Theory for the dynamics of clusters near the critical point. I. Relaxation of the Glauber kinetic Ising model

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A semiphenomenological cluster theory is developed for dynamic critical properties, which is not limited to small deviations from equilibrium. Explicit numerical expressions are derived for linear and nonlinear response functions of the kinetic Ising model, which are compatible with dynamic scaling and recent calculations of dynamic critical exponents. An uniaxial magnet (or Ising system) and its critical fluctuations are approximated by a "droplet" or "cluster" model, the critical part of the free energy being the sum of the contributions of clusters each containing l spins. These clusters are assumed to grow and shrink with a phenomenological rate $\propto l^r$, $r \approx 1$. These rates are compatible with direct Monte Carlo simulations in two and three dimensions. The resulting cluster reaction and diffusion equation is an approximation to the master equation of the Glauber kinetic Ising model (in which the magnetization is not conserved), and has the same structure as used in nucleation theories. In linear response to space- and time-dependent fields the relaxation times of energy and magnetization are expressed as triple integrals. All relaxation times diverge as $|1 - T/T_c|^{-(2-r)\beta\delta}$. This treatment is consistent with dynamic scaling and universality, and is more general than the conventional (Van Hove) theory of critical slowing down. Also the (smaller) exponents found for the response to localized variations ("autocorrelation functions") agree with dynamic scaling. The wave-vector dependence of the relaxation times arises from the static correlation function only, and not from the cluster diffusion term. If the equilibrium cluster distribution is assumed to be that of the Fisher droplet model, and for special values of r, the (discrete) eigenvalue spectrum and eigenfunctions (generalized Laguerre polynomials) of the cluster reaction equation are found explicitly. Relaxation functions and frequency-dependent susceptibilities are expressed by hypergeometric series, even in the case of nonlinear response. At the critical point, the spectrum becomes continuous and the exponential decay for large times is replaced by power-law behavior. Similarly, it is found that dynamic scaling applies also to the nucleation rate for this model, and the scaling exponent of the nucleation rate is $2 - \alpha + (2 - r)\beta\delta$.

I. INTRODUCTION

Studying dynamic critical phenomena it is important to establish relations between singular transport coefficients (or relaxation times) and the critical singularities of static properties.[1] While mean-field theories and related "conventional theories of critical slowing down"[2] are not very satisfactory near the critical point, more successful treatments have been based on hydrodynamic approaches and scaling arguments,[3–5] on mode-mode coupling approximations,[6,7] and renormalization-group expansions.[8–10] Although predictions are rich in detail, our understanding of dynamic critical phenomena is still somewhat incomplete, and hence the application of additional methods seems desirable.

Such a method is described in the present paper, where we interpret dynamic critical fluctuations in terms of a "cluster dynamics."[11] Near T_c the dynamics are dominated by long-range fluctuations, which may alternatively be described by large "clusters." It will be pointed out that these large clusters may be characterized by one coordinate only, while the contributions of small clusters can essentially be neglected, so that an important reduction of the mathematical complexity is obtained. Although the uncertainty of both what is meant precisely by a "cluster," and what the static properties of clusters are, is a serious drawback of this attempt, this approach may also have some important advantages: (i) Similar concepts are already used to describe relaxation phenomena far from thermal equilibrium, e.g., by nucleation theory.[12] Thus the cluster-reaction theory is a description, where both the critical and the nonequilibrium relaxation are special cases of a more general approach. In many cases the theory of nonequilibrium relaxation is still in a rather crude stage, and thus this general treatment may help to clarify some of the open problems. (ii) In some cases computer experiments on critical phenomena[13] have yielded "raw data," where an immediate intuitive interpretation in terms of "clusters" is possible.[14–16] Such qualitative observations may lead to questionable conclusions if they

are not substantiated by a more quantitative description for the cluster dynamics, as first tried in Ref. 15. (iii) In a few cases, a simple qualitative understanding for the singularities of the kinetic coefficients is lacking, like the Glauber model discussed here, and hence the cluster dynamics may help to elucidate these problems. In addition, it may also provide a more convenient technique for calculating dynamic correlation functions, etc.

In the present paper we show that the cluster dynamics indeed permits some progress on all these counts, treating a rather simple case only, the Glauber kinetic Ising model.[17] Other systems will be treated elsewhere.[18] The Ising model may describe reasonably the static aspects of critical phenomena[19] both for anisotropic magnets, liquid-gas systems, binary alloys, and perhaps even biochemical objects such as membranes,[20] chain molecules,[21] etc. But the dynamics of the kinetic Ising model is at best a very crude approximation to the actual dynamics of the above systems. Therefore it is important to note that the phenomenological cluster-reaction theory may be far more general than the kinetic Ising model, although we shall only consider this model explicitly.

In spite of its unrealistic character, the Glauber kinetic Ising model found great attention,[1,8,9,11,13-15,22-30] due to several reasons: (i) Methods can be applied which are not available for most other dynamic problems, as high-temperature-series expansions,[23] exact inequalities,[26] and Monte Carlo calculations[27]; (ii) while the mode-mode coupling work suggests[7] that the exponent $\Delta_{\mu\mu}$ of the order-parameter relaxation time is equal to the exponent γ of the susceptibility, i.e., that the conventional theory[2] should be valid in this case, the high-temperature-series work,[23] the renormalization-group approach,[8] and some of the Monte Carlo calculations[27] showed that, in fact, $\Delta_{\mu\mu} > \gamma$, for dimensionality $d < 4$. Both the fact that mode-mode theory does not hold and that a great variety of results are available make a test of the cluster-reaction theory very interesting.

The outline of this paper is as follows. In Sec. II, we briefly define the kinetic Ising model and summarize the most relevant critical properties.[31] We also discuss the derivation of the cluster-reaction theory from the master equation.[15] In Sec. III, we summarize the static properties of clusters. Section IV contains the calculation of the relaxation times according to our model. Section V shows how explicit results for the eigenvalue spectrum of the Liouville operator and the dynamic scaling functions are obtained, if an explicit formula for the static cluster concentration n_l such as the Fisher droplet model[32] is used. Section VI then contains the extension to the case of nonlinear response and nucleation, while Sec. VII summarizes our conclusions.

II. KINETIC ISING MODEL AND DERIVATION OF CLUSTER-REACTION THEORY

A. Master equation and relaxation functions

We consider a system of N Ising spins ($\mu_i = \pm 1$) on a lattice,

$$\mathcal{H} = -\sum_{i \neq j} J_{ij} \mu_i \mu_j - \mu_B H \sum_i \mu_i \,, \tag{2.1}$$

denoting the exchange constants by J_{ij} and the magnetic field by H, μ_B being the magnetic moment of a spin, and assume that these spins are coupled to some "heat bath" (e.g., lattice vibrations) which produces random spin flips. Following Glauber, it is assumed that only one spin (e.g., μ_j) is flipped at once, according to a transition probability $W(\mu_1, \ldots, \mu_j, \ldots, \mu_N)$ which does not depend explicitly on time. Then the dynamics is described by the master equation for the probability $P(\mu_1, \ldots, \mu_j, \ldots, \mu_N, t)$ that the system is found in a state $\{\mu_1, \ldots, \mu_j, \ldots, \mu_N\}$ at time t,[17,22]

$$\begin{aligned} \frac{d}{dt} P(\mu_1, \ldots, \mu_j, \ldots, \mu_N, t) &= -\sum_j W(\mu_j \to -\mu_j) P(\mu_1, \ldots, \mu_j, \ldots, \mu_N, t) \\ &\quad + \sum_j W(-\mu_j \to \mu_j) P(\mu_1, \ldots, -\mu_j, \ldots, \mu_N, t) \\ &\equiv -L_\mu P(\mu_1, \ldots, \mu_j, \ldots, \mu_N, t) \,. \end{aligned} \tag{2.2}$$

This single spin-flip model does not have any conserved quantities, in particular neither energy nor magnetization are constants of the motion. Now time-dependent averages are defined by

$$\langle A(t) \rangle \equiv \sum_{\{\mu_i\}} A(\{\mu_i\}) P(\{\mu_i\}, t) \,. \tag{2.3}$$

The thermal-equilibrium state is a stationary solution of Eq. (2.2) due to the detailed balance condition

$$\begin{aligned} &W(\mu_j \to -\mu_j) P_0(\mu_1, \ldots, \mu_j, \ldots, \mu_N) \\ &\quad = W(-\mu_j \to \mu_j) P_0(\mu_1, \ldots, -\mu_j, \ldots, \mu_N) \,, \end{aligned} \tag{2.3a}$$

where $P_0(\mu_1, \ldots, \mu_j, \ldots, \mu_N)$ is the distribution

$$\begin{aligned} P_0(\{\mu_i\}) &= (1/Z)\, e^{-(1/k_B T)\mathcal{H}} \,, \\ Z &= \mathrm{Tr}\, e^{-(1/k_B T)\mathcal{H}} \,. \end{aligned} \tag{2.3b}$$

As an example we mention the ansatz for W suggested by Suzuki and Kubo,[24]

$$W(\mu_j \to -\mu_j) = (1/2\tau_s)[1 - \tanh(\delta\mathcal{H}_j / 2k_B T)] \,, \tag{2.4}$$

where $\delta\mathcal{H}_j$ is the change in energy associated with the slip of the jth spin [to be computed according to Eq. (2.1)], and τ_s is the relaxation time of a single spin interacting with the heat bath. This

arbitrary parameter fixes our time scale. We will not make any specific use of this arbitrary form for W [Eq. (2.4)], however.

