Calculation of dynamic critical properties from a cluster-reaction theory

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Critical fluctuations in kinetic Ising models are interpreted in terms of cluster reactions. The basic assumption, that clusters with l spins grow at a rate $\propto l^r$, is tested by Monte Carlo computations in the single-spin-flip case. The dynamic susceptibilities associated with order parameter and energy are then calculated also for nonzero magnetic field, and are shown to fulfill dynamic scaling. The exponent $(2-r)\beta\delta$ of the relaxation times can be different from the susceptibility exponent γ.

I. INTRODUCTION

The critical dynamics of kinetic Ising models[1] has recently received great attention: (i) In contrast to models where the dynamics is governed by critical propagating modes, the dynamic critical exponents cannot in general be expressed only by static ones by use of dynamic scaling.[2,3] In particular, dynamic scaling does not determine $\Delta_{\mu\mu}$, the exponent of the order-parameter relation time $\tau_{\mu\mu}$,

$$\tau_{\mu\mu} = \int_0^\infty \phi_{\mu\mu}(t)\,dt\,, \tag{1}$$

$$\phi_{\mu\mu}(t) = \frac{\langle \mu(0)\mu(t)\rangle - \langle\mu\rangle^2}{\langle\mu^2\rangle - \langle\mu\rangle^2}\,, \qquad \tau_{\mu\mu} = \tau^0_{\mu\mu}\epsilon^{-\Delta_{\mu\mu}}, \quad \epsilon = 1 - T/T_c\,. \tag{2}$$

(ii) Renormalization-group techniques,[4,5] high-temperature series,[6] and Monte Carlo studies[7,8] have shown that $\Delta_{\mu\mu}$ differs from the susceptibility exponent γ in single-spin-flip (Glauber[9]) models. (iii) This result is in contrast to mode-mode coupling theories of anisotropic magnets,[10] which predict

$$\Delta_{\mu\mu} = \gamma\,, \tag{3}$$

which is also the result of the conventional theory.[1]

No simple physical interpretation for the result $\Delta_{\mu\mu} > \gamma$ and no detailed satisfactory theory of the slowing down exists. In the present work we treat this problem with a cluster reaction model.[11] In Sec. II, we introduce the model, and briefly discuss the basic approximations involved in it. In Sec. III, we derive the dynamic critical properties of this model. For an explicit calculation of critical amplitudes we use the Fisher droplet model[12] as an example, and we compare these explicit results to corresponding Monte Carlo calculations. Section IV then contains our conclusions.

II. CLUSTER-REACTION MODEL

The kinetic Ising model is described by a master equation for the probability distribution $|P_\mu(t)\rangle$ of the spin

$$\frac{d}{dt}\,|P_\mu(t)\rangle = L_\mu\,|P_\mu(t)\rangle\,, \tag{4}$$

where the operator L_μ can be specified explicitly in terms of spin-flip transition probabilities.[1] Denoting a group of l reversed spins linked together by nearest-neighbor bonds as a "cluster," one may describe the state of the system by its cluster configuration. Spin flips then produce cluster reactions (we also count the creation of a single reversed spin—a $l=1$ cluster—as a cluster reaction). Discussing the nonequilibrium relaxation[11,13] it was pointed out that it is most important to derive the averaged cluster concentrations $\bar{n}_l(t)$. Considering now the response of the cluster concentration to a small change δe of an external parameter e, we denote by $n_l^{\delta e}$ the cluster distribution which is in thermal equilibrium with the applied parameter $e+\delta e$, and $n_l = n_l^{\delta e=0}$. It was shown[12,13] that it is then reasonable to replace Eq. (4) by

$$\frac{\partial}{\partial t}\,|P_l(t)\rangle = L_l\,|P_l(t)\rangle\,, \tag{5}$$

where $|P_l(t)\rangle$ means a distribution of cluster concentrations $\{\bar{n}_l(t)\}$, and L_l is given by

$$L_l = \frac{\partial}{\partial l}\,D_l n_l^{\delta e}\left(\frac{\partial}{\partial l}\right)\frac{1}{n_l^{\delta e}}\,. \tag{6}$$

The growth rate D_l is given by the probability $W(l,l')$ that a cluster with l' spins is incorporated[11,14] into the l cluster,

$$D_l \equiv \frac{1}{n_l}\sum_{l'} l'^2 W(l,l')\,. \tag{7}$$

We expect D_l to have asymptotically a power-law

behavior

$$D_l = \tilde{D}(l) l^r , \tag{8}$$

with $\tilde{D}(\infty)$ being finite and nonzero.

