions are in good agreement with Eq. (4.16), as shown in Fig. 5.

V. DISCUSSION

In order to study the spin autocorrelation function $C(t)$ in the ordered stated in the kinetic Ising system, we have analyzed the Langevin equation for fluctuations of the radius of a single spherical droplet, assuming that deformations of the shape and interactions between droplet are not important.

The relaxation-rate spectra for the Fokker-Planck equation corresponding to this Langevin equation were shown to be (1) continuous from zero for $d=2$ (2) continuous with a finite gap of magnitude $\sigma^2/4k_BT$ for $d=3$, and (3) discrete for $d\geq4$. The gaps in the spectra for $d\geq3$ in the Fokker-Planck equation seem to correspond to the gap found by Ogielski in his Monte Carlo simulations in $d=3$;[4] but it is not clear that we should compare the relaxation-rate spectrum of the Fokker-Planck equation for a droplet to that of the time-evolution operator for the Glauber model.

We have found that the asymptotic form of the autocorrelation function $C(t)$ in this model is a simple exponential for $d\geq3$ and a stretched exponential with $\beta=\frac{1}{2}$ for $d=2$. The survival probability for a droplet of almost any initial size is similar to $C(t)$ for large enough times. For $d\geq3$, the large droplets are never statistically important enough to contribute significantly to $C(t)$, and there is a finite gap in the relaxation-rate spectrum. For $d=2$, on the other hand, large droplet fluctuations do play an important role in long-time equilibrium dynamics, and there is a stretched exponential decay of $C(t)$.

Our results for $C(t)$ are the same as those of Ref. 1. The physical pictures are somewhat different, however. In particular, Huse and Fisher assumed that the survival probability vanishes after the deterministic lifetime, and that the droplet approximation by itself predicts the decay of $C(t)$ to be faster than exponential for $d>3$. Our analysis predicts exponential decay for $d \geq 3$ without appealing to any considerations beyond the droplet model.

The above results for $C(t)$ in the droplet model are distinctly different from the results of Ogielski's simulations[3] for the Glauber model. Although $C(t)$ had become as small as 10^{-3} in his simulations, Ogielski conjectured that he might not have reached the asymptotic region in the time that was computationally accessible. As shown in Fig. 2, however, the asymptotic region starts quite early in the droplet model. We conclude that the present simple-droplet picture must be insufficient and that the effects neglected (the shape effects, the interaction between droplets, etc.) are important at least in the region where the simulations were performed.

On the other hand, the exponents β predicted by Takano *et al.*[3] agree reasonably well with Ogielski's results for all dimensions. Their argument, however, is based on the simple assignment of gapless modes in the Glauber model to the droplet motion. Such a naive assignment seems dubious because the relaxation-rate spectra for droplet fluctuations were shown to have gaps for $d \geq 3$. Although the gapless modes themselves can be expected from the $\kappa^2 t$ scaling law in the quenched system, these gapless modes may not have weight in the states of broken symmetry. We think it might be interesting to see whether the gap that has been found in Monte Carlo simulations for the Glauber model in $d=3$ actually corresponds to the gap of the droplet fluctuations by examining its temperature dependence.[4]

ACKNOWLEDGMENTS

This research was supported by the National Science Foundation Grant No. PHY82-17853 with supplementary support from the National Aeronautics and Space Administration and by U.S. Department of Energy Grant No. DE-FG03-84ER45108.

APPENDIX

In this appendix we derive Eqs. (4.13)–(4.16) using a WKB approximation for (4.11).

Define

$$p(x)=\sqrt{\varepsilon - V(x)}\ . \tag{A1}$$

Then the WKB solution is[6]

$$\varphi_\varepsilon(x)=\frac{1}{[4\pi|p(x)|\theta^{1/2}]^{1/2}}\exp\left[-\frac{1}{\sqrt{\theta}}\int_x^a |p(x')|dx'\right]$$

$$\equiv \varphi_\varepsilon^L(x) \quad (0<x<a)\ , \tag{A2a}$$

$$\varphi_\varepsilon(x)=\frac{1}{[\pi p(x)\theta^{1/2}]^{1/2}}\cos\left[\frac{1}{\sqrt{\theta}}\int_a^x p(x')dx' - \frac{\pi}{4}\right]$$

$$\equiv \varphi_\varepsilon^R(x) \quad (x>a)\ , \tag{A2b}$$

where a is the classical turning point, $V(a)=\varepsilon$. Note that we have chosen to normalize φ_ε so that sums over k in equations such as (2.15) or (2.22) become integrals over ε.