The slowing down of our model is now conveniently described in terms of relaxation functions. Considering thermal fluctuations in our equilibrium state, these functions are defined by

$$\Phi_{B,C}(\vec{q}, t) = S_{BC}(\vec{q}, t)/S_{BC}(\vec{q}, 0) , \tag{2.5a}$$

$$S_{B,C}(\vec{q}, t) = \sum_j e^{i\vec{q}\cdot\vec{x}_j}[\langle B(0,0)C(\vec{x}_j, t)\rangle - \langle B\rangle\langle C\rangle] , \tag{2.5b}$$

where B, C can be the magnetization μ_j, or energy

$$E_j = \sum_{i(\neq j)} J_{ij}\mu_i\mu_j ,$$

etc. It is also interesting to consider the slowing down of these local quantities, and describe it in terms of autocorrelation functions

$$\Phi^A_{B,C}(t) = \frac{\langle B(0,0)\,C(0,t)\rangle - \langle B\rangle\langle C\rangle}{\langle B(0,0)\,C(0,0)\rangle - \langle B\rangle\langle C\rangle} , \tag{2.6a}$$

which can be expressed also in terms of Eq. (2.5), of course, i.e.,

$$\Phi^A_{BC}(t) = \sum_q S_{BC}(\vec{q}, t)\Big/\sum_q S_{BC}(\vec{q}, 0) . \tag{2.6b}$$

As an alternative to this description in terms of time-dependent correlation functions, one can also consider the response of the system to a time-dependent ($\propto e^{i\omega t}$) infinitesimal change of an external parameter e conjugate to C and calculate the susceptibility $\chi_{BC}(\vec{q}, \omega)$ in linear response

$$\langle B(t)\rangle = \langle B\rangle + \chi_{BC}(\vec{q}, \omega)\delta e\, e^{i(\vec{q}\cdot\vec{x} + \omega t)} . \tag{2.7}$$

Of course, these descriptions are related by the fluctuation-dissipation theorem[24]

$$\chi_{BC}(\vec{q}, \omega) = \chi_{BC}(\vec{q}, 0) - \frac{i\omega}{k_B T}\int_0^\infty S_{BC}(\vec{q}, t)\, e^{-i\omega t}\, dt , \tag{2.8a}$$

while for the static susceptibility we have

$$\chi_{BC}(\vec{q}, 0) = (1/k_B T)\, S_{BC}(\vec{q}, 0) . \tag{2.8b}$$

Being interested also in situations far from thermal equilibrium, we consider the (nonlinear) response to finite changes Δe of an external parameter $e(t)$. Assuming that the system is in equilibrium for $t < 0$ [where $e(t) = e + \Delta e\, e^{i\vec{q}\cdot\vec{x}}$], we describe the relaxation for $t > 0$ [where $e(t) = e$] by a nonequilibrium relaxation function[29]

$$\Phi^{\Delta e}_B(\vec{q}, t) = \frac{\langle B_q(t)\rangle - \langle B_q(\infty)\rangle}{\langle B_q(0)\rangle - \langle B_q(\infty)\rangle} , \tag{2.9}$$

defining $B_q(t) \equiv \sum e^{i\vec{q}\cdot\vec{x}} B(\vec{x}, t)$. This relaxation function reduces to Eq. (2.5) for very small Δe.[29]

Equations (2.5) and (2.8) suggest that a reasonable definition for a relaxation time $\tau_{BC}(\vec{q})$ of the system is given in terms of the small-frequency behavior of the dynamic susceptibility,

$$\chi_{BC}(\vec{q}, \omega) = \chi_{BC}(\vec{q})[1 - i\omega\tau_{BC}(\vec{q}) + O(\omega^2)] , \tag{2.10a}$$

where

$$\tau_{BC}(\vec{q}) = \int_0^\infty dt\, \Phi_{BC}(\vec{q}, t) . \tag{2.10b}$$

Similarly, Eqs. (2.8) and (2.10) can be generalized to the response to local fields

$$\chi^A_{BC}(\omega) = \chi^A_{BC}[1 - i\omega\tau^A_{BC} + O(\omega^2)] , \tag{2.11a}$$

$$\tau^A_{BC} = \int_0^\infty dt\, \Phi^A_{BC}(t) , \tag{2.11b}$$

and finally one defines a nonequilibrium relaxation time by

$$\tau^{\Delta e}_B(\vec{q}) = \int_0^\infty dt\, \Phi^{\Delta e}_B(\vec{q}, t) . \tag{2.12}$$

Of course, infinitely many other relaxation times can be defined[25] and investigated,[28] but this seems to us of little physical interest here.

B. Critical slowing down

A calculation of these relaxation times and functions from Eqs. (2.1)–(2.4) is easy in the framework of the mean-field approximation, which yields[15,24]

$$\chi_{\mu\mu}(\vec{q}, \omega) = \frac{\chi_{\mu\mu}(\vec{q})}{1 + i\omega\tau_{\mu\mu}(\vec{q})} ,$$
$$\tau_{\mu\mu}(\vec{q}) = \frac{\tau_s k_B T \chi_{\mu\mu}(\vec{q})}{1 - \langle\mu\rangle^2} . \tag{2.13}$$

Equation (2.13) implies that μ_q is an exact eigenvector of the "Liouville operator" L_μ [Eq. (2.2)],

$$L_\mu\mu_q(t) = \lambda_q\mu_q(t) , \quad \lambda_q = \tau^{-1}_{\mu\mu}(\vec{q}) , \quad \Delta_{\mu\mu} = \gamma , \tag{2.14}$$

while the (generalized)[33] time-dependent Ginzburg-Landau theory requires Eq. (2.13) only for $\omega \to 0$, or sufficiently large times. Abe and Hatano[26] investigated the validity of Eq. (2.14) by moment calculations, which agree with Eq. (2.14) for the first moment only,

$$\frac{d}{dt}\Phi_{BC}(\vec{q}, t)\Big|_{t=0} \equiv \frac{1}{\tau^I_{BC}(\vec{q})} = \frac{\text{const}}{\chi_{BC}(\vec{q})} , \tag{2.15}$$

but disagree for higher moments. These moment calculations thus show that Eq. (2.14) is invalid in the general case, although it holds if either the dimensionality or the range of the interaction are infinite. Of course, this argument does not rule out that the conventional theory[2] [i.e., Eq. (2.13) for $\omega \to 0$] holds; in fact, one expects it to become valid for $d \geq 4$.[8]

The singularities near T_c are described in terms of the critical exponents ($H = 0$),

$$\chi_{BC}(0)\underset{\epsilon\to 0}{\longrightarrow}\tilde{\chi}_{BC}\,|1-T/T_c|^{-\gamma_{BC}}\equiv\tilde{\chi}_{BC}\,|\epsilon|^{-\gamma_{BC}}\,, \tag{2.16a}$$

$$\tau_{BC}(0)\underset{\epsilon\to 0}{\longrightarrow}\tilde{\tau}_{BC}\,|\epsilon|^{-\Delta_{BC}}\,,$$

$$\tau_B^{\Delta e}(0)\underset{\epsilon\to 0}{\longrightarrow}\tilde{\tau}_B^{\Delta e}\,|\epsilon|^{-\Delta_B^{\Delta e}}\,,$$

$$\tau_{BC}^{A}\underset{\epsilon\to 0}{\longrightarrow}\tilde{\tau}_{BC}^{A}\,|\epsilon|^{-\Delta_{BC}^{A}}\,, \tag{2.16b}$$

with constant prefactors $\tilde{\tau}$. Since in the Glauber model L_μ is a Hermitian operator, it follows that[26]

$$\Delta_{BC}\geq\gamma_{BC}\,, \tag{2.17}$$

while Eq. (2.15) shows that the initial relaxation time τ_{BC}^{I} always diverges with the same exponent as the associated static susceptibility.

The dynamic scaling hypothesis implies that near T_c,[3,4]

$$\chi_{BC}(\vec{q},\omega)=|\epsilon|^{-\gamma_{BC}}\tilde{\chi}_{BC}(\vec{q}\,|\epsilon|^{-\nu},\omega\,|\epsilon|^{-\Delta_{BC}},H|\epsilon|^{-\beta\delta})$$
$$+|\epsilon|^{-\gamma_{BC}+x_1}\tilde{\chi}_{BC}^{1}+\cdots\,, \tag{2.18}$$

where β and δ are the order-parameter exponents

$$\langle\mu\rangle\underset{\epsilon\to 0,H=0}{\longrightarrow}B|\epsilon|^{\beta}\,,$$
$$\langle\mu\rangle\underset{H\to 0,\epsilon=0}{\longrightarrow}DH^{1/\delta}\,, \tag{2.19a}$$

and ν is the exponent of the correlation length

$$\xi\to\tilde{\xi}\,|\epsilon|^{-\nu}\,. \tag{2.19b}$$

Since the dynamic scaling hypothesis is based on the physical idea that there is only one characteristic frequency important near T_c,[3,4] one expects the various exponents Δ_{BC} to be equal.[34] Furthermore, it is easy to relate the exponents Δ_{BC} and the associate Δ_{BC}^{A} by use of Eq. (2.18); if the denominator of Eq. (2.6a) remains nonzero at T_c, this relation is[35]

$$\Delta_{BC}-\Delta_{BC}^{A}=d\nu-\gamma_{BC}\,. \tag{2.20}$$

Finally, one expects that the scaling function $\tilde{\chi}$ in Eq. (2.18) is universal, i.e., apart from some scale factors, $\tilde{\chi}$ should depend only on the dimensionality of the system, but not on irrelevant details such as lattice structure, spin quantum number, precise choice of W in Eq. (2.2), nor on the precise behavior of the exchange integral J_{ij} as a function of distance $\vec{x}_i-\vec{x}_j$, etc. It should also make no difference if more than one spin is flipped at a time, as long as there are no constants of the motion.

It will turn out that all these conjectures are supported by the cluster dynamics treatment, to be discussed below.

C. Derivation of cluster dynamics

In this subsection we obtain the main starting point of our treatment, the cluster reaction and diffusion equation [Eq. (2.33)]. The main approximation made is to characterize each cluster by one coordinate only (denoted as l), since we argue that for very large clusters, i.e., $l\to\infty$, other coordinates become irrelevant parameters. We now derive Eq. (2.33) from the master equation (2.3) by a sequence of approximations, which we attempt to justify in detail.