Approximating Eq. (4) by Eq. (5) one is free to redefine what is meant precisely by a cluster; thus we may consider various models for the static cluster distribution n_l with different meanings of the "cluster index" l. In the following we consider a class of models, where n_l depends on the magnetic field h only in the combination hl

$$n_l = f(l, \epsilon, hl) , \tag{9}$$

and where the critical part of the magnetization is given by

$$m \propto -\sum_l l n_l . \tag{10}$$

The physical meaning of Eq. (10) is that the system can essentially be interpreted in terms of a "non-interacting cluster gas." Thus the contribution of cluster interactions to the free energy (e.g., due to excluded volume effects) have to be taken into account already by a suitable choice of the n_l in Eq. (9). Equations (9) and (10) are consistent with static scaling, if we have

$$n_l = \tilde{q}(l) l^{-2-1/\delta} \tilde{f}(\epsilon l^{1/\beta\delta}, hl) , \tag{11}$$

with $\tilde{q}(\infty)$ finite and nonzero. Obviously, the Fisher cluster model[12]

$$n_l = \tilde{q}(\infty) l^{-2-1/\delta} \exp(-b\epsilon l^{1/\beta\delta} - hl) , \tag{12}$$

which we use below as an explicit example, is a special case of Eq. (11). In Eq. (12), $\tilde{q}(\infty)$ and b are related to critical amplitudes and hence the static scaling function is uniquely determined [using Eqs. (9), (10), and (12)], which is of course only an approximation. Equation (11), on the other hand, could still be chosen in full accordance with the exact static critical behavior. Hence, the shortcomings of Eq. (12) do not apply on Eq. (11), on which most of our calculations will be based.

We consider now time-dependent changes of field h or temperature T,

$$\delta h(t) = e^{i\omega t} \delta h_\omega , \tag{13}$$

$$\delta T(t) = e^{i\omega t} \delta T_\omega , \tag{14}$$

and introduce response functions $g_l^\mu(\omega)$ and $g_l^E(\omega)$

$$\bar{n}_l(t) = n_l [1 - \delta h(t) g_l^\mu(\omega)] \tag{15}$$

or

$$\bar{n}_l(t) = n_l \{1 + [b\delta T(t)/T_c] g_l^E(\omega)\} . \tag{16}$$

From Eqs. (10), (11), (15), and (16) one derives the dynamic susceptibilities

$$\chi_{\mu\mu}(\omega) = \sum_l l n_l g_l^\mu(\omega) , \tag{17}$$

$$\chi_{\mu E}(\omega) = \sum_l l n_l g_l^E(\omega) , \tag{18}$$

and subsequently one obtains the relaxation times expanding[15]

$$\chi_{\mu\mu}(\omega) = k_B T \chi_{\mu\mu} (1 - i\omega\tau_{\mu\mu} + - \cdots) . \tag{19}$$

If the critical part of the free energy is given by

$$-k_B T \sum_l n_l$$

as in the case of the Fisher droplet model, then it is straightforward to derive also $\chi_{EE}(\omega)$:

$$\chi_{EE}(\omega) = \sum_l \frac{\partial n_l}{\partial(1/T)} g_l^E(\omega) . \tag{20}$$

Equations (5)–(7) and Eqs. (15) and (16) yield differential equations for g_l^μ and g_l^E:

$$\frac{d^2}{dl^2}[g_l^\mu(\omega) - g_l^\mu(0)] + \left\{\frac{d}{dl}[g_l^\mu(\omega) - g_l^\mu(0)]\right\} \frac{d}{dl} \times [\ln(n_l D_l)] - \frac{i\omega g_l^\mu}{D_l} = 0 \tag{21}$$

and

$$\frac{d^2}{dl^2}[g_l^E(\omega) - g_l^E(0)] + \left\{\frac{d}{dl}[g_l^E(\omega) - g_l^E(0)]\right\} \frac{d}{dl} \times [\ln(n_l D_l)] - \frac{i\omega g_l^E}{D_l} = 0 . \tag{22}$$