We are interested in the behavior of $C(t)$ for large t, which is determined by the properties of $\varphi_\varepsilon(x)$ for small ε. In the subsequent discussion we assume that t is large enough that the relevant ε's satisfy

$$\varepsilon < \frac{3\alpha^3}{8\theta^2}\ . \tag{A3}$$

Then the first term on the right-hand side of (3.1) is dominant near the turning point, and $a \approx (\alpha^2/4\theta\varepsilon)^{3/2}$. This will simplify some calculations. The regions in which the WKB eigenfunctions (A2a) and (A2b) are good approximations are

$$\left[\frac{2\theta}{3\alpha}\right]^{3/2} < x < a-\delta\ , \tag{A4a}$$

and

$$x > a+\delta\ , \tag{A4b}$$

respectively, where $\delta \approx 3^{1/3}\alpha/4\theta^{1/6}\varepsilon^{5/6}$ is the width of the region in which the WKB approximation breaks down near the turning point.

We now proceed to calculate the autocorrelation function $C(t)$, the distribution function $\rho(x,t)$, the weighted survival probability $\bar{P}(x_0,t)$, and the half-time history $H(x,t/2|x_0 t;x_0 0)$ via Eqs. (2.19)–(2.23). Let us first evaluate the integral

$$\int_0^\infty \varphi_\varepsilon(x)e^{-U(x)/2\theta}dx$$

$$=\int_0^a \varphi_\varepsilon^L(x)\exp\left[-\frac{3\alpha}{4\theta}x^{2/3}\right]dx + \int_a^\infty \varphi_\varepsilon^R(x)\exp\left[-\frac{3\alpha}{4\theta}x^{2/3}\right]dx\ .$$

The second term can be ignored in the energy region defined by Eq. (A3). We estimate the first term by a saddle-point method:

$$\int_0^\infty \varphi_\varepsilon(x)e^{-U(x)/2\theta}dx \approx \int_0^a \exp\left[-\frac{1}{\sqrt{\theta}}\int_x^a \sqrt{V(x')-\varepsilon}\,dx' - \frac{3\alpha}{4\theta}x^{2/3}\right]dx$$

$$\equiv \int_0^a e^{-S(x)}dx$$

$$\approx e^{-S(x^*)} \approx \exp(-\alpha^3/8\theta^2\varepsilon)\ , \tag{A5}$$

where

$$x^* = \left[\frac{6\varepsilon}{\alpha}\right]^{-3/4} \tag{A6}$$

is the saddle point, and

$$S(x^*) \approx \frac{\alpha^3}{8\theta^2\varepsilon}\ . \tag{A7}$$

One can easily check that x^* is within the region specified by Eq. (A4a). The function $\exp[-S(x)]$ is sharply peaked for small θ, but $(\Delta x/x^*)^2 \sim [(x^*)^2 S''(x^*)]^{-1} \sim \theta$ is independent of ε, as $\varepsilon \to 0$. Substituting Eq. (A5) into Eq. (2.22), we have

$$C(t)\approx\int_0^\infty \exp\left[-\frac{\alpha^3}{4\theta^2\varepsilon}-\varepsilon\Gamma t\right]d\varepsilon .$$

Again we evaluate the above integral via the saddle-point method. We find the saddle point

$$\varepsilon^*=\frac{\alpha^{3/2}}{2\theta\sqrt{\Gamma t}} , \qquad (A8)$$

and in this way obtain Eq. (4.13). From Eqs. (A3) and (A8), we get Eq. (4.14). The most probable droplet size x^* at time t, Eq. (4.15), is easily obtained from Eqs. (2.20), (A5), (A7), and (A8).