Clearly, given a suitable prescription for deciding which cluster any given spin belongs to, one can give an exact description of our system in terms of clusters. The usefulness of this description, however, lies in the approximation of considering only one coordinate, since then a strongly correlated system can be described as being composed of essentially noninteracting clusters. In this sense our theory is analogous to Landau's theory of quantum liquids, in which the system is composed of quasiparticles whose interactions have a particularly simple form.

In order to give a specific example, we mention the definition of clusters in terms of the set of contours, that is the set of nonintersecting polygons (in two dimensions; in three dimensions: polyhedra) separating opposite spins[36] [Fig. 1(a)]; we interpret each contour as a cluster surface. Each spin flip then produces cluster reactions [Fig. 1(b)]. Obviously, it is then convenient to choose as "coordinates" describing a cluster the number l of reversed spins which it contains, the surface area of its contour, etc. Another important cluster coordinate is, for instance, its "center of gravity" $\vec{x}$,

$$\vec{x}=\frac{\sum_i\vec{x}_i m_i}{\sum_i m_i}=\frac{1}{l}\sum_i\vec{x}_i\,, \tag{2.21}$$

where we have associated a mass $m_i=1$ with each reversed spin, and the sums include all spins of the considered cluster.

Of course this "contour picture" is only an example, and it is, in fact, inappropriate for our pur-

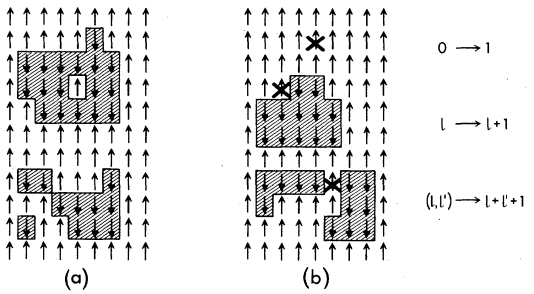

FIG. 1. (a) Small parts of an arbitrary two-dimensional Ising spin configuration. Contours form the boundaries of clusters (dashed regions) of l reversed spins ($l=1$, 2, 8, 17 in this example). In two dimensions, clusters can touch at most at isolated points. (b) Flip of an up spin (denoted by a cross) can, e.g., either create a $l=1$ cluster, or produce a $l\to l+1$ or $(l,l')\to l+l'+1$ reaction. Reactions with more than two clusters are rather unimportant and therefore not shown.

pose (see Sec. III). Therefore we will in the following describe each cluster by a set of coordinates $\{l, \alpha\}$, where l is the excess number of reversed spins within the cluster, and α symbolically accounts for the other coordinates.[37] Similar to Ref. 15, we consider now the average cluster concentration $\bar{n}_l(t) = \langle n_l^\alpha(t) \rangle_\alpha$ in the system, where the various cluster configurations described by different $\{\alpha\}$ are summed over and weighted with the appropriate Boltzmann factors. Equation (2.2) then implies

$$\frac{d\bar{n}_l}{dt} = \sum_{l'=0}^{\infty} \langle n_{l+l'+1}^{\alpha} (t)\, S_{l+l'+1,l'}^{\alpha;\alpha'}(t) \rangle_{\alpha,\alpha'} - \sum_{l'} \langle n_l^\alpha(t)\, S_{l,l'}^{\alpha;\alpha'}(t) \rangle_{\alpha,\alpha'}$$
$$+ \sum_{l'} \langle n_{l-l'-1}^{\alpha}(t) n_{l'}^{\alpha'}(t)\, C_{l-l'-1,l'}^{\alpha;\alpha'}(t) \rangle_{\alpha,\alpha'} - \sum_{l'} \langle n_l^\alpha(t) n_{l'}^{\alpha'}(t) C_{l,l'}^{\alpha;\alpha'}(t) \rangle_{\alpha,\alpha'} + \cdots \,. \tag{2.22}$$

Here $S_{l,l'}^{\alpha;\alpha'}(t)$ denotes the rate at which a cluster $\{l', \alpha'\}$ is splitting off from a cluster $\{l, \alpha\}$; the case $l' = 0$ then describes ordinary "shrinking" of the l cluster [reverse process to Fig. 1(b)]. $C_{l,l'}^{\alpha;\alpha'}(t)$ describes the rate at which clusters $\{l, \alpha\}$ and $\{l', \alpha'\}$ coalesce; the case $l' = 0$ describes ordinary growing of the l cluster [Fig. 1(b)]. For simplicity the reactions involving more than two clusters at the same time have not been written down in Eq. (2.22), but otherwise Eq. (2.22) is exact. In principle, one can determine the $C_{l,l'}^{\alpha;\alpha'}$ and $S_{l,l'}^{\alpha;\alpha'}$ from the master equation [Eqs. (2.2) and Eq. (2.4)].

We will see later (Secs. III and IV) that the divergence of susceptibilities and relaxation times is due only to the contributions of very large clusters. Therefore it is appropriate for our purpose to consider only the behavior of $d\bar{n}_l/dt$ for $l \to \infty$ and we may also use a coarse grained time scale.

From general thermodynamic fluctuation theory it is clear that in this limit the probability distribution for n_l^α at fixed large l will be very sharply peaked at the mean values $\{\bar{\alpha}(l)\}$; therefore we shall now make a factorization approximation

$$\langle n_l^\alpha(t) S_{l,l'}^{\alpha;\alpha'}(t) \rangle_{\alpha,\alpha'} \cong \langle n_l^\alpha(t) \rangle_\alpha \langle S_{l,l'}^{\bar{\alpha}(l),\alpha'}(t) \rangle_{\alpha'}$$
$$\equiv \bar{n}_l(t) \bar{S}_{l,l'}(t) \,, \tag{2.23a}$$
$$\langle n_l^\alpha(t) n_{l'}^{\alpha'}(t)\, C_{l,l'}^{\alpha;\alpha'}(t) \rangle_{\alpha,\alpha'} \approx \langle n_l^\alpha(t) \rangle_\alpha \langle n_{l'}^{\alpha'} C_{l,l'}^{\bar{\alpha}(l),\alpha'}(t') \rangle_{\alpha'}$$
$$\equiv \bar{n}_l(t) \bar{C}_{l,l'}(t) \,. \tag{2.23b}$$

Since we wish to use these relations from $l = \infty$ down to $l \cong l_t$ (see Sec. III) where the relative fluctuation in other coordinates (e.g., the cluster surface area) is no longer negligible, we have also to perform time averaging over some interval Δt. In order that the error made by the approximation Eq. (2.23) be negligibly small, we must choose $\Delta t \gg 1$, which is legitimate near T_c where the time scales of interest diverge. Note that the cluster concentrations decrease rapidly with increasing l' (see Sec. III); therefore the l' in Eqs. (2.22) and (2.23) are typically much smaller than l and a further factorization[15] of $\bar{C}_{l,l'}(t)$ defined in Eq. (2.23b) may be inaccurate and is avoided here. Hence we get

$$\frac{d\bar{n}_l}{dt} = \sum_{l'} \bar{n}_{l+l'+1}(t) \bar{S}_{l+l'+1,l'}(t) - \sum_{l'} \bar{n}_l(t) \bar{S}_{l,l'}(t)$$
$$+ \sum_{l'} \bar{n}_{l-l'-1}(t) \bar{C}_{l-l'-1,l'}(t) - \sum_{l'} \bar{n}_l(t) \bar{C}_{l,l'}(t) \,. \tag{2.24}$$

We now denote the cluster concentrations of the final equilibrium state by $\{n_l^f\}$, and the equilibrium values of $\bar{S}_{l,l'}(t)$ and $\bar{C}_{l,l'}(t)$ by $S_{l,l'}^f$ and $C_{l,l'}^f$. Because of the detailed balance condition [Eq. (2.3)] we have also

$$n_{l+l'+1}^f S_{l+l'+1,l'}^f = n_l^f C_{l,l'}^f \equiv W(l, l') \,. \tag{2.25}$$

Introducing

$$g_l(t) = [\bar{n}_l(t) - n_l^f]/n_l^f \,,$$
$$\Delta S_{l,l'}(t) = [\bar{S}_{l,l'}(t) - S_{l,l'}^f]/S_{l,l'}^f \,,$$

we then obtain in linear response

$$n_l^f \frac{d}{dt} g_l(t) = \sum_{l'} W(l, l')[g_{l+l'+1}(t) - g_l(t)] - \sum_{l'} W(l - l' - 1, l')[g_l(t) - g_{l-l'-1}(t)]$$
$$+ \sum_{l'} W(l - l')[\Delta S_{l+l'+1,l'}(t) - \Delta C_{l,l'}(t)] + \sum_{l'} W(l - l' - 1, l')[\Delta C_{l-l'-1,l'}(t) - \Delta S_{l,l'}(t)] \,. \tag{2.26}$$

Since our treatment refers to large clusters $l \gg l'$, we expand in $(l' + 1)$,

$$g_{l-l'-1}(t) = g_l(t) - (l' + 1) \frac{\partial}{\partial l} g_l(t) + \tfrac{1}{2}(l' + 1)^2 \frac{\partial^2}{\partial l^2} g_l(t) + \cdots \,, \tag{2.27}$$

to get

$$n_l^f \frac{\partial}{\partial t} g_l(t) = \left(\sum_{l'} W(l, l')\,(l'+1)^2\right) \frac{\partial^2}{\partial l^2} g_l(t) + \frac{\partial}{\partial l}\left(\sum_{l'} W(l, l')\,(l'+1)^2\right) \frac{\partial}{\partial l} g_l(t)$$
$$+ \sum_{l'} (l'+1) \frac{\partial}{\partial l} \{W(l, l')[\Delta S_{l,l'}(t) - \Delta C_{l,l'}(t)]\} . \qquad (2.28)$$