For $\omega \to 0$ the solution for g_l^μ [and thus $\tau_{\mu\mu}$, from Eqs. (17) and (19)] is

$$g_l^\mu(\omega) = g_l^\mu(0) + i\omega \left\{ C_0^\mu + C_1^\mu \int_0^l (D_{l_1} n_{l_1})^{-1} dl_1 - \int_0^l \left[(D_{l_1} n_{l_1})^{-1} \int_{l_1}^\infty l_2 n_{l_2} dl_2 \right] dl_1 \right\} , \tag{23}$$

the expression for g_l^E being similar. The constants C_0^μ and C_0^E yield irrelevant corrections to the critical behavior.[14] In Glauber models[9] neither energy E nor magnetization μ are constants of the motion and $C_1^\mu = 0$, $C_1^E = 0$. If μ is a constant of the motion, $C_1^\mu \neq 0$ and hence $\tau_{\mu\mu} = \infty$, while $C_1^E \neq 0$ and $\tau_{EE} = \infty$ if E is a constant of the motion.[14] While our treatment allows for all these four types of kinetic Ising models,[5] here[16] only the Glauber case will be discussed, deferring a more detailed discussion including the case of nonzero wave vectors to Ref. 14.

Finally, we stress the fact that our basic results [Eqs. (17), (18) and (21)–(23)] do not make use of any specific assumption, neither Eq. (8) nor (12), and hence are far more general than the Fisher droplet model.

III. CRITICAL DYNAMICS AND COMPARISON TO MONTE CARLO CALCULATIONS

Near T_c, the important contribution in Eqs. (17), (18), and (23) comes from large clusters, $l \to \infty$. It is easy to show from Eqs. (8), (11), (21), and (22) the dynamic scaling behavior

$$\chi_{AB}(\omega) = \epsilon^{-\gamma_{AB}} \tilde{\chi}(h\epsilon^{-\beta\delta}, \omega\epsilon^{-\Delta_{AB}}), \tag{24}$$

with

$$\Delta_{\mu\mu} = \Delta_{\mu E} = (2-\gamma)\beta\delta, \tag{25}$$

and if in addition Eq. (20) is invoked

$$\Delta_{EE} = (2-\gamma)\beta\delta. \tag{26}$$

In a naive geometrical interpretation of the droplet model, l is the number of reversed spins in the cluster. Then the two terms in the exponential in Eq. (12) represent the contribution of cluster surface and cluster volume to the cluster free energy. Since one expects the cluster surface to vary in d dimensions as

$$S_l \propto l^{\sigma_G}, \quad \sigma_G = \frac{d-1}{d}, \tag{27}$$

one would expect geometrically $\sigma \equiv 1/\beta\delta$ to be equal to σ_G, implying,

$$\sigma_G = \tfrac{1}{2}, \ \tfrac{2}{3}, \text{ and } \tfrac{3}{4} \text{ for } d = 2, 3, \text{ and } 4, \tag{28}$$

while the correct values are[12]

$$\sigma = \tfrac{8}{15}, \ \tfrac{16}{25}, \text{ and } \tfrac{2}{3}. \tag{29}$$

As expected, the geometric predictions are quite good for $d = 2$, 3 but worse for higher d.

A similar argument can be suggested for γ. Here one expects to find

$$\gamma_G = 1, \tag{30}$$

since all spins of the cluster may be flipped. From $\Delta^G_{\mu\mu} = (2-\gamma_G)/\sigma_G$ and Eqs. (28) and (30) one would get

$$\Delta^G_{\mu\mu} = 2, \ \tfrac{3}{2}, \text{ and } \tfrac{4}{3}, \tag{31}$$

while the correct values are[4,6]

$$\Delta_{\mu\mu} = 2.00 \pm 0.05, \ 1.35 \pm 0.05, \text{ and } 1. \tag{32}$$

Again the geometric prediction becomes worse for higher d.

