Finite-Size Effects on Critical Diffusion and Relaxation Towards Metastable Equilibrium

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We present the first analytic study of finite-size effects on critical diffusion above and below $T_c$ of three-dimensional Ising-like systems whose order parameter is coupled to a conserved density. We also calculate the finite-size relaxation time that governs the critical order-parameter relaxation towards a metastable equilibrium state below $T_c$. Two new universal dynamic amplitude ratios at $T_c$ are predicted and quantitative predictions of dynamic finite-size scaling functions are given that can be tested by Monte-Carlo simulations.

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The dissipative critical dynamics of bulk systems with a non-conserved order parameter are reasonably well understood. Depending on whether the order parameter is governed by purely relaxational dynamics or whether it is coupled to a hydrodynamic (conserved) density such systems belong to the universality classes of models A or C [1]. The fundamental dynamic quantities of these systems are the relaxation and diffusion times which diverge as the critical temperature $T_c$ is approached.

For finite systems, these times are expected to become smooth and finite throughout the critical region and to depend sensitively on the geometry and boundary conditions. These finite-size effects are particularly large in Monte Carlo (MC) simulations of small systems. On a qualitative level, they can be interpreted on the basis of phenomenological finite-size scaling assumptions. For a more stringent analysis the knowledge of the shape of universal finite-size scaling functions is necessary. So far there exist reliable theoretical predictions on finite-size dynamics in three dimensions only on two relaxation times $\tau_1$ and $\tau_2$ determining the long-time behavior of the order parameter and the square of the order parameter $\langle \varphi^2 \rangle$.

In this Letter we present the first prediction of the finite-size scaling function for the critical diffusion time of three-dimensional systems above and below $T_c$. Furthermore we shall present the analytic identification and quantitative calculation of a new leading relaxation time that governs the critical order-parameter relaxation towards a metastable equilibrium state of finite systems below $T_c$. Our predictions contain no adjustable parameters other than two amplitudes of the bulk system.

We start from model C [2], i.e., from the relaxational and diffusive Langevin equations for the one-component order-parameter field $\varphi(x, t)$ and for the density $\rho(x, t) = \langle \varphi \rangle + m$ in a finite volume $V$,

$$\frac{\partial \varphi(x, t)}{\partial t} = -\Gamma_0 \frac{\delta H}{\delta \varphi(x, t)} + \Theta_\varphi(x, t),$$

$$\frac{\partial m(x, t)}{\partial t} = \lambda_0 \nabla^2 \frac{\delta H}{\delta m(x, t)} + \Theta_m(x, t),$$

$$H = \int_V d^dx \left[ \frac{1}{2} \tau_0 \varphi^2 + \frac{1}{2} \nabla \varphi \nabla \varphi + \bar{u}_0 \varphi^4 + \frac{1}{2} m^2 + \gamma_0 m \varphi^2 - h_0 m \right]$$

where $\Theta_\varphi$ and $\Theta_m$ are Gaussian $\delta$-correlated random forces. We consider an equilibrium ensemble near $T_c(\bar{\rho})$ at fixed $\bar{\rho} = V^{-1} \int_V d^dx \rho(x, t)$. This corresponds to the experimental situation of keeping the conserved quantity (e.g., number of impurities) fixed when changing the reduced temperature $\hat{t} = [T - T_c(\bar{\rho})]/T_c(\bar{\rho})$. The latter enters through $\tau_0$. Because of $\langle \varphi \rangle = \bar{\rho}$ we have $\langle m \rangle = m = 0$. Eqs. (1) - (3) describe the dynamics of relaxational and diffusive modes that are coupled through $\gamma_0$. We are interested in the long-time behavior of the diffusive modes above, at and below $T_c$ as well as in the order-parameter relaxation on an intermediate time scale below $T_c$. We shall begin with the diffusive modes. For simplicity and for the purpose of a comparison with MC simulations, we assume cubic geometry, $V = L^d$, with periodic boundary conditions.

For the bulk system, the diffusion constants $D^\pm(\hat{t})$ above and below $T_c$ appear in the small $k$ limit of the long-time behavior of the correlation function

$$C_n(k, \hat{t}) = V^{-1} \langle m_k(t) n_{-k}(0) \rangle \sim \exp \left[-D^\pm(\hat{t}) k^2 \hat{t} \right]$$

where $m_k(t) = m_k(t) + c_n(k) \psi_k(t)$ is an appropriate linear combination of $m_k(t) = \int_V d^dx m(x, t) e^{-ikx}$ and $\psi_k(t) = \int_V d^dx [\varphi(x, t) - \langle \varphi \rangle] e^{-ikx}$ with $c_n(k) = \bar{c}_n k^2 + O(k^4)$. The coefficient $c_n$ can be identified by linearizing Eqs. (1) - (3). Above $T_c$, $c_n = 0$ because of $\langle \varphi \rangle = 0$. At $T_c$, the long-time behavior of $C_n$ is non-exponential (power law) for the bulk system.