From Eqs. (2.23b) and (2.25) we see that for $l, l' \to \infty$ we should have $W(l, l') \propto n_l^f n_{l'}^f$. Thus $W(l, l')$ decreases rapidly as a function of l' (see Sec. III). On the other hand, near T_c we will only be interested in solutions which satisfy the boundary condition (see Secs. V and III)

$$g_{l=0}(t) = 0 \quad \text{or} \quad g_l(t) \approx 0 ;$$
$$l \ll l_\xi \begin{cases} l_\xi \propto |\epsilon|^{-1/\beta\delta}, & H = 0 \\ l_\xi \propto |H|^{-1}, & \epsilon = 0 . \end{cases} \qquad (2.29)$$

Since the order of magnitude of $\Delta S_{l,l'}(t)$ and $\Delta C_{l,l'}(t)$ must be the same as $g_{l'}(t)$ [see Eqs. (2.23b) and (2.25)], the last term in Eq. (2.28) is also negligible; it yields only irrelevant corrections to the critical behavior.[38] Defining a cluster reaction rate R_l by[11,15]

$$R_l = \frac{1}{n_l^f} \sum_{l'=0}^{\infty} W(l, l')\,(l'+1)^2 , \qquad (2.30)$$

Eq. (2.28) reduces to an equation well known from nucleation theory,[12]

$$\frac{\partial}{\partial t} g_l(t) = R_l\left(\frac{\partial^2}{\partial l^2} g_l(t) + \frac{\partial}{\partial l}(\ln R_l n_l^f) \frac{\partial}{\partial l} g_l(t)\right) . \qquad (2.31)$$

Allowing for reactions where more than two clusters are involved at the same time changes only the precise definition of R_l but not Eq. (2.31). Therefore Eq. (2.31) should have a rather general validity. It is also important to note that we have made little use of the specific dynamics of the kinetic Ising model [e.g., Eq. (2.4)]. Therefore we expect Eq. (2.31) to be a valid phenomenological description for a much wider class of physical systems.

Since we are interested also in the response to "fields" varying in space $\vec{x}$, we must consider also a space-dependent cluster concentration $\bar{n}_l(\vec{x}, t)$ and its relative deviation from equilibrium n_l^f (where no $\vec{x}$ dependence exists owing to translational symmetry). Note that each spin flip also shifts the center of gravity $\vec{x}$ [Eq. (2.21)] of each cluster involved in this reaction. These shifts are random in orientation and the amount of a single shift is a/l,[39] where a is the nearest-neighbor distance. These random walks of the center of gravity give rise to a cluster diffusion constant[40]

$$D_l = R_l (a/l)^2 c_1 , \qquad (2.32)$$

where c_1 is a constant of order unity which depends on the amount of coarse graining of our time scale. Thus the generalization of Eq. (2.31) is found to be

$$\frac{\partial}{\partial t} g_l(\vec{x}, t) = R_l\left(\frac{\partial^2}{\partial l^2} g_l(\vec{x}, t) + \frac{\partial}{\partial l}(\ln R_l n_l^f) \frac{\partial}{\partial l} g_l(\vec{x}, t)\right)$$
$$+ D_l \nabla^2 g_l(\vec{x}, t) . \qquad (2.33)$$

This cluster reaction and diffusion equation is the basic starting point of our calculations. It involves two functions, the cluster concentration in equilibrium n_l^f and the cluster reaction rate R_l which are, in general, unknown. In the next parts of our paper we will discuss the properties of these two functions. If they are assumed to be known, then the critical dynamics can be derived from Eq. (2.33) by mathematical manipulations which are in principle straightforward, as will become evident in Secs. IV–VI.

D. Cluster reaction rate

If we were to use the "contour picture" to define clusters (Fig. 1) without any coarse graining of the time scale, R_l would be essentially proportional to the number of reactions per unit time in which a cluster with l spins is involved, according to Eq. (2.30). Since each of the l spins of such a cluster has a nonzero probability to flip and hence react, we would expect

$$R_l \propto l , \quad l \to \infty . \qquad (2.34)$$

Of course there is no reason to assume that this relation remains strictly true if we use a more general definition for the clusters (Sec. III), where the volume V_l of an l cluster is no longer proportional to l. But owing to the general similarity of the various possible cluster definitions, we assume[41]

$$R_l = \tilde{R}(l) l^r , \quad 0 < \tilde{R}(\infty) < \infty , \quad l \to \infty , \qquad (2.35)$$

where r is a general exponent. In Eq. (2.35), we do not *a priori* exclude that the amplitude $\tilde{R}(\infty)$ vanishes or diverges right at the critical point.

If we did not perform any coarse graining of our time scale, we would expect a relation $R_l \propto V_l$, since each of the V_l spins has a nonzero probability to flip and hence react, and each spin flips on the average once per unit time. We are not allowed to assume $R_l \propto V_l \Delta t$ in the case of coarse graining, however, since the Δt configurations over which the coarse grainings is performed are highly correlated. This will affect the rate R_l at which an l cluster grows or shrinks (in fact, a significant number of spin flips will rather contribute to average out the significant fluctuations in surface area, etc., mentioned above and thus does not contribute to a change in l). Owing to these "memory effects" we are unable to obtain the value of r explicitly or relate it

to any static properties of clusters. Since it will turn out that the exponent r is related to dynamic critical exponents, we are prevented from estimating these exponents reliably. We can only estimate their rough order of magnitude [using $r \approx 1$, Eq. (2.34), or $r \approx 1 + 1/\delta$, Eq. (3.5)]. Of course, the same difficulty arises in the description of static cluster properties with respect to static critical exponents: e.g., the rough estimate for $\beta\delta$ would be $d/(d-1)$, where d is the dimensionality.[32]

It is also interesting to investigate the range of validity of the asymptotic description (2.35). By Monte Carlo computer experiments[13] it is possible to obtain information on the behavior of R_l also for small l and not too close to T_c. For practical reasons,[13] such computations can be carried out only when $\langle \mu \rangle$ is still rather large. Then the fluctuations of $\langle \mu \rangle$ consist of few well-separated clusters of reversed spins, and the contour description of clusters can be used conveniently. In Fig. 2 we present some results for a 110×110 square Ising lattice with nearest-neighbor interactions and periodic boundary conditions. In this special case Eq. (2.4) was used, and no coarse graining of time was performed. $W(l, l')$ was obtained for $l, l' < 30$ and $\epsilon = 1 - T/T_c \cong 0.04$ and 0.02. It is seen that a power-law behavior with an exponent r close to 1 [Eq. (2.34)] is, in fact, a reasonable description for R_l even in the small-l range. Correspondingly, the critical behavior of the relaxation functions obtained here should be visible in a rather broad region around T_c. This prediction is indeed supported by some of the numerical calculations.[23,27] Similar results have been obtained for three dimensions previously.[11] The numerical results also suggest that $\tilde{R}(\infty)$ does not depend on ϵ in a critical manner.[11]

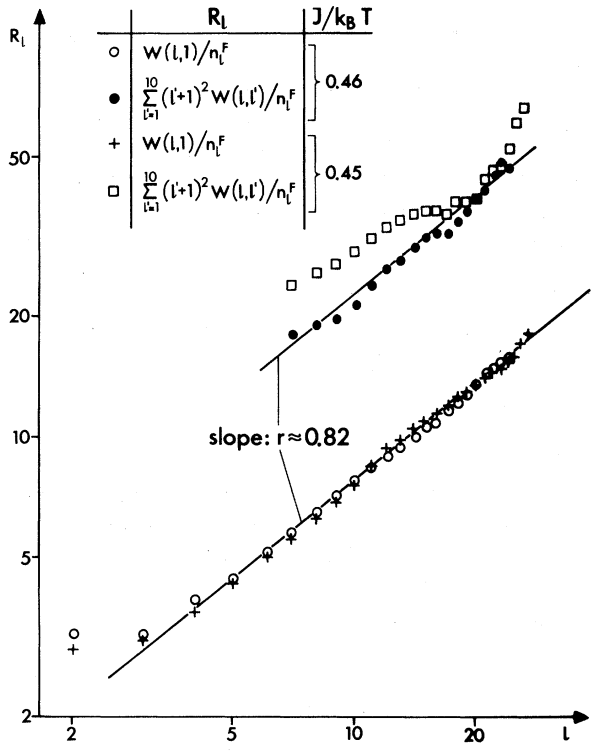

FIG. 2. Log–log plot of reaction rate R_l vs number l of spins for a 110×110 square Ising lattice. The partial reaction rates involving only single steps [$W(l,1)$] are also included. Two temperatures ($J/k_BT = 0.46$ and 0.45) close to the critical point ($J/k_BT_c = 0.4407$) are shown. All data for $l > 5$ have been smoothed over an interval $\Delta l = 7$; nevertheless strong critical flucatutions do not allow very accurate estimates, especially in the case $J/k_BT = 0.45$, where reactions up to $l' = 10$ are included. But at least the more accurate single-step data suggest that a power-law behavior is indeed a valid description, with r close to the theoretical value ($r_{\text{th}} = 0.93$). Note that the units of the ordinate axis are arbitrary.