In an alternative interpretation of the droplet model[17] l is the excess number of reversed spins in a correlated volume V_l,

$$V_l \propto l^{1+1/\delta} \tag{33}$$

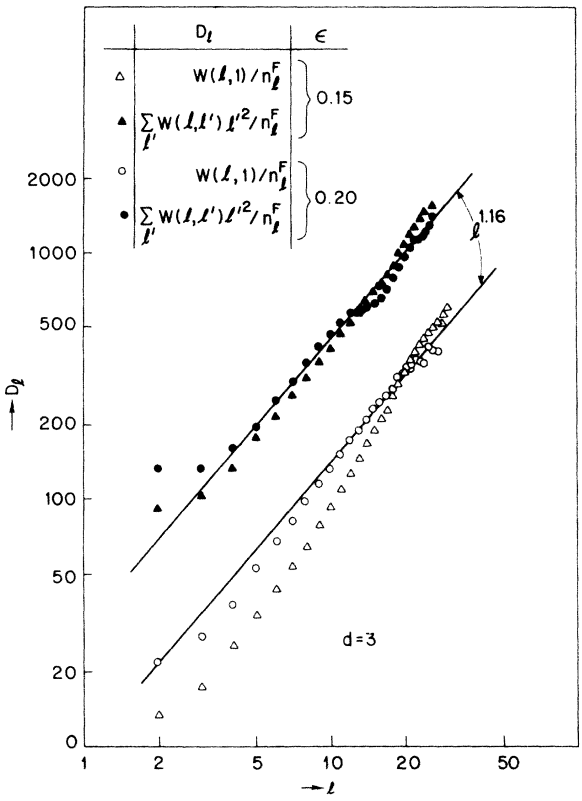

FIG. 1. Growth rate D_l of clusters plotted vs number l of spins, for two values of ϵ, on a sc lattice with $30\times30\times30$ spins, nearest-neighbor interactions, and periodic boundary conditions. The partial growth rates involving only single-spin steps $[W(l,1)]$ are included and show that it would be a poor approximation to neglect higher-order cluster reactions. For the normalizations we did not use the actual cluster distribution n_l, but the Fisher model n_l^F, in order to make the effect of the small-l corrections as small as possible. The data are derived from 10 000 Monte Carlo steps per spin. For $l > 5$, smoothing with a Lorentzian (half-width $\Delta l = 5$) was necessary; and the summation in Eq. (2) was truncated at $l' = 5$ in order to avoid "noise" effects due to finite-time averaging (Refs. 14 and 19).

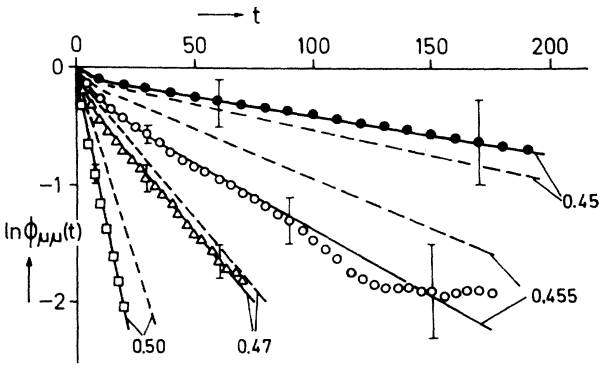

FIG. 2. $\ln \phi_{\mu\mu}(t)$ vs time, in units of "Monte Carlo steps per spin" for a 55×55 square lattice with periodic boundary conditions and nearest-neighbor interactions for some temperatures J/k_BT. Error estimates are indicated for some points. The dashed curves show our cluster model treatment, with b and q_0 fitted to the amplitudes of coexistence curve and critical isotherm, respectively.

which has the surface

$$S_l \propto l^{\sigma+1/\delta} . \tag{34}$$

Geometrically, in this interpretation one would expect the relations to hold:

$$S_l \propto V_l^{(d-1)/d} \tag{35}$$

and

$$D_l \propto V_l . \tag{36}$$

Both relations Eqs. (35) and (36) are found to be correct for $d=4$, but violated for $d<4$. Equation (36) and (33) would yield $\Delta_{\mu\mu}=\gamma$.