For the finite system, the coefficient $c_n(k)$ is modified [via the replacement $\langle \varphi \rangle \to M_0$ as defined in Eqs. (1)]
and (8) below and the long-time behavior of $C_n$ remains exponential,
\[
C_n(k, \tilde{t}, L, t) \sim \exp \left[ -\Omega_n(k, \tilde{t}, L) t \right],
\]
even in the non-hydrodynamic region at bulk $T_c$ where the small-$k$ approximation is no longer justified. As a conceptual complication there exists a smallest nonzero value $k_{min}^2 = 4\pi^2/L^2$ of $k^2$ which prevents us to perform the limit $k \to 0$ for the finite system. Therefore we need to derive the finite-size scaling function for $\Omega_n(k, \tilde{t}, L)$ at finite $k$. Nevertheless we may define an effective diffusion time $\tau_D = \Omega_n(2\pi L^{-1}, \tilde{t}, L)^{-1}$ or a diffusion constant $D = \Omega_n/k^2$ at $k = k_{min} = 2\pi/L$ of the finite system by
\[
D(\tilde{t}, L) = (2\pi)^{-2}L^2 \Omega_n(2\pi L^{-1}, \tilde{t}, L)
\]
which interpolates smoothly between the bulk result $D(\tilde{t}, \infty) = D^+(\tilde{t})$ above and below $T_c$ (Fig. 1).

In the spirit of finite-size theory [9] we decompose $\varphi(x, t) = M_0 + \delta\varphi(x, t)$ with the zero-mode average
\[
M_0 = \int_{-\infty}^{\infty} dM \int_{-H_0}^{H_0} dM e^{-H_0} / \int_{-\infty}^{\infty} dM e^{-H_0} \tag{7}
\]
where $H_0(M) = L^d \left( \frac{1}{2} \sigma \varphi M^2 + \tilde{u}_0 M^4 \right)$ is the $k = 0$ part of $H$, with $M = V^{-1} \int_V d^d x \varphi$. For the finite system, the quantity $M_0(\sigma_0, L)$ is non-zero for all $T$ and interpolates smoothly between $T > T_c$ and $T < T_c$. Linearization of Eqs. (1) - (3) with respect to $\delta\varphi_k(t)$ and $m_k(t)$ leads to
\[
c_n(k) = (w_0 \gamma_0)^{-1} \left\{ b_0 - \left[ (b_0^2 + w_0 \gamma_0^2 k^2)^{1/2} \right] \right\},
\]
\[
\Omega_n(k, \tilde{t}, L) = \frac{1}{2} \lambda_0 \left[ b_0^2 - \left[ (b_0^2 + w_0 \gamma_0^2 k^2)^{1/2} \right] \right],
\]
\[
b_0(k) = w_0 \left[ \tau_0 + 12 \tilde{u}_0 M^2 + k^2 \right] \pm k^2,
\]
with $w_0 = \Gamma_0/\lambda_0$ and $\gamma_0 = 4\gamma_0 M_0$.