III. STATIC PROPERTIES OF CLUSTERS

In order to make practical use of the cluster reaction and diffusion equation (2.33), we have to discuss the properties of the static "cluster concentration" n_l^f. Let us start with an "expansion" of the free energy per spin of the following type:

$$F = F_0 - k_B T \sum_{l=1}^{\infty} c(l, \epsilon, hl) , \quad h \equiv 2\mu_B H/k_B T \tag{3.1}$$

where F_0 is a background term nonsingular at T_c. In order to satisfy static scaling

$$F(\epsilon, h) = h^{1+1/\delta} \tilde{F}(\epsilon h^{-1/\beta\delta}) , \tag{3.2}$$

one finds that the singular parts of the expansion coefficients $c(l, \epsilon, hl)$ must have the form

$$c(l, \epsilon, hl) = \tilde{Q}(l) l^{-2-1/\delta} \tilde{c}(\epsilon l^{1/\beta\delta}, hl) ,$$
$$\tilde{Q}(\infty) \neq 0 , \infty . \tag{3.3}$$

Then similar expansions are obtained for the magnetization and energy per spin; of course

$$\langle \mu \rangle = -\frac{\partial F}{\partial H} = -\frac{\partial F_0}{\partial H} - 2\mu_B \sum_{l=1}^{\infty} l n_l ,$$
$$n_l \equiv -\left.\frac{\partial c(l, \epsilon, z)}{\partial z}\right|_{z=hl} \tag{3.4a}$$

and

$$E \equiv \frac{\partial}{\partial(1/k_BT)} \frac{F}{k_BT} = \frac{\partial}{\partial(1/k_BT)} \frac{F_0}{k_BT}$$
$$+ k_B T_c \sum_{l=1}^{\infty} l^{1/\beta\delta} n_l^E , \tag{3.4b}$$
$$n_l^E \equiv -\tilde{q}(l) l^{-2-1/\delta} \left.\frac{\partial \tilde{c}(y, hl)}{\partial y}\right|_{y=\epsilon l^{1/\beta\delta}} .$$

It is now appealing to interpret this (exact) expansion as a representation of the free energy in

terms of an "ideal gas of clusters," where $c(l, \epsilon, hl)$ is then the contribution of clusters labeled by l. Since h occurs in the combination hl only, there should be l more down spins than up spins in an l cluster. Studying the relaxation of the magnetization [Eq. (3.4a)], we are hence able to identify the coefficient n_l in Eq. (3.4a) with the "cluster concentration" n_l^f tentatively introduced in Sec. II. We now discuss the general properties of n_l which can be inferred from this definition [Eq. (3.1)].

According to the contour representation mentioned in Sec. II (see Fig. 1), there would not be many up spins belonging to a cluster, and therefore we would have $V_l \propto l$. However, we also require that the expansions Eqs. (3.1)–(3.4) be convergent series, i.e., that the contribution from large enough l be negligible. This convergence property is not always possible in the contour picture, owing to the percolation problem.[42] It turns out that above T_c in three dimensions a finite fraction of the reversed spins are part of one infinite percolating cluster.[43] Therefore near T_c the contour picture has to be abandoned, and we have to use a different prescription for the construction of clusters. Thus it is possible that $V_l = \tilde{V}(l) l^z$, with $z \geq 1$, $\tilde{V}(\infty)$ being finite and nonzero.[44]

Since we need z to discuss the local response (Sec. IV B), we now show that there is only one reasonable choice for z, consistent with our convergence requirements, in spite of the considerable arbitrariness in what is meant precisely by a cluster. Remember that the contour picture was obtained by considering order-parameter fluctuations on a length scale given by the lattice spacing. Since we are not interested in any details on such a fine scale, we rather consider fluctuations of a coarse-grained order parameter, Fig. 3. Fluctuations of various linear dimensions occur up to a linear dimension of about the correlation length ξ, while fluctuations which are considerably larger than ξ are very rare. *If we choose a definition of clusters such that they are a set of "basis functions" to represent these fluctuations rather directly, the series Eq.* (3.1) *would have the desired rapid convergence* (except right at T_c, where $\xi \to \infty$). From the requirement[45]

$$\langle V_l \rangle_{av} \equiv \frac{\int_0^\infty dl\, V_l l^K n_l}{\int_0^\infty dl\, l^K n_l} \propto \xi_d \propto |\epsilon|^{-\beta(\delta+1)}, \qquad (3.5a)$$

we therefore determine the exponent z, i.e.,[48]

$$V_l = \tilde{V}(l) l^{1+1/\delta}, \quad m_l = \tilde{m}(l) l^{-1/\delta}, \quad \tilde{m}(\infty) \neq 0, \infty, \qquad (3.5b)$$

where we introduced the averaged relative magnetization m_l of a cluster.

For numerical calculations we will use some explicit form for n_l.[32] One can make a guess for this form of c or $\tilde{c}$ in Eqs. (3.1) and (3.3) on the basis of Figs. 3 and 4. First, introducing the free energy F_l necessary to build up a cluster with l spins, we rewrite c as $c \propto e^{-F_l/k_B T}$. Second, the main contribution to F_l will be a "cluster volume energy" and a "cluster surface energy." Of course, for $V_l \gg \xi^d$ these two terms are given in terms of macroscopic quantities, i.e.,

$$F_l/k_B T = V_l h \langle \mu \rangle + V_l^{1-1/d} \epsilon^{2-\alpha-\nu} \times \text{const}$$
$$= lh + l^{1-1/d} \epsilon^{2-\alpha-\nu-(d-1)\beta/d} \times \text{const}.$$

Third, if we want to keep only two leading terms corresponding to these contributions also for $V_l \lesssim \xi^d$, the only possibility consistent both with Eq. (3.3) and a matching condition is

$$F_l/k_B T = hl + b\epsilon l^{1/\beta\delta},$$

which results in the Fisher cluster model,[32]

$$c(l, \epsilon, hl) = n_l = (1/b) n_l^E$$
$$= \tilde{Q}(\infty) l^{-2-1/\delta} e^{-b\epsilon l^{1/\beta\delta} - hl}, \qquad (3.6)$$

where the constants b and $\tilde{Q}(\infty)$ are related to critical amplitudes.

Although the same result was obtained by rather different arguments,[32] clearly the specific assumptions built into Eq. (3.6) are far more restrictive than our general arguments which we used for Eq. (3.5) or (2.32), respectively, and therefore the validity of Eq. (3.6) is rather questionable. In fact, Eq. (3.6) does not make any sense for $T > T_c$. Nevertheless, Monte Carlo simulations[15,43,46] have shown that Eq. (3.6) is fairly accurate for $T < T_c$

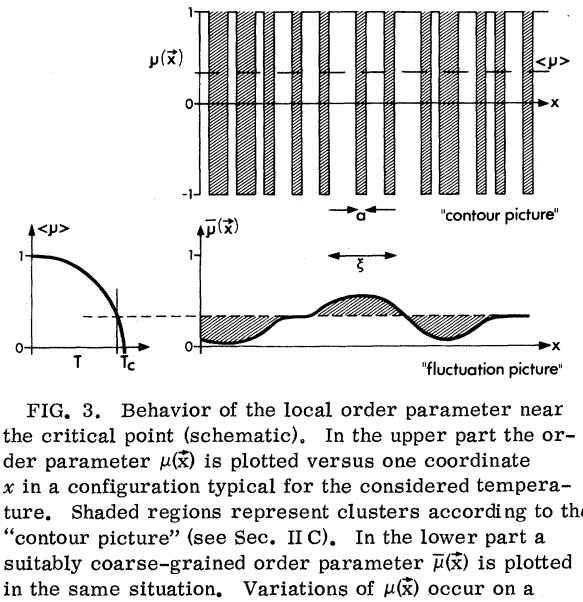

FIG. 3. Behavior of the local order parameter near the critical point (schematic). In the upper part the order parameter $\mu(\vec{x})$ is plotted versus one coordinate x in a configuration typical for the considered temperature. Shaded regions represent clusters according to the "contour picture" (see Sec. II C). In the lower part a suitably coarse-grained order parameter $\bar{\mu}(\vec{x})$ is plotted in the same situation. Variations of $\mu(\vec{x})$ occur on a length scale of the lattice spacing a, while the characteristic length for variations of $\bar{\mu}(\vec{x})$ is the correlation length ξ. Shaded regions then represent clusters according to the "fluctuation picture."

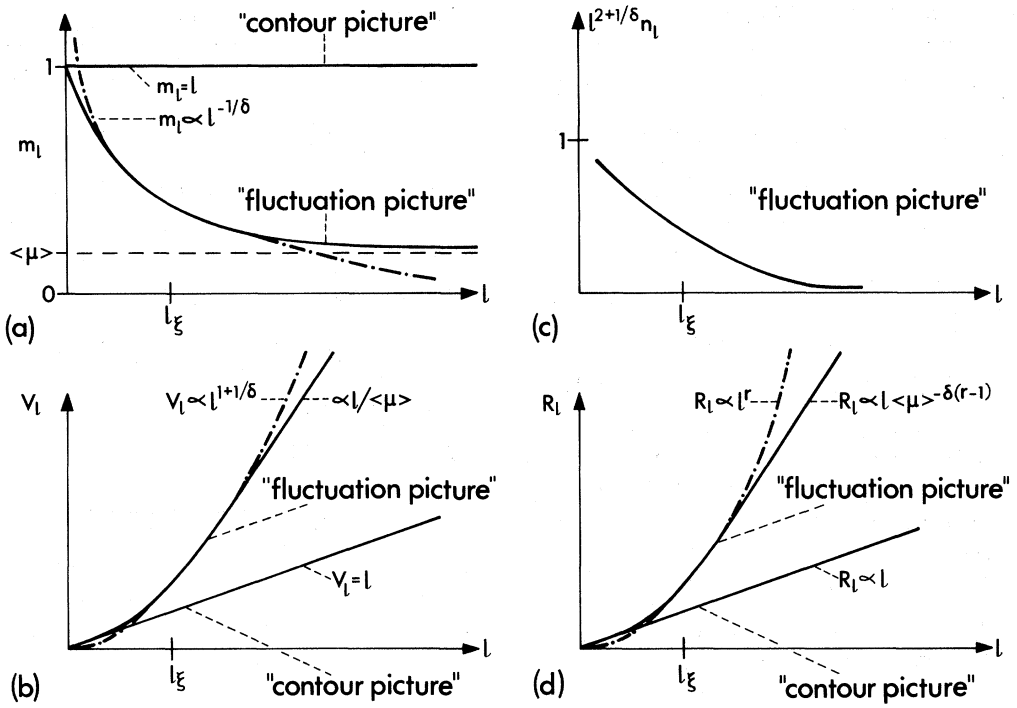

FIG. 4. Behavior of static cluster properties as a function of l (schematic): (a) cluster magnetization m_l; (b) cluster volume V_l; (c) normalized cluster concentration $l^{2+1/\delta} n_l$; (d) cluster reaction rate R_l. The full curves give the actual behavior of these respective quantities, both according to the "contour interpretation" and to the "fluctuation interpretation" of clusters, while the dash-dotted curve gives the power law which is valid in the vicinity of l_ξ. The behavior of $l^{2+1/\delta} n_l$ for the "contour picture" is rather uncertain; but it must contain a singularity (probably of δ-function type) for $l \to \infty$ in the paramagnetic region, owing to percolation effects (Ref. 43).

both in two and three dimensions,[47] and hence we will use Eq. (3.6) as an explicit example. Several generalizations of Eq. (3.6) consistent with Eq. (3.1) have been suggested,[48,49] but the validity of these approaches is too questionable to use them here. Hence most of our results will be based on the more general relations Eqs. (2.32) and (3.1)–(3.5) only.

