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RAPID COMMUNICATIONS

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Finite-size effects on critical diffusion and relaxation towards metastable equilibrium

W. Koch and V. Dohm
Institut für Theoretische Physik, Technische Hochschule Aachen, D-52056 Aachen, Germany
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We present an 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 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 nonconserved order parameter is fairly 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 [1,2]. The fundamental dynamic quantities of these systems are the relaxation and diffusion times that 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 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 for two relaxation times τ_1 and τ_2, which determines the long-time behavior of the order parameter and the square of the order parameter [3,4]. No analytic work exists, to the best of our knowledge, on the important universality class [1] of diffusive finite-size behavior near T_c. This is of relevance, e.g., to magnetic systems with mobile impurities [1], to binary alloys with an order-disorder transition [5,6], to uniaxial antiferromagnets [1], or to systems in which the order parameter is coupled to the conserved energy density [1,2].

In this Rapid Communication we present a 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 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 $\varphi(\mathbf{x},t)$ and for the density $\rho(\mathbf{x},t)=\langle\rho\rangle+m(\mathbf{x},t)$ in a finite volume V,

$$\frac{\partial\varphi(\mathbf{x},t)}{\partial t}=-\Gamma_0\frac{\delta H}{\delta\varphi(\mathbf{x},t)}+\Theta_\varphi(\mathbf{x},t), \tag{1}$$

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

$$H=\int_V d^dx[\tfrac{1}{2}\tau_0\varphi^2+\tfrac{1}{2}(\nabla\varphi)^2+\tilde{u}_0\varphi^4+\tfrac{1}{2}m^2 + \gamma_0 m\varphi^2-h_0 m], \tag{3}$$

where Θ_φ and Θ_m are Gaussian δ-correlated random forces. We consider an equilibrium ensemble near $T_c(\overline{\rho})$ at fixed

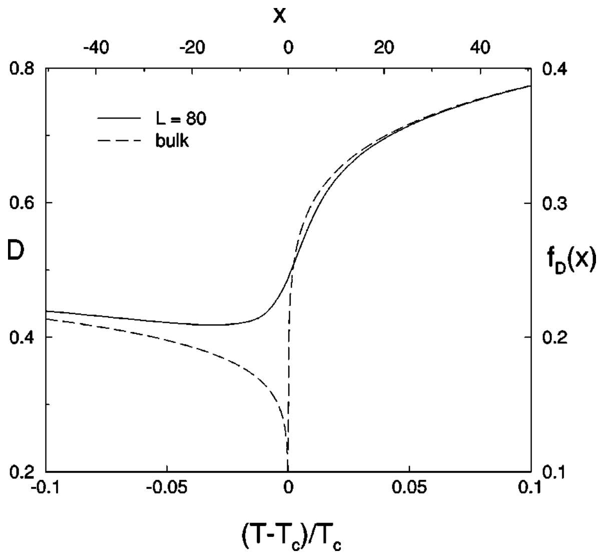

FIG. 1. Diffusion constant $D(\tilde{t},L)/A_D^+$ for $L=80\xi_0$ (solid line) vs $\tilde{t}$, and of the scaling function $f_D(x)$ [Eq. (12)] vs $x=\tilde{t}L^{(1-\alpha)/\nu}$ (solid line), with L in units of ξ_0. The dashed lines represent the bulk diffusion constants $D^\pm(\tilde{t})/A_D^+$.

$\bar{\rho}=V^{-1}\int_V d^dx\rho(\mathbf{x},t)$. This corresponds to the experimental situation of keeping the conserved quantity (e.g., number of impurities) fixed when changing the reduced temperature $\tilde{t}=[T-T_c(\bar{\rho})]/T_c(\bar{\rho})$. The latter enters through τ_0. Because of $\langle\rho\rangle=\bar{\rho}$ we have $\langle m\rangle=\bar{m}=0$. Equations (1)–(3) describe the dynamics of relaxational and diffusive modes that is coupled through γ_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 $D^\pm(\tilde{t})$ above and below T_c appear in the small-k limit of the long-time behavior of the correlation function