An application of these unrenormalized expressions to the critical region requires us to turn to the renormalized theory. The strategy of the field-theoretic renormalization-group (RG) approach at $d = 3$ dimensions is well established in bulk statics [8] and dynamics [9] and has been successfully applied recently to the model-A finite-size dynamics [9]. The details of its application to model $C$ will be given elsewhere [10]. Here we only present the asymptotic finite-size scaling form
\[
\Omega_n(k, \tilde{t}, L) = L^{-z} f_n(iL^{1-\alpha/\nu}, kL) \tag{11}
\]
as derived from Eqs. (3) and (8) - (10), with the dynamic critical exponent $z = 2 + \alpha/\nu$ [10]. The scaling function reads in three dimensions
\[
f_n(x, \nu) = A_n \tilde{\ell}^{\nu} \left\{ b_+ - \left[ (b_+^2 + w^* c^* \tilde{\ell}^{1/2} \kappa^2 \tilde{y}(\tilde{y}) \right]^{1/2} \right\},
\]
\[
\tilde{y}(\tilde{y}) = (4\pi \tilde{u}^*)^{-1/2} \left\{ \tilde{y}(\tilde{y}) + 12 \tilde{u}(\tilde{y}) \right\},
\]
\[
\tilde{\ell}(\tilde{y}) = (\int_0^{\infty} ds \int_{-\infty}^{\infty} s^2 e^{-\frac{1}{2}y s^2 - s^2})/(\int_0^{\infty} ds e^{-\frac{1}{2}y s^2 - s^2}),(c^*) = 16(\gamma^*)^2(4\pi \tilde{u}^*)^{1/2} and \tilde{x} = x \tilde{\ell}^{\nu} \tilde{y}(\tilde{y}).
\]
This yields the scaling form $D(\tilde{t}, L) = L^{-2-z} f_D(x)$ for the diffusion constant, Eq. (11), with
\[
f_D(x, \nu) = (2\pi)^{-2} f_n(x, 2\pi) \tag{12}
\]
The static parameters are [9] $\tilde{u}^* = u^* + (\gamma^*)^2/2$ and $(\gamma^*)^2 = \alpha(4\nu B(u^*))^{-1}$ with [11] $u^* = 0.0404$ and $B(u^*) = 0.502$ in three dimensions. For $\alpha$ and $\nu$ we take 0.6335 and 0.100 [12]. The dynamic parameter is $w^* = 1$ in one-loop order. The two non-universal bulk amplitudes $\xi_0(\tilde{\rho})$ and $A_n = \gamma_0^2 \xi_0^{1-\alpha} \tau_0^{1-\alpha}$ are defined by the asymptotic behavior $\xi(\tilde{\rho}) \sim \xi_0(\tilde{\rho})$ and $D(\tilde{t}) \sim \tilde{A}_n \tilde{L}^{1-\gamma_0^{1-\alpha}}$ of the correlation length and diffusion constant at fixed $\tilde{\rho}$ above $T_c$. The exponent $\nu/(1 - \alpha)$ instead of $\nu$ is due to Fisher renormalization [9].

The solid line in Fig. 3 shows $D(\tilde{t}, L)/A_D^+ \tilde{t}$ vs $\tilde{t}$ for the example $L = 80$ and the same line represents $f_D(x)$ vs $x$ (top scale). For comparison the bulk limits $D^\pm$ (dashed lines) are also shown, with $A_D^- = \gamma_0^2 \xi_0^{1-\alpha} \tau_0^{1-\alpha} = 0.55$. We expect the accuracy of these results to be of $O(10\%)$. These predictions can be tested by MC simulations, after adjusting $\xi_0(\tilde{\rho})$ and $A_D^+$ in the bulk region $x \gg 1$ above $T_c$. In addition to the finite-size effect on the diffusive modes there exists an interesting finite-size effect on the relaxational modes below $T_c$ that has so far not been investigated analytically. It is well known that no spontaneous symmetry breaking can take place in finite systems below $T_c$ because of ergodicity. For Ising-like systems, ergodicity implies a “tunneling” between metastable states...
of opposite orientation of the magnetization as observed in MC simulations. On an intermediate time scale \( t < t_x(L) \), however, the magnetization does not change sign and its magnitude relaxes towards a finite value that characterizes such a metastable state. This relaxation process is important for large systems since the crossover time \( t_x(L) \) is expected to grow with the size \( L \) as \( \sim L^z \) where \( z \) is the dynamic critical exponent. This process occurs both in model A and model C, therefore we confine ourselves to the simpler model A in the following. We stress that the relaxation process for \( t < t_x(L) \) is fundamentally different from the ultimate long-time behavior for \( t \gg t_x(L) \) studied previously.

Model A is defined by Eq. (1), where \( H \) is replaced by

\[
H_{\varphi} = \int d^d x \left[ \frac{1}{2} r_0 \varphi^2 + \frac{1}{2} (\nabla \varphi)^2 + u_0 \varphi^4 \right].
\]  

(13)

We consider the time-dependent spatial average \( M(t, L) = L^{-d} \int d^d x \varphi(x, t) \). We are primarily interested in the long-time behavior of the equilibrium correlation function \( \langle M(t, L)M(0, L) \rangle \equiv C(t, L) \) for \( d = 3 \). For the bulk system, this behavior is

\[
C(t, \infty) \sim A_6^+ \exp(-t/\tau_6^+) ,
\]

\[
C(t, \infty) - M^2_{\text{sp}} \sim A_6^- \exp(-t/\tau_6^-)
\]

(15)

above and below \( T_c \), where \( M_{\text{sp}} = \lim_{L \to \infty} \langle M(t, \infty) \rangle \) is the spontaneous order parameter and \( \tau_6^\pm \) are the bulk relaxation times. For the finite system above \( T_c \), the leading time dependence is still a single exponential \( \sim c_1 e^{-t/\tau_1(L)} \) with a relaxation time \( \tau_1(L) \) whose finite-size scaling function is known both analytically and numerically. In particular, \( \lim_{L \to \infty} \tau_1(L) = \tau_6^+ \).