Note that we do not imply that Eqs. (2.32) and (3.5b) are valid for $l \to \infty$ away from the critical point. We rather require the validity of these relations in the vicinity of $l = l_\xi$, which is defined by

$$V_{l_\xi} = \xi^d \,, \tag{3.7}$$

where then both arguments of $\tilde{c}(\epsilon l^{1/\beta\delta}, hl)$ are not yet large compared to unity. For $l \gg l_\xi$ (or, equivalently, $\epsilon l^{1/\beta\delta} \gg 1$, or $hl \gg 1$) both cluster volume, magnetization, and reaction rate will be determined by geometric relations[50]; see Fig. 4. Since the cluster concentration is negligible in this regime, this regime does not contribute significantly to the response functions we are going to calculate, and we leave this geometric regime henceforth outside of concentration.

Discussing $\vec{q}$-dependent response we also need information on the static correlation functions in the cluster representation, and hence we define the functions $p_{\mu\mu}(l, \vec{q})$, $p_{\mu E}(l, \vec{q})$, $p_{E\mu}(l, \vec{q})$, and $p_{EE}(l, \vec{q})$ by

$$n_l(\vec{x}) = n_l[1 - \delta h e^{i\vec{q}\cdot\vec{x}} p_{\mu\mu}(l, \vec{q})] \,, \tag{3.8a}$$

or

$$n_l(\vec{x}) = n_l[1 + b(\delta T/T_c)\, e^{\vec{q}\cdot\vec{x}} p_{\mu E}(l, \vec{q})] \,, \tag{3.8b}$$

and

$$n_l^E(\vec{x}) = n_l^E[1 - \delta h e^{i\vec{q}\cdot\vec{x}} p_{E\mu}(l, \vec{q})] \,, \tag{3.8c}$$

or

$$n_l^E(\vec{x}) = n_l^E[1 + b(\delta T/T_c) e^{i\vec{q}\cdot\vec{x}} p_{EE}(l, \vec{q})] \,. \tag{3.8d}$$

From the definitions for the susceptibilities, $\langle \mu(\vec{x}) \rangle = \langle \mu \rangle + \delta H e^{i\vec{q}\cdot\vec{x}} \chi_{\mu\mu}(\vec{q})$, etc., [cf., Eq. (2.7)], we get immediately

$$\chi_{\mu\mu}(\vec{q}) \equiv \frac{4\mu_B^2}{k_B T} \sum_{l=1}^{\infty} l n_l\, p_{\mu\mu}(l, \vec{q}) \,, \tag{3.9a}$$

$$\chi_{\mu E}(\vec{q}) = -\frac{2\mu_B b}{T_c} \sum_{l=1}^{\infty} l n_l\, p_{\mu E}(l, \vec{q}) \,, \tag{3.9b}$$

$$\chi_{E\mu}(\vec{q}) = -2\mu_B \sum_{l=1}^{\infty} l^{1/\beta\delta} n_l^E p_{E\mu}(l, \vec{q}) \,, \tag{3.9c}$$

$$\chi_{EE}(\vec{q}) \equiv C(\vec{q}) = b k_B \sum_{l=1}^{\infty} l^{1/\beta\delta} n_l^E p_{EE}(l, \vec{q}) \,. \tag{3.9d}$$

Because $\partial^2 F/\partial H \partial T = \partial^2 F/\partial T \partial H$, we have $T\chi_{\mu E}(\vec{q}) = \chi_{E\mu}(\vec{q})$ and hence[51]

$$bn_l l p_{\mu E}(l, \vec{q}) = l^{1/\beta\delta} n_l^E p_{E\mu}(l, \vec{q}) \ . \tag{3.10}$$

It is easy to see that the scaling of the susceptibilities [Eqs. (2.18) and (2.8a)] and of the cluster concentrations [Eqs. (3.3) and (3.4)] implies the following scaling relations for the cluster transforms p_{BC} of the correlation functions,

$$p_{\mu\mu}(l, \vec{q}) = l\tilde{p}_{\mu\mu}(|\epsilon|^{\beta\delta} l, hl, \vec{q}\,|\epsilon|^{-\nu}) \ , \tag{3.11a}$$

$$p_{\mu E}(l, \vec{q}) = l^{1/\beta\delta}\tilde{p}_{\mu E}(|\epsilon|^{\beta\delta} l, hl, \vec{q}\,|\epsilon|^{-\nu}) \ , \tag{3.11b}$$

$$p_{EE}(l, \vec{q}) = l^{1/\beta\delta}\tilde{p}_{EE}(|\epsilon|^{\beta\delta} l, hl, \vec{q}\,|\epsilon|^{-\nu}) \ , \tag{3.11c}$$

with all the $\tilde{p}_{BC}(0, 0, 0) = \text{const}$.

As an illustration of these formal definitions, we consider as a simple example the case $d = 4$, $h = 0$ explicitly, where ($\gamma = 1$, $\nu = \frac{1}{2}$)

$$\chi(\vec{q}) = \frac{4\mu_B^2}{k_B T}\,|\epsilon|^{-\gamma}\tilde{\chi}\,\frac{1}{1 + (\vec{q}\,|\epsilon|^{-\nu}/\kappa_0)^2} \ . \tag{3.12}$$

Assuming the Fisher cluster model [Eq. (3.6)] to be valid, one finds for $p_{\mu\mu}(l, \vec{q})$ the result

$$p_{\mu\mu}(l, \vec{q}) = [b/\tilde{Q}(\infty)]\,\tilde{\chi} l \times \exp[-l^{1/\beta\delta}(\vec{q}/\kappa_0)^2 b] \ . \tag{3.13}$$

In this way we express the static correlation functions in terms of wave-vector-dependent cluster concentrations.

IV. RELAXATION TIMES IN CLUSTER-DYNAMICS FORMULATION

A. Wave-vector-dependent relaxation times

In this section we calculate the relaxation times defined in Sec. II [Eqs. (2.10)], i.e., the initial relaxation times and then the "total" relaxation times as functions of field and wave vector from the cluster-reaction equation (2.33), making use of the scaling behavior of the static cluster properties derived in Sec. III [Eqs. (3.4) and (3.11)]. In order to fix the undetermined temperature dependence of $\tilde{R}(\infty)$, we first consider the initial slowing down.

If the equilibrium is slightly disturbed, we may express the time dependence of order parameter and energy by[52]

$$\langle \mu(\vec{x}, t)\rangle = -\frac{\partial F_0}{\partial H} - 2\mu_B \sum_{l=1}^{\infty} l\bar{n}_l(\vec{x}, t) \ , \tag{4.1a}$$

$$E(\vec{x}, t) = \frac{\partial}{\partial(1/k_B T)}\,\frac{F_0}{k_B T} + k_B T_c \sum_{l=1}^{\infty} l^{1/\beta\delta}\bar{n}_l^E(\vec{x}, t) \ . \tag{4.1b}$$

This generalization of Eq. (3.4) is a definition for the time-dependent cluster concentrations $\bar{n}_l^E(t)$ of the order parameter and energy, respectively. The basic physical assumption, that the time dependence of $\bar{n}_l(\vec{x}, t)$ is given by Eq. (2.33) [and an analogous equation for $\bar{n}_l^E(\vec{x}, t)$], has been justified in Sec. II. Using Eq. (2.9) for a small change of the field δh, we find from Eq. (2.15),

$$\frac{1}{\tau_{\mu\mu}^I(0)} = \frac{1}{\chi\delta H}\,\frac{d}{dt}\,\langle \mu(t)\rangle\big|_{t=0} \ , \tag{4.2a}$$

and using Eqs. (4.1a) and (2.31) this is expressed as

$$\frac{1}{\tau_{\mu\mu}^I(0)} = -\frac{1}{\chi\delta H}\,2\mu_B \sum_{l=1}^{\infty} l n_l R_l \times\left(\frac{\partial^2}{\partial l^2}\,g_l'(0) + \frac{\partial}{\partial l}\,(\ln R_l n_l)\,\frac{\partial}{\partial l}\,g_l(0)\right) \ . \tag{4.2b}$$