We now calculate the time dependence of the weighted survival probability $\bar{P}(x_0,t)$. Substituting Eq. (A5) into (2.21), we have

$$\bar{P}(x_0,t)\approx e^{-U(x_0)/2\theta}\left[\int_0^{V(x_0)}\varphi_\varepsilon^L(x_0)\exp(-\varepsilon\Gamma t-\alpha^3/8\theta^2\varepsilon)d\varepsilon+\int_{V(x_0)}^\infty\varphi_\varepsilon^R(x_0)\exp(-\varepsilon\Gamma t-\alpha^3/8\theta^2\varepsilon)d\varepsilon\right] . \qquad (A9)$$

There is an energy gap $V(x_0)$ in the second integral, so it decays exponentially for large t. The first integral gives an anomalous decay. For

$$x_0\gg\left[\frac{2\theta}{3\alpha}\right]^{3/2} , \qquad (A10a)$$

or in a dimensionless form

$$r_0\gg 1 , \qquad (A10b)$$

$$\int_{x_0}^a\sqrt{V(x')-\varepsilon}\,dx'\approx\int_{x_0}^a\left[\frac{\alpha^2}{4\theta x^{2/3}}-\varepsilon\right]^{1/2}dx'$$
$$=\frac{1}{\varepsilon}\left[\frac{\alpha^2}{4\theta}-\varepsilon x_0^{2/3}\right]^{3/2} . \qquad (A11)$$

Substituting Eq. (A11) into $\varphi_\varepsilon^L(x_0)$ in the first term of Eq. (A9), Eq. (4.16) is obtained via the saddle-point estimation.

Finally, we calculate the half-time history, Eq. (2.23), which is the square of the following integral ($x>x_0$):

$$\int_0^\infty d\varepsilon\,\varphi_\varepsilon(x_0)\varphi_\varepsilon(x)e^{-\varepsilon\Gamma t/2}=\int_0^{V(x)}\varphi_\varepsilon^L(x_0)\varphi_\varepsilon^L(x)e^{-\varepsilon\Gamma t/2}d\varepsilon+\int_{V(x)}^{V(x_0)}\varphi_\varepsilon^L(x_0)\varphi_\varepsilon^R(x)e^{-\varepsilon\Gamma t/2}d\varepsilon$$
$$+\int_{V(x_0)}^\infty\varphi_\varepsilon^R(x_0)\varphi_\varepsilon^R(x)e^{-\varepsilon\Gamma t/2}d\varepsilon$$
$$\equiv I_1+I_2+I_3 . \qquad (A12)$$

What we have in mind is that x_0 is small compared with x ($x_0\ll x$). I_3 can then be ignored. Let us now consider I_1:

$$I_1\approx\int_0^{V(x)}\exp\left[-\frac{1}{\sqrt{\theta}}\int_{x_0}^a\sqrt{V-\varepsilon}\,dx'-\frac{1}{\sqrt{\theta}}\int_x^a\sqrt{V-\varepsilon}\,dx'-\frac{\varepsilon\Gamma t}{2}\right]d\varepsilon$$
$$\equiv\int_0^{V(x)}\exp[-S_1(\varepsilon,x)]d\varepsilon . \qquad (A13)$$

Using Eq. (A11), we get ($x_0\ll x$)

$$S_1(\varepsilon,x)\approx\frac{1}{\varepsilon\theta^2}\left[\frac{\alpha^2}{2}\right]^3+\frac{1}{\varepsilon\sqrt{\theta}}\left[\frac{\alpha^2}{4\theta}-\varepsilon x^{2/3}\right]^{3/2}+\frac{\varepsilon\Gamma t}{2} . \qquad (A14)$$

To study Eqs. (A13) and (A14), it is convenient to use the variables ξ and u defined by $V(x)=\xi$ and $\varepsilon=\xi(1-u)$. Equations (A13) and (A14) then become

$$I_1\approx\int_0^1\exp[-\tilde{S}_1(u,\xi)]du , \qquad (A15)$$

and

$$\tilde{S}_1(u,\xi)=\frac{A}{\xi(1-u)}(1+u^{3/2})+\frac{\Gamma t\xi}{2}(1-u)$$
$$\approx\frac{A}{\xi}+\frac{\Gamma t\xi}{2}+\left[\frac{A}{\xi}-\frac{\Gamma t\xi}{2}\right]u+\frac{A}{\xi}u^{3/2}$$
$$+\frac{A}{\xi}u^2+\cdots , \qquad (A16)$$

where $A\equiv(\alpha/2)^3/\theta^2$. $\tilde{S}_1(0,\xi)$ has minimum at $\xi^*=\sqrt{2A/\Gamma t}$. Letting $\xi=\xi^*+w$, we have

$$\tilde{S}_1(u,\xi)\approx\sqrt{2A\Gamma t}+\frac{(\Gamma t)^{3/2}w^2}{2^{3/2}\sqrt{A}}-\Gamma twu$$
$$+\left[\frac{A\Gamma t}{2}\right]^{1/2}(u^{3/2}+u^2)+\cdots , \qquad (A17)$$

and

$$I_1\approx\exp\left[-\sqrt{2A\Gamma T}-\frac{(\Gamma t)^{3/2}w^2}{2^{3/2}\sqrt{A}}\right]$$
$$\times\int_0^1\exp[\Gamma twu-\sqrt{A\Gamma t/2}(u^{3/2}+u^2)]du . \qquad (A18)$$