Just as the droplet model is unable to fix precisely the static exponents, it cannot fix precisely the dynamic exponent r introduced in Eq. (8). But this approach elucidates the fact that there is no reason for $\Delta_{\mu\mu}$ and γ to be equal. And with D_l given, $\chi_{AB}(\omega)$ can be obtained explicitly.

For numerical calculations, n_l and D_l must be specified. Previous Monte Carlo work[11,18] showed that the Fisher model[12] is a reasonable description of the actual cluster distribution, and that one may then take

$$q(l)=q_0=\text{ const for } l \gtrsim 7 . \tag{37}$$

Here we extend this work to obtain $W(l,l')$ and hence D_l. Figure 1 shows some results on a log-log plot. The slope shown is derived from the renormalization-group result[4] for $\Delta_{\mu\mu}$ via Eq. (26). Figure 1 proves the consistency of our description, and that again it is reasonable to take

$$\tilde{D}(l) \approx D_0 = \text{ const} . \tag{38}$$

If Eqs. (37) and (38) are used down to $l=1$ one may express $\chi_{\mu\mu}(\omega)$ in terms of confluent hypergeometric functions in some cases.[14]

In Fig. 2 some results are shown for $d=2$ and compared to data obtained from Monte Carlo simulations, continuing earlier work.[7,19] Here $\phi_{\mu\mu}(t)$ [Eq. (1)], the Fourier transform of $\mathrm{Im}\chi_{\mu\mu}(\omega)$, is plotted versus time. In order to fix D_0, the initial relaxation time[7] was fitted to the data. Since they are not very close to T_c, we use an "effective" value $\Delta_{\mu\mu}=1.8$ for a simple fit for all times and temperatures shown. The large statistical errors of the Monte Carlo work, increasing critically[19] for $T \to T_c$, make definite statements about the deficiencies of this treatment difficult. While for the temperatures not close to T_c the discrepancies seem to be well outside the error limits, this fact may also be due to correction terms to scaling, since small values of l there still make important contributions, and for the smaller values of l Eqs. (37) and (38) do not hold. It is hoped that extensions of the renormalization-group approach[4] will provide a more sensitive test of the limitations of our numerical results.

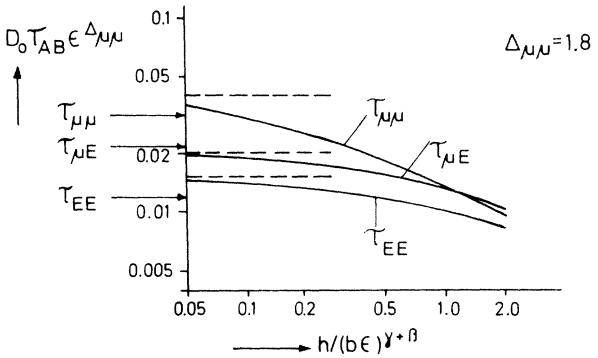

FIG. 3. Scaled relaxation times vs scaled field. The dashed lines indicate the results for $h=0$, while the arrows indicate the corresponding Monte Carlo results.

Figure 3 gives the critical amplitudes as a function of the magnetic field, calculated from the triple integral resulting from Eqs. (17), (18), and (23). The Monte Carlo estimates of Ref. 7 are included; again the initial relaxation time was fitted. While the cluster treatment correctly predicts $\tau_{\mu\mu} > \tau_{\mu E} > \tau_{EE}$, slight discrepancies remain, as it occurs already for static amplitudes.