For the finite system below \( T_c \), however, the situation is more complicated and considerably less well explored. MC simulations and phenomenological considerations suggest that there should exist an L dependent generalization \( \tau^{-}(L) \) of \( \tau_6^- \), with \( \lim_{L \to \infty} \tau^{-}(L) = \tau_6^- \), which should describe (i) the exponential relaxation of \( \langle M(t, L) \rangle \) towards a metastable finite value on an intermediate time scale \( t < t_x(L) \) before tunneling sets in, and (ii) a corresponding exponential decay of \( C(t, L) \) on this time scale. The question arises whether and how this important relaxation time \( \tau^{-}(L) \) can be identified analytically within models A and C. This question was left unanswered in the previous literature. In particular, neither \( \tau_1(L) \) nor \( \tau_2(L) \) as calculated previously, can be identified with \( \tau^{-}(L) \). Below \( T_c \), \( \tau_1(L) \) describes the decay of \( \langle M(t, L) \rangle \) and of \( C(t, L) \) towards zero for \( t \gg t_x(L) \) due to tunneling processes, and \( \tau_2(L) \) describes the decay of \( \langle M(t)^2 \rangle \) and of \( \langle M(t)^2M(0)^2 \rangle \) towards \( \langle M^2 \rangle_{eq} \) and \( \langle M^2 \rangle_{eq}^2 \), respectively, for \( t \gg t_x(L) \). In the following we establish an analytic identification of \( \tau^{-}(L) \) and present the first quantitative prediction for its finite-size scaling behavior.

To elucidate the main features we first neglect the inhomogeneous fluctuations \( \sigma(x, t) = \varphi(x, t) - M(t) \). Then Eq. (1) is equivalent to the Fokker-Planck equation

\[
\frac{\partial P(M, t)}{\partial t} = -L_0 P(M, t) \text{ for the probability distribution} \ P(M, t)
\]

with \( c_6(L) = \int \frac{\partial M}{\partial M} (\partial H_0(M) + \partial M) \)

(16)

where \( H_0(M) = \frac{1}{2} r_0 M^2 + u_0 M^4 \). It is well known that \( C(t, L) \) is determined by the eigenvalues \( \epsilon_k \) and eigenfunctions \( \phi_k(M) \) of \( L_0 \) according to

\[
C(t, L) = \sum_{k=1}^\infty c_k(L) \exp[-t/\tau_k(L)] , \ t > 0
\]

(17)

with \( c_k(L) = [\int_{-\infty}^{\infty} dM \ M \phi_k(M)]^2 \) and \( \tau_k(L) = \epsilon_k^{-1} \), \( \epsilon_0 = 0 \leq \epsilon_1 \leq \epsilon_2 \ldots \) By symmetry, \( c_k = 0 \) for even values of \( k \). Below \( T_c \), \( \tau_1(L) \) diverges in the bulk limit and \( \lim_{L \to \infty} c_1 e^{-t/\tau_1} = M_{\text{sp}}^2 \) becomes time-independent, thus an analysis of the \( k = 3 \) term in Eq. (17) becomes indispensable. From the spectrum of \( L_0 \) we find a degeneracy for \( k = 3 \) and \( k = 5 \) in the bulk limit for \( r_0 < 0 \). This requires to take the \( k = 5 \) term into account as well. We have found, however, that the coefficient \( c_5 \) vanishes in the bulk limit below \( T_c \) whereas \( c_3 \) remains finite. For finite \( L \) near \( T_c \), \( \tau_3 \) is well separated from \( \tau_5 \) as shown below. Thus it suffices to describe the time dependence of \( C(t, L) \) on intermediate time scales \( t \sim O(\tau^{-}(L)) \) and \( O(\tau^{-}(L)) < t < O(\tau_3(L)) \) as well as on the long-time scale \( t \gg \tau_3(L) \) as

\[
C(t, L) \sim c_1(L)e^{-t/\tau_1(L)} + c_3(L)e^{-t/\tau_3(L)}
\]

(18)

where \( c_1(\infty) = M_{\text{sp}}^2 \) and \( c_3(\infty) = A_6^- \) below \( T_c \). In particular we arrive at the desired identification

\[
\tau^{-}(L) \equiv \tau_3(L) , \ \lim_{L \to \infty} \tau_3(L) = \tau_6^- .
\]

(19)

We conclude that, although \( \tau_3(L) \) represents only a subleading relaxation time above \( T_c \), \( \tau_3(L) \) governs the leading time dependence of \( C(t, L) \) of large finite systems below \( T_c \) (Fig. 3).