$$C_n(k,\tilde{t},t)=V^{-1}\langle n_{\mathbf{k}}(t)n_{-\mathbf{k}}(0)\rangle\sim\exp[-D^\pm(\tilde{t})k^2t], \quad (4)$$

where $n_{\mathbf{k}}(t)=m_{\mathbf{k}}(t)+c_n(k)\psi_{\mathbf{k}}(t)$ is an appropriate linear combination of $m_{\mathbf{k}}(t)=\int_V d^dx m(\mathbf{x},t)e^{-i\mathbf{k}\cdot\mathbf{x}}$ and $\psi_{\mathbf{k}}(t)=\int_V d^dx[\varphi(\mathbf{x},t)-\langle\varphi\rangle]e^{-i\mathbf{k}\cdot\mathbf{x}}$ with $c_n(k)=\tilde{c}_nk^2+O(k^4)$. 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 nonexponential (power law) for the bulk system.

For the *finite* system, the coefficient $c_n(k)$ is modified [via the replacement $\langle\varphi\rangle\rightarrow M_0$ as defined in Eqs. (7) and (8) below] and the long-time behavior of C_n remains exponential;

$$C_n(k,\tilde{t},L,t)\sim\exp[-\Omega_n(k,\tilde{t},L)t], \quad (5)$$

even in the nonhydrodynamic 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 from performing the limit $k\rightarrow 0$ 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

$$D(\tilde{t},L)=(2\pi)^{-2}L^2\Omega_n(2\pi L^{-1},\tilde{t},L), \quad (6)$$

which interpolates smoothly between the bulk result $D(\tilde{t},\infty)=D^\pm(\tilde{t})$ above and below T_c (Fig. 1).

In the spirit of finite-size theory [3,7] we decompose $\varphi(\mathbf{x},t)=M_0+\delta\varphi(\mathbf{x},t)$ with the zero-mode average

$$M_0^2=\int_{-\infty}^{\infty}dM M^2e^{-H_0}\Big/\int_{-\infty}^{\infty}dMe^{-H_0}, \quad (7)$$

where $H_0(M)=L^d(\frac{1}{2}\tau_0M^2+\tilde{u}_0M^4)$ is the $k=0$ part of H, with $M=V^{-1}\int_V d^dx\varphi$. For the finite system, the quantity $M_0(\tau_0,L)$ is nonzero for all T and interpolates smoothly between $T>T_c$ and $T<T_c$. Linearization of Eqs. (1)–(3) with respect to $\delta\varphi_{\mathbf{k}}(t)$ and $m_{\mathbf{k}}(t)$ leads to

$$c_n(k)=(w_0\tilde{\gamma}_0)^{-1}\{b_0^- -[(b_0^-)^2+w_0\tilde{\gamma}_0^2k^2]^{1/2}\}, \quad (8)$$

$$\Omega_n(k,\tilde{t},L)=\tfrac{1}{2}\lambda_0\{b_0^+ -[(b_0^-)^2+w_0\tilde{\gamma}_0^2k^2]^{1/2}\}, \quad (9)$$

$$b_0^\pm(k)=w_0(\tau_0+12\tilde{u}_0M_0^2+k^2)\pm k^2, \quad (10)$$

with $w_0=\Gamma_0/\lambda_0$ and $\tilde{\gamma}_0=4\gamma_0M_0$.