IV. CONCLUSIONS

First we note that Eq. (25) may also be obtained from a more direct argument with the formal solution of Eq. (1),

$$|P_l(t)\rangle = e^{L_l t}\,|P_l(0)\rangle . \tag{39}$$

Near T_c, the most important eigenvalues λ of L_l are associated with "critical clusters" with $l^\sigma \epsilon \approx 1$, i.e.,

$$l = l_\xi \propto \epsilon^{-1/\sigma} . \tag{40}$$

From Eqs. (6), (8), and (40) one then derives

$$\lambda \propto l_\xi^{r-2} \propto \epsilon^{(2-r)/\sigma} . \tag{41}$$

Another interesting prediction follows in the case where the Fisher model[12] and a simple power law for D_l [Eqs. (8) and (38)] is assumed: The dynamic scaling function depends on only three "amplitudes," D_0, q_0, and b; i.e., one has three scale factor universality, in generalization to two scale factor universality for static critical phenomena.

In summary, a rather explicit theory for the critical dynamics of kinetic Ising models has been obtained, which is general enough to be consistent with recent expansion estimates,[4-6] in contrast to the mode-mode coupling theory.[10] Furthermore, it offers a simple physical picture for the interpretation of the result $\Delta_{\mu\mu} \neq \gamma$ in terms of cluster reactions. As an example, Fishers' cluster model[12] was used to obtain numerical results, which

compare favorably to available Monte Carlo work. Of course, then the dynamics cannot be better than the basic model taken for the static cluster distribution, Eq. (12). We hope to extend[14] our approach to the explicit use of more general cluster models, and also to dynamic critical phenomena in liquid-gas systems, binary alloys, and structural phase transitions.

ACKNOWLEDGMENT

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[1]For a review see, e.g., K. Kawasaki, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, New York, 1972); Vol. II.

[2]B. I. Halperin and P. C. Hohenberg, Phys. Rev. 177, 952 (1969).

[3]R. A. Ferrell *et al.*, Phys. Rev. Lett. 18, 891 (1967).

[4]B. I. Halperin, P. C. Hohenberg, and S.-K. Ma, Phys. Rev. Lett. 29, 1548 (1972).

[5]B. I. Halperin, Proceedings of the 24th Nobel Symposium held at Lerum, Sweden, June 1973 (Academic, New York, to be published).

[6]H. Yahata and M. Suzuki, J. Phys. Soc. Jap. 27, 1421 (1969); H. Yahata, *ibid.* 30, 657 (1971).

[7]E. Stoll, K. Binder, and T. Schneider, Phys. Rev. B 8, 3266 (1973).

[8]T. Schneider, E. Stoll, and K. Binder, Phys. Rev. Lett. 29, 1080 (1972). The doubts raised on the result $\Delta_{\mu\mu}\neq\gamma$ in this paper have been removed by the reanalysis of Ref. 7.

[9]R. J. Glauber, J. Math. Phys. 4, 294 (1963).

[10]K. Kawasaki, Progr. Theor. Phys. 40, 706 (1968); G. Laramore and L. P. Kadanoff, Phys. Rev. 187, 619 (1969); E. Riedel and F. Wegner, Phys. Lett. A 32, 273 (1970).

[11]K. Binder and H. Müller-Krumbhaar, Phys. Rev. B 9, 2328 (1974).

[12]M. E. Fisher, Physics 3, 255 (1967).

[13]K. Binder and E. Stoll, Phys. Rev. Lett. 31, 47 (1973).

[14]K. Binder, D. Stauffer, and H. Müller-Krumbhaar, (unpublished).

[15]Note that $\tau_{\mu E}$ and τ_{EE} are defined in complete analogy to $\tau_{\mu\mu}$.

[16]Equations (5)–(7) provide immediately the result for the energy relaxation of the spin-exchange model (Ref. 1), where μ = const. But note the differences in the critical behavior: While spin flips in the interior of the cluster provide a cluster reaction, spin-exchange processes in the interior do not. Hence the values of γ in Eq. (2) are different. While D_l remains finite for $T\to T_c$ in the Glauber case, it diverges in other cases (Ref. 14).

[17]D. Stauffer, C. S. Kiang, and G. H. Walker, J. Stat. Phys. 3, 323 (1971); K. Binder and D. Stauffer, *ibid.* 6, 49 (1972).

[18]E. Stoll, K. Binder, and T. Schneider, Phys. Rev. B 6, 2777 (1972); H. Müller-Krumbhaar (unpublished).

[19]H. Müller-Krumbhaar and K. Binder, J. Stat. Phys. 8, 1 (1973).