These results yield also the key to the interpretation of \( \tau_3(L) \) as the relaxation time governing the approach of the non-equilibrium quantity \( \langle M(t, L) \rangle \) towards a metastable finite value before \( M(t, L) \) starts to change sign. This interpretation is based on the fact that the leading relaxation times of \( \langle M(t, L) \rangle \) are determined by the same eigenvalues of \( L_0 \) as the long-time behavior of the equilibrium correlation function \( C \), i.e.,

\[
\langle M(t, L) \rangle \sim \hat{c}_1(L)e^{-t/\tau_1(L)} + \hat{c}_3(L) e^{-t/\tau_3(L)}
\]

(20)

The basic difference between Eqs. (14) and (15) is that the coefficients \( \hat{c}_k \) depend on the initial (non-equilibrium) state.
We proceed by presenting the results of a quantitative calculation of $\tau_2(L)$ and $\tau_3(L)$ including the effect of the inhomogeneous fluctuations $\sigma(x)$ to one-loop order. This calculation is parallel to that performed previously [3] and is expected to be as reliable as the previous results [3]. It is based on the Fokker-Planck equation $\partial P(M,t)/\partial t = -\nabla_1 P(M,t)$ where $\nabla_1$ has the same structure as $\mathcal{L}_0$, Eq. (13), but with $\Gamma_0, u_0, \Gamma_0$ replaced by (positive) effective parameters $\Gamma_0^{eff}, u_0^{eff}, \Gamma_0^{eff}$. In terms of the eigenvalues $\mu_3(\kappa)$ and $\mu_5(\kappa)$ of the equivalent Schrödinger equation [4] we determine the relaxation times $\tau_2$ and $\tau_3$ as

$$\tau_i = (2\Gamma_0^{eff})^{-1}L^{d/2}(u_0^{eff})^{-1/2} \mu_i(\kappa)$$

with $\kappa = 1/2 \Gamma_0^{eff} L^{d/2} (u_0^{eff})^{-1/2}$. In the asymptotic region the field-theoretic RG approach at $d = 3$ [3,15,16] yields the finite-size scaling form $\tau_i = L^2 f_i(x), i = 3, 5$, with the scaling variable $x = t L^{1/\nu}, t = (T - T_c)/T_c$. The analytic expressions for $f_i(x)$ are analogous to those given previously [4] and will be given elsewhere [10]. At $T_c$ we predict the universal ratios $\tau_1/\tau_3 = 8.5$ and $\tau_3/\tau_5 = 2.3$.

![Figure 2](image-url)

**FIG. 2.** Relaxation times $\tau_i(\tilde{t},L)/A_{\tilde{t}}$ for $L = 80\tilde{a}$ (solid lines) vs $\tilde{t} = (T - T_c)/T_c$, and of their scaling functions $f_i(x)$ vs $x = t L^{1/\nu}$ (solid lines), with $L$ in units of the lattice constant $\tilde{a}$. Dashed lines: bulk relaxation times $\tau_i^c(\tilde{t})/A_{\tilde{t}}^c$.

The results are shown in Fig. 2. For an application to the Ising model we have taken $\xi_0/\tilde{a} = 0.495$ [12] where $\tilde{a}$ is the lattice spacing. The relaxation times $\tau_i$ in Fig. 2 are normalized to the bulk amplitude $A_{\tau}^{c}$ of $\tau_0 = A_{\tau} \tilde{t}^{-\nu z}$, $z = 2.04$ (dashed line above $T_c$). Below $T_c$, our theory yields the expected exponential decay, Eq. (13), for the $d = 3$ bulk system, in agreement with Ref. [22]. The dashed line below $T_c$ represents the bulk relaxation time $\tau_b = A_{\tau b} \tilde{t}^{-\nu z}$ with $A_{\tau b}/A_{\tau}^{c} = 2^{-\nu z} (1 + 18u^*)/(1 + 18u^*) = 0.26$ in three dimensions. Unlike for $\tau_1$ and $\tau_2$ [3,15,16], no MC data are presently available for $\tau_3$.

In summary we have presented the first quantitative predictions for the finite-size effects on critical diffusion and order-parameter relaxation towards metastable equilibrium in three-dimensional systems near $T_c$. It would be interesting to test the predicted universal ratios $\tau_1/\tau_3$, $\tau_3/\tau_5$ and the finite-size scaling functions $f_2(x)$ and $f_3(x)$ (Figs. 1 and 2) by MC simulations. This appears to be within reach of present simulation techniques [23].

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