Now Eqs. (3.4) and (3.3) imply

$$g_l(0) \equiv [n_l(h + \delta h) - n_l(h)]/n_l = -l\delta h\,\frac{d}{dz}\ln\frac{d}{dz}\,\tilde{c}(y, z) \xrightarrow[l\to\infty]{} l\delta h \times \text{const} \ , \tag{4.3}$$

where the constant in Eq. (4.3) is, in general, nonzero, since it is related to the critical amplitude of the susceptibility. Then we get

$$\frac{1}{\tau_{\mu\mu}^I(0)} = \frac{1}{\chi}\,\tilde{R}(\infty)\sum_{l=1}^{\infty} l^{-1-1/\delta+\gamma}\,\frac{d\tilde{c}}{dz} \times\left(\frac{2 + 1/\delta - \gamma}{l} + \frac{d}{dl}\ln\frac{d\tilde{c}}{dz}\right) \times \text{const}, \tag{4.4}$$

which should be compared with Eq. (2.15). We evaluate the sum in Eq. (4.4) right at the critical point. If $\gamma < 1 + 1/\delta$, the sum is finite and hence $\tilde{R}(\infty)$ must also be finite, which was already suggested on the basis of the Monte Carlo results (Sec. II D). In the other case we would get from Eq. (2.15) that

$$R(\infty) \propto |\epsilon|^{(\gamma-1-1/\delta)\beta\delta} \ , \quad \gamma > 1 + 1/\delta \tag{4.5}$$

but this case is highly unlikely [cf., Eq. (2.32) and Sec. II D], since[15] the dominant contributions to the reactions of an l cluster come from l' clusters with $l' \approx 1$, and hence $\tilde{R}(\infty)$ should not be critical. Next we consider the linear response to time-dependent changes of magnetic field or temperature [cf. Eq. (2.7)] and generalize Eq. (3.8) to[53]

$$\bar{n}_l(\vec{x}, t) = n_l[1 - \delta h e^{i(\vec{q}\cdot\vec{x} + \omega t)} p_{\mu\mu}(l, \vec{q}, \omega)] \ , \tag{4.6a}$$

or

$$\bar{n}_l(\vec{x}, t) = n_l[1 + b(\delta T/T_c) e^{i(\vec{q}\cdot\vec{x} + \omega t)} p_{\mu E}(l, \vec{q}, \omega)] \ , \tag{4.6b}$$

and

$$\bar{n}_l^E(\vec{x}, t) = n_l^E[1 - \delta h e^{i(\vec{q}\cdot\vec{x} + \omega t)} p_{E\mu}(l, \vec{q}, \omega)] \ , \tag{4.6c}$$

or

$$\bar{n}_l^E(\vec{x}, t) = n_l^E[1 + b(\delta T/T_c) e^{i(\vec{q}\cdot\vec{x} + \omega t)} p_{EE}(l, \vec{q}, \omega)] \ . \tag{4.6d}$$

The dynamic susceptibilities are then given by the generalization of Eq. (3.9), i.e.,

$$\chi_{\mu\mu}(\vec{q},\omega)=\frac{4\mu_B^2}{k_B T}\sum_{l=1}^{\infty} l n_l\, p_{\mu\mu}(l,\vec{q},\omega)\,, \tag{4.7a}$$

$$\chi_{\mu E}(\vec{q},\omega)=\frac{-2\mu_B b}{T_c}\sum_{l=1}^{\infty} l n_l p_{\mu E}(l,\vec{q},\omega)\,, \tag{4.7b}$$

$$\chi_{E\mu}(\vec{q},\omega)=-2\mu_B\sum_{l=1}^{\infty} l^{1/\beta\delta} n_l^E p_{E\mu}(l,\vec{q},\omega)\,, \tag{4.7c}$$

and

$$\chi_{EE}(\vec{q},\omega)=bk_B\sum_{l=1}^{\infty} l^{1/\beta\delta} n_l^E p_{EE}(l,\vec{q},\omega)\,. \tag{4.7d}$$

Owing to the symmetry relation[54]

$$\langle \mu(0,0)\mathcal{H}(\vec{x},t)\rangle=\langle \mathcal{H}(0,0)\mu(\vec{x},t)\rangle\,,$$

we require as a generalization of Eq. (3.10) that

$$l b n_l p_{\mu E}(l,\vec{q},\omega)=l^{1/\beta\delta} n_l^E p_{E\mu}(l,\vec{q},\omega)\,. \tag{4.8}$$

From Eq. (2.10a) we see that the desired relaxation times are [$p_{BC}(l,\vec{q},0)=p_{BC}(l,\vec{q})$]

$$\tau_{\mu\mu}(\vec{q})=-\frac{1}{\sum_{l=1}^{\infty} l n_l p_{\mu\mu}(l,\vec{q})}\sum_{l=1}^{\infty} l n_l p_{\mu\mu}^{(1)}(l,\vec{q})\,,$$
$$p_{\mu\mu}^{(1)}(l,\vec{q})=\frac{\partial}{\partial(i\omega)}\,p_{\mu\mu}(l,\vec{q},\omega)\Big|_{\omega=0}\,, \tag{4.9}$$

the expression for $\tau_{\mu E}(\vec{q})$, etc. being analogous.

Now we determine $p_{\mu\mu}(l,\vec{q},\omega)$, etc., combining Eq. (4.6) with Eqs. (2.33) and (2.32), which gives

$$\frac{i\omega}{R_l}\,p_{\mu\mu}(l,\vec{q},\omega)=\frac{d}{dl}\left[p_{\mu\mu}(l,\vec{q},\omega)-p_{\mu\mu}(l,\vec{q},0)\right]\frac{d}{dl}\left(\ln R_l n_l\right)+\frac{d^2}{dl^2}\left[p_{\mu\mu}(l,\vec{q},\omega)-p_{\mu\mu}(l,\vec{q},0)\right]$$
$$-q^2a^2\,\frac{c}{l^2}\left[p_{\mu\mu}(l,\vec{q},\omega)-p_{\mu\mu}(l,\vec{q},0)\right]\,, \tag{4.10}$$

the equation for $p_{\mu E}$ being completely analogous. Using an equation similar to Eq. (2.33) for $\bar{n}_l^E(\vec{x},t)$ gives analogous equations for $p_{E\mu}(l,\vec{q},\omega)$ and $p_{EE}(l,\vec{q},\omega)$, and because of Eq. (4.8) we must have $R_l^E\propto R_l$, i.e., the same exponent r applies for order parameter and energy relaxation which will give us the equality Eq. (4.18a) for the exponents $\Delta_{\mu\mu}$, etc. In deriving Eq. (4.10) it is important to note that the final equilibrium state n_l^f to be used in Eq. (2.38) is not n_l but rather

$$n_l^f=n_l\left[1-\delta h e^{i(\vec{q}\vec{x}+\omega t)}p_{\mu\mu}(l,\vec{q})\right]\,, \tag{4.11}$$

i.e., the cluster distribution which would be in equilibrium with the applied field at every instant of time. Next we transform variables by $X=|\epsilon|^{\beta\delta}l$, and using $p_{\mu\mu}(l,\vec{q},\omega)\equiv P_{\mu\mu}(X,\vec{q},\omega)$, $n(X)\equiv n_l$, $R(X)\equiv R_l$, we find with the help of Eq. (2.35) that

$$\frac{i\omega}{\tilde{R}(\infty)\,|\epsilon|^{(2-\tau)\beta\delta}}\,X^{-\tau}P_{\mu\mu}(X,\vec{q},\omega)=\frac{d}{dX}\left[P_{\mu\mu}(X,\vec{q},\omega)-P_{\mu\mu}(X,\vec{q},0)\right]\frac{d}{dX}\left[\ln R(X)n(X)\right]$$
$$+\frac{d^2}{dX^2}\left[P_{\mu\mu}(X,\vec{q},\omega)-P_{\mu\mu}(X,\vec{q},0)\right]-\frac{q^2a^2c}{X^2}\left[P_{\mu\mu}(X,\vec{q},\omega)-P_{\mu\mu}(X,\vec{q},0)\right]\,. \tag{4.12}$$

Since critical phenomena deal with the limit $q\epsilon^{-\nu}=\text{const}$, $\epsilon\to 0$, $q\to 0$, the last term in Eq. (4.12) which is of order q^2a^2 may be neglected. Hence, near T_c this diffusive contribution yields unimportant corrections to scaling only. From Eqs. (3.3), (3.4), and (3.11a) it then follows that the solution of Eq. (4.12) must have the form

$$P_{\mu\mu}(X,\vec{q},\omega)=l\tilde{p}_{\mu\mu}\left(|\epsilon|\,l^{1/\beta\delta},\,hl,\,\vec{q}\,|\epsilon|^{-\nu},\,\frac{\omega}{\tilde{R}(\infty)}\,|\epsilon|^{-(2-\tau)\beta\delta}\right)\,, \tag{4.13}$$

and the generalizations of Eqs. (3.11b) and (3.11c) are analogous. Combining Eqs. (4.13), (4.7), and (3.3), one finds that the susceptibilities are consistent with extended dynamic scaling hypotheses[4]

$$\chi_{\mu\mu}(\vec{q},\omega)=\frac{4\mu_B^2}{k_B T}\,|\epsilon|^{-\gamma}\int_0^{\infty} dX\,X^2 n(X)\tilde{p}_{\mu\mu}\left(X,\,h\,|\epsilon|^{-\beta\delta}X,\,\vec{q}\,|\epsilon|^{-\nu},\,\frac{\omega}{\tilde{R}(\infty)}\,|\epsilon|^{-(2-\tau)\beta\delta}\right)\,, \tag{4.14a}$$

$$\chi_{\mu E}(\vec{q},\omega)=-2\mu_B\,\frac{b}{T_c}\,|\epsilon|^{-1+\beta}\int_0^{\infty} dX\,X^{1+1/\beta\delta}\,n(X)\tilde{p}_{\mu E}\left(X,\,h\,|\epsilon|^{-\beta\delta}X,\,\vec{q}\,|\epsilon|^{-\nu},\,\frac{\omega}{\tilde{R}(\infty)}\,|\epsilon|^{-(2-\tau)\beta\delta}\right)\,, \tag{4.14b}$$

and

$$\chi_{EE}(\vec{q},\omega)=bk_B\,|\epsilon|^{-\alpha}\int_0^{\infty} dX\,X^{2/\beta\delta}\,n^E(X)\tilde{p}_{EE}\left(X,\,h\,|\epsilon|^{-\beta\delta}X,\,\vec{q}\,|\epsilon|^{-\nu},\,\frac{\omega}{\tilde{R}(\infty)}\,|\epsilon|^{-(2-\tau)\beta\delta}\right)\,. \tag{4.14c}$$

In Eq. (4.14c) we have assumed that the specific-heat exponent $\alpha>0$, since otherwise the summation in Eq. (4.7d) could not be replaced by an integration for $\omega=0$.

Note that Eqs. (4.12) and (4.14) also imply universality properties—apart from just one scale factor for the $\tilde{p}_{BC}$ and each of its arguments, the $\tilde{p}_{BC}$ are universal functions of any set of exponents β, δ, and r.