The integral in (A18) gives an unimportant prefactor for

$w<0$. For $w>0$, it is $\exp[8(\Gamma t)^2 w^3/27A]$ by a saddle-point estimation. Thus for $w \ll \sqrt{A/\Gamma t}$, which is the region we are interested in,

$$I_1 \approx \exp\left[-\sqrt{2A\Gamma t} - \frac{(\Gamma t)^{3/2} w^2}{2^{3/2}\sqrt{A}}\right] = \exp\left[-\frac{2}{3}r_h - \frac{(r-r_h)^2}{3r_h}\right], \tag{A19}$$

where

$$r_h = (\tfrac{3}{8}\tau)^{1/2}, \tag{A20}$$

and $r = 3\alpha x^{2/3}/2\theta$ and $\tau = 3\alpha^3\Gamma t/2\theta^2$ is the dimensionless droplet radius and the dimensionless time, respectively. Equation (A20) applies for any $r_0 \ll r_h$. The width of the Eq. (A19) is

$$\Delta r \approx r_h^{1/2} \approx \tau^{1/4}. \tag{A21}$$

I_2 can be calculated in the same manner if one notices that

$$I_2 \approx \int_0^1 \{\exp[-\tilde{S}_2^+(u,\xi)] + \exp[-\tilde{S}_2^-(u,\xi)]\}du, \tag{A22}$$

where $\varepsilon \equiv \xi(1+u)$ and

$$\tilde{S}_2^{\pm}(u,\xi) = \frac{A}{\xi(1+u)}(1 \pm iu^{3/2}) + \frac{\Gamma t \xi}{2}(1+u). \tag{A23}$$

Comparing Eqs. (A22) and (A23) with (A15) and (A16), we find that $I_2 \approx I_1$. So for $r_0 \ll r_h$,

$$H(r,\tau/2|r_0\tau;r_0 0) \approx \exp\left[-\frac{4}{3}r_h - \frac{2(r-r_h)^2}{3r_h}\right]. \tag{A24}$$

Thus H is peaked at $r_h \approx \sqrt{\tau}$ with its maximum value being $\sim \exp(-4r_h/3) \sim \exp(-\sqrt{2\tau/3})$ and width $\sim \tau^{1/4}$.

---

[1] D. A. Huse and D. S. Fisher, Phys. Rev. B **35**, 6841 (1987).

[2] H. Takano, H. Nakanishi, and S. Miyashita, Phys. Rev. B **37**, 3716 (1988).

[3] A. T. Ogielski, Phys. Rev. B **36**, 7315 (1987).

[4] Ogielski (see Ref. 3) finds a gap in relaxation rate spectra and expects exponential decay for $t \gg 10^4$. However, he obtained this estimate by assuming that $G(k,t) \equiv \langle S_k(t)S_{-k}(0)\rangle \to G(k,0)\exp(-\lambda_0(k)t)$ $(t \to \infty)$, which is not necessarily true and should be examined carefully. To test this assumption, Ogielski computed $y \equiv -t/\ln[G(k,t)/G(k,0)]$ and found it to be roughly constant as a function of t. But y is insensitive to a prefactor $a(k,t)$ defined by $G(k,t) = a(k,t)G(k,0)\exp[-\lambda_0(k)t]$, which is expected in Ref. 2.

[5] K. Kawasaki and T. Ohta, Prog. Theor. Phys. **67**, 147 (1982).

[6] We neglected a very small term,

$$\{C/[4\pi|p(x)|\theta^{1/2}]^{1/2}\}\exp\left[+(1/\sqrt{\theta})\int_x^a |p(x')|dx'\right],$$

in (A2a), where

$$|C| \approx \exp\left[-(2/\sqrt{\theta})\int_0^a |p(x')|dx'\right].$$