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 [3]. The details of its application to model C will be given elsewhere [10]. Here we only present the asymptotic finite-size scaling form

$$\Omega_n(k,\tilde{t},L)=L^{-z}f_n(\tilde{t}L^{(1-\alpha)/\nu},kL) \quad (11)$$

as derived from Eqs. (5) and (8)–(10), with the dynamic critical exponent $z=2+\alpha/\nu$ [2]. The scaling function reads in three dimensions:

$$f_n(x,\kappa)=A_n\tilde{\ell}^{\alpha/\nu}\{b_+-[b_-^2+w^*c^*\tilde{\ell}^{1/2}\kappa^2\vartheta_2(\tilde{y})]^{1/2}\},$$

$$b_\pm(x,\kappa)=w^*[\tilde{\ell}(x)^2+\kappa^2]\pm\kappa^2,$$

$$\tilde{\ell}(x)^{3/2}=(4\pi\tilde{u}^*)^{1/2}\{\tilde{y}(x)+12\vartheta_2[\tilde{y}(x)]\},$$

$$\tilde{y}(x)=(4\pi\tilde{u}^*)^{-1/2}\tilde{\ell}(x)^{(3/2)-(1-\alpha)/\nu}\hat{x},$$

$$\vartheta_2(y)=\left(\int_0^\infty ds\ s^2e^{-1/2ys^2-s^4}\right)\Big/\left(\int_0^\infty ds\ e^{-1/2ys^2-s^4}\right),$$

with $c^*=16(\gamma^*)^2(4\pi/\tilde{u}^*)^{1/2}$ and $\hat{x}=x\xi_0^{-(1-\alpha)/\nu}$. This yields the scaling form $D(\tilde{t},L)=L^{2-z}f_D(x)$ for the diffusion constant [Eq. (6)] with

$$f_D(x)=(2\pi)^{-2}f_n(x,2\pi). \tag{12}$$

The static parameters are [8] $\tilde{u}^*=u^*+(\gamma^*)^2/2$ and $(\gamma^*)^2=\alpha[4\nu B(u^*)]^{-1}$ with [11] $u^*=0.0404$ and $B(u^*)=0.502$ in three dimensions. For ν and α we take 0.6335 and 0.100 [12]. The dynamic parameter is $w^*=1$ in one-loop order. The two nonuniversal bulk amplitudes $\xi_0(\overline{\rho})$ and $A_n=\frac{1}{2}A_D^+\xi_0^{z-2}$ are defined by the asymptotic behavior $\xi=\xi_0\tilde{t}^{-\nu/(1-\alpha)}$ and $D^+(\tilde{t})=A_D^+\tilde{t}^{(z-2)\nu/(1-\alpha)}$ of the correlation length and diffusion constant at fixed $\overline{\rho}$ above T_c. The exponent $\nu/(1-\alpha)$ instead of ν is due to Fisher renormalization [13].

The solid line in Fig. 1 shows $D(\tilde{t},L)/A_D^+$ vs $\tilde{t}$ for the example $L=80\xi_0$. The same line represents $f_D(x)$ vs x (top scale). For comparison the bulk limits $D^\pm$ (dashed lines) are also shown, with $A_D^-/A_D^+=2^{\alpha/(1-\alpha)}(1+\frac{1}{2}\gamma^{*2}/u^*)^{-1}=0.55$. We expect the accuracy of these results to be of O (10%). These predictions can be tested by MC simulations, after adjusting $\xi_0(\overline{\rho})$ and A_D^+ in the bulk region $x\gg1$ 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 to our knowledge not been investigated analytically so far. 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 [14–16]. 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 [14]. 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 [3,4].

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

$$H_\varphi=\int_V d^dx[\tfrac{1}{2}r_0\varphi^2+\tfrac{1}{2}(\nabla\varphi)^2+u_0\varphi^4]. \tag{13}$$

We consider the time-dependent spatial average $M(t,L)=L^{-d}\int_V d^dx\ \varphi(\mathbf{x},t)$. 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

$$C(t,\infty)\sim A_b^+\exp(-t/\tau_b^+), \tag{14}$$

$$C(t,\infty)-M_{sp}^2\sim A_b^-\exp(-t/\tau_b^-) \tag{15}$$

above and below T_c, where $M_{sp}=\lim_{t\to\infty}\langle M(t,\infty)\rangle$ is the spontaneous order parameter and $\tau_b^\pm$ are the bulk relaxation times. For the finite system *above* T_c, the leading time dependence is still a single exponential $\sim c_1e^{-t/\tau_1(L)}$ with a relaxation time $\tau_1(L)$ whose finite-size scaling function is known both analytically [3,4] and numerically [3,16]. In particular, $\lim_{L\to\infty}\tau_1(L)=\tau_b^+$.

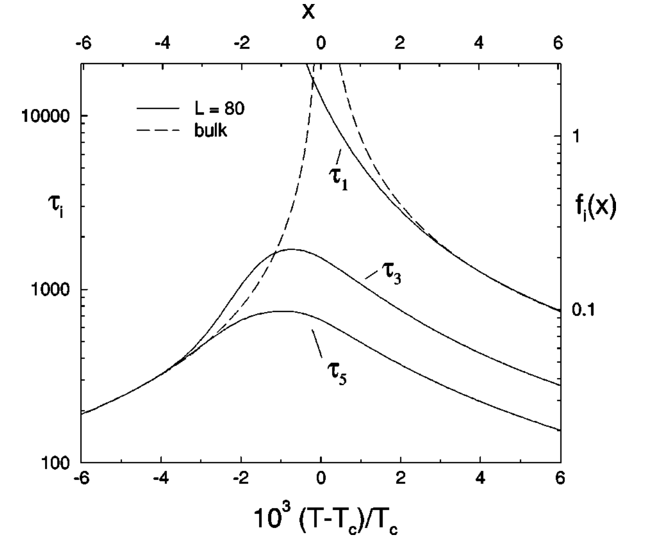

FIG. 2. Relaxation times $\tau_i(\tilde{t},L)/A_{\tau b}^+$ for $L=80\tilde{a}$ (solid lines) vs $\tilde{t}=(T-T_c)/T_c$, and of their scaling functions $f_i(x)$ vs $x=\tilde{t}L^{1/\nu}$ (solid lines), with L in units of the lattice constant $\tilde{a}$; dashed lines, bulk relaxation times $\tau_b^\pm(\tilde{t})/A_{\tau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

For the finite system *below* T_c, however, the situation is more complicated and considerably less well explored. MC simulations [14] and phenomenological considerations suggest that there should exist an L dependent generalization $\tau^-(L)$ of τ_b^-, with $\lim_{L\to\infty}\tau^-(L)=\tau_b^-$, 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 [3], can be identified with $\tau^-(L)$. [Below T_c,τ_1 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 τ_2 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 a quantitative prediction for its finite-size scaling behavior.

To elucidate the main features we first neglect the inhomogeneous fluctuations $\sigma(\mathbf{x},t)=\varphi(\mathbf{x},t)-M(t)$. Then Eq. (1) is equivalent to the Fokker-Planck equation $\partial P(M,t)/\partial t=-\mathcal{L}_0P(M,t)$ for the probability distribution $P(M,t)$ with the operator

$$\mathcal{L}_0=-\frac{\Gamma_0}{L^d}\frac{\partial}{\partial M}\left(\frac{dH_0(M)}{dM}+\frac{\partial}{\partial M}\right), \tag{16}$$

where $H_0(M)=L^d(\frac{1}{2}r_0M^2+u_0M^4)$. It is well known [17] that $C(t,L)$ is determined by the eigenvalues ϵ_k and eigenfunctions $\phi_k(M)$ of $\mathcal{L}_0$ according to

$$C(t,L)=\sum_{k=1}^{\infty} c_k(L)\exp[-t/\tau_k(L)], \quad t>0, \tag{17}$$

with $c_k(L)=[\int_{-\infty}^{\infty} dMM\phi_k(M)]^2$ and $\tau_k(L)=\epsilon_k^{-1}$, $\epsilon_0=0 \leqslant \epsilon_1 \leqslant \epsilon_2 \ldots$. By symmetry, $c_k=0$ for even values of k. Below T_c, $\tau_1(L)$ diverges in the bulk limit and $\lim_{L\to\infty} c_1 e^{-t/\tau_1}=M_{sp}^2$ becomes time independent, thus an analysis of the $k=3$ term in Eq. (17) becomes indispensable. From the spectrum of $\mathcal{L}_0$ [17] we find a degeneracy for $k=3$ and $k=5$ in the bulk limit for $r_0<0$. This requires us 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, τ_5 is well separated from τ_3, 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 $O[\tau^-(L)]<t<O[\tau_1(L)]$ as well as on the long-time scale $t\gg\tau_1(L)$ as

$$C(t,L)\sim c_1(L)e^{-t/\tau_1(L)}+c_3(L)e^{-t/\tau_3(L)}, \tag{18}$$

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

$$\tau^-(L)\equiv\tau_3(L), \quad \lim_{L\to\infty}\tau_3(L)=\tau_b^- . \tag{19}$$

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. 2).

These results also yield the key to the interpretation of $\tau_3(L)$ as the relaxation time governing the approach of the *nonequilibrium* 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 [18] that the leading relaxation times of $\langle M(t,L)\rangle$ are determined by the same eigenvalues of $\mathcal{L}_0$ as the long-time behavior of the equilibrium correlation function C, i.e.,

$$\langle M(t,L)\rangle\sim\tilde{c}_1(L)e^{-t/\tau_1(L)}+\tilde{c}_3(L)e^{-t/\tau_3(L)}. \tag{20}$$

The basic difference between Eqs. (20) and (18) is that the coefficients $\tilde{c}_k$ depend on the initial (nonequilibrium) state.

We proceed by presenting the results of a quantitative calculation of $\tau_3(L)$ and $\tau_5(L)$, including the effect of the inhomogeneous fluctuations $\sigma(\mathbf{x})$ 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 $\partial P(M,t)/\partial t=-\mathcal{L}_1 P(M,t)$, where $\mathcal{L}_1$ has the same structure as $\mathcal{L}_0$ [Eq. (16)] but with r_0,u_0,Γ_0 replaced by (positive) effective parameters $r_0^{eff},u_0^{eff},\Gamma_0^{eff}$ [3,7,19]. In terms of the eigenvalues $\mu_3(\kappa)$ and $\mu_5(\kappa)$ of the equivalent Schrödinger equation [17] we determine the relaxation times τ_3 and τ_5 as

$$\tau_i=(2\Gamma_0^{eff})^{-1}L^{d/2}(u_0^{eff})^{-1/2}\mu_i(\kappa), \tag{21}$$

with $\kappa=\frac{1}{2}r_0^{eff}L^{d/2}(u_0^{eff})^{-1/2}$. In the asymptotic region the field-theoretic RG approach at $d=3$ [3, 7–9] yields the finite-size scaling form $\tau_i=L^z f_i(x)$, $i=3,5$, with the scaling variable $x=\tilde{t}L^{1/\nu}$, $\tilde{t}=(T-T_c)/T_c$. The analytic expressions for $f_i(x)$ are analogous to those given previously [3] and will be given elsewhere [10]. At T_c we predict the universal ratios $\tau_1/\tau_3=8.5$ and $\tau_3/\tau_5=2.3$.

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 τ_i in Fig. 2 are normalized to the bulk amplitude $A_{\tau b}^+$ of $\tau_b^+=A_{\tau b}^+\tilde{t}^{-\nu z}$, $z=2.04$ (dashed line above T_c). Below T_c, our theory yields the expected [20,21] exponential decay [Eq. (15)] for the $d=3$ bulk system, in disagreement with Ref. [22]. The dashed line below T_c represents the bulk relaxation time $\tau_b^-=A_{\tau b}^-|\tilde{t}|^{-\nu z}$ with $A_{\tau b}^-/A_{\tau b}^+=2^{-\nu z}(1+\frac{9}{4}u^*)/(1+18u^*)=0.26$ in three dimensions. Unlike for τ_1 and τ_2 [3,15,16], no MC data are presently available for τ_3.

In summary we have presented 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 τ_1/τ_3, τ_3/τ_5 and the finite-size scaling functions $f_D(x)$ and $f_i(x)$ (Figs. 1 and 2) by MC simulations. This appears to be within reach of present simulation techniques [23].

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