In order to obtain somewhat more explicit results, we use the expansion [cf., Eq. (4.9)]

$$p_{\mu\mu}(l, \vec{q}, \omega) = p_{\mu\mu}(l, \vec{q}) + i\omega p^{(1)}_{\mu\mu}(l, \vec{q}) + \frac{(i\omega)^2}{2!} p^{(2)}_{\mu\mu}(l, \vec{q}) + \cdots \quad (4.15)$$

and analogous expansions for $p_{\mu E}(l, \vec{q}, \omega)$ and $p_{EE}(l, \vec{q}, \omega)$ to solve Eq. (4.10). One first obtains

$$\frac{d^2}{dl^2} p^{(1)}_{\mu\mu}(l, \vec{q}) + \frac{d}{dl} p^{(1)}_{\mu\mu}(l, \vec{q}) \frac{d}{dl} (\ln R_l n_l) - \frac{p_{\mu\mu}(l, \vec{q})}{R_l} = 0 , \quad (4.16a)$$

and analogous equations for $p^{(1)}_{\mu E}(l, \vec{q})$ and $p^{(1)}_{EE}(l, \vec{q})$.

Equation (4.16a) is solved by

$$p^{(1)}_{\mu\mu}(l, \vec{q}) = C^0_{\mu\mu}(\vec{q}) + C^1_{\mu\mu}(\vec{q}) \int_0^l \frac{dl_1}{R_{l_1} n_{l_1}} - \int_0^l \frac{dl_1}{R_{l_1} n_{l_1}} \int_{l_1}^{\infty} dl_2 \, n_{l_2} p_{\mu\mu}(l_2, \vec{q}) , \quad (4.16b)$$

and analogous expressions for $p^{(1)}_{\mu E}(l, \vec{q})$ and $p^{(1)}_{EE}(l, \vec{q})$. The integration constants $C^1_{BC}(\vec{q})$ must be zero, since otherwise the relaxation times $\tau_{BC}(\vec{q})$ would be always infinite. Also, $C^0_{BC}(\vec{q})$ must be zero, since physically the boundary condition $p_{BC}(l=0, \vec{q}, \omega) = 0$ is required. This follows from the fact that $\tilde{C}$ depends on ϵ and h in the combination $|\epsilon| l^{1/\beta\delta}$ and hl only, and hence $\tilde{C}$ becomes unsensitive to changes of ϵ and h for $l \to 0$. From Eqs. (4.16) and (4.9) we get the desired relaxation times, their amplitudes being expressed as triple integrals,

$$\tau_{\mu\mu}(\vec{q}) = |\epsilon|^{-(2-r)\beta\delta} \Bigg(\int_0^{\infty} dX \, X n(X) \int_0^X \frac{dX_1}{R(X_1) n(X_1)} \int_{X_1}^{\infty} dX_2 \, n(X_2) X_2 \tilde{p}_{\mu\mu}(X_2, h|\epsilon|^{-\beta\delta} X_2, \vec{q}|\epsilon|^{-\nu}) \Big/ \int_0^{\infty} dX \, X^2 n(X) \tilde{p}_{\mu\mu}(X, h|\epsilon|^{-\beta\delta} X, \vec{q}|\epsilon|^{-\nu}) \Bigg) , \quad (4.17a)$$

$$\tau_{\mu E}(\vec{q}) = |\epsilon|^{-(2-r)\beta\delta} \Bigg(\int_0^{\infty} dX \, X n(X) \int_0^X \frac{dX_1}{R(X_1) n(X_1)} \int_{X_1}^{\infty} dX_2 \, n(X_2) X_2^{1/\beta\delta} \tilde{p}_{\mu E}(X_2, h|\epsilon|^{-\beta\delta} X_2, \vec{q}|\epsilon|^{-\nu}) \Big/ \int_0^{\infty} dX \, X^{1+1/\beta\delta} n(X) \tilde{p}_{\mu\mu}(X, h|\epsilon|^{-\beta\delta} X, \vec{q}|\epsilon|^{-\nu}) \Bigg) , \quad (4.17b)$$

and

$$\tau_{EE}(\vec{q}) = |\epsilon|^{-(2-r)\beta\delta} \Bigg(\int_0^{\infty} dX \, X^{1/\beta\delta} n^E(X) \int_0^X \frac{dX_1}{R^E(X_1) n^E(X_1)} \int_{X_1}^{\infty} dX_2 \, n^E(X_2) X_2^{1/\beta\delta} \tilde{p}_{EE}(X_2, h|\epsilon|^{-\beta\delta} X_2, \vec{q}|\epsilon|^{-\nu}) \Big/ \int_0^{\infty} dX \, X^{2/\beta\delta} n^E(X) \tilde{p}_{EE}(X, h|\epsilon|^{-\beta\delta} X, \vec{q}|\epsilon|^{-\nu}) \Bigg) . \quad (4.17c)$$

Of course, the higher-order coefficients in the frequency expansion of $\chi_{BC}(\vec{q}, \omega)$ can be obtained similarly, but the coefficient of ω^m involves a $(2m+1)$-fold integration. In Eq. (4.17c) we used the fact that the exponents of R^E_l and R_l are the same; in fact, the symmetry relation Eq. (4.8) also implicitly relates the amplitudes $\tilde{R}_l(\infty)$ and $\tilde{R}^E_l(\infty)$; therefore there is only one dynamic scale factor, and the amplitude ratios $\tau_{\mu E}(\vec{q})/\tau_{\mu\mu}(\vec{q})$, $\tau_{EE}(\vec{q})/\tau_{\mu\mu}(\vec{q})$ are given in terms of static quantities only.

For $r < 1 + 1/\delta$, $\tilde{R}(\infty)$ is finite at the critical point, and hence the critical exponents are

$$\Delta_{\mu\mu} = \Delta_{\mu E} = \Delta_{EE} = (2 - r)\beta\delta , \quad (4.18a)$$

while for the case $r \geq 1 + 1/\delta$, Eq. (4.5) has to be taken into account and then

$$\Delta_{\mu\mu} = \Delta_{\mu E} = \Delta_{EE} = \gamma ; \quad (4.18b)$$

therefore the cluster-reaction equation also implies that the Abe Hatano[26] inequality Eq. (2.17) holds, if γ_{BC} is the exponent of the initial slowing down. As discussed before, the case $r > 1 + 1/\delta$ is highly unlikely, and thus the result $\Delta_{\mu\mu} > \gamma$ is made plausible.

We calculated numerically the relaxation times $\tau_{\mu\mu}(0)$, $\tau_{\mu E}(0)$, and $\tau_{EE}(0)$ as functions of the scaled magnetic field for $d = 2$, 3, and 4,[55] where for $n(X)$ the Fisher cluster model[32] [Eq. (3.6)] was used, and then we also have simply

$$\tilde{p}_{\mu\mu}(X, h|\epsilon|^{-\beta\delta} X, 0) = \tilde{p}_{\mu E}(X, h|\epsilon|^{-\beta\delta} X, 0) = \tilde{p}_{EE}(X, h|\epsilon|^{-\beta\delta} X, 0) = 1 ,$$

as can be seen by deriving the corresponding static susceptibilities [Eq. (3.9)] directly from Eq. (3.6). We have shown previously[11] that these results are in reasonable agreement with Monte Carlo calculations for $d = 2$. A further example is given in Fig. 5 for $d = 3$.

B. Local relaxation times

Here we give the extension of our method by which the autocorrelation times defined in Eq. (2.11) are obtained, and discuss their scaling and universality behavior.

First we note from Eqs. (2.6), (2.8), (2.10), and (2.11), that τ^A_{BC} may be expressed in terms of the corresponding $\tau_{BC}(\vec{q})$,

$$\tau^A_{BC} = \sum_q S_{BC}(\vec{q})\tau_{BC}(\vec{q}) \Big/ \sum_q S_{BC}(\vec{q}, 0) \,. \tag{4.19}$$

Since the static local response

$$\sum_q S_{BC}(\vec{q}, 0) = \langle B(0,0)C(0,0)\rangle - \langle B\rangle\langle C\rangle$$

stays finite at T_c, it cannot be calculated by our scaling expressions for the cluster concentrations which are valid for large l only. However, in the cases we are considering here, this static local response can be calculated directly from its definition [Eq. (2.11b)] and hence we get

$$\tau^A_{\mu\mu} = \frac{|\epsilon|^{-\Delta^A_{\mu\mu}} U_d}{\mu_B^2 - \langle\mu\rangle^2} \int_0^\infty dY\, Y^{d-1} \int_0^\infty dX\, X n(X) \int_0^X \frac{dX_1}{R(X_1)n(X_1)} \int_{X_1}^\infty dX_2\, X_2\, n(X_2)\tilde{p}_{\mu\mu}(X_2, h|\epsilon|^{-\beta\delta}X_2, Y)\tilde{\chi}(Y, h|\epsilon|^{-\beta\delta}) \,, \tag{4.20a}$$

$$\tau^A_{\mu E} = \frac{|\epsilon|^{-\Delta^A_{\mu E}} U_d}{\mu_B B[J(0) - |E|]} \int_0^\infty dY\, Y^{d-1} \int_0^\infty dX\, X n(X) \int_0^X \frac{dX_1}{R(X_1)n(X_1)} \int_{X_1}^\infty dX_2\, X_2^{1/\beta\delta} \times n(X_2)\tilde{p}_{\mu E}(X_2, h|\epsilon|^{-\beta\delta}X_2, Y)\tilde{\chi}_{\mu E}(Y, h|\epsilon|^{-\beta\delta}) \,, \tag{4.20b}$$

and

$$\tau^A_{EE} = \frac{|\epsilon|^{-\Delta^A_{EE}} U_d}{\langle \mathcal{H}(0,0)^2\rangle - \langle\mathcal{H}\rangle^2} \int_0^\infty dY\, Y^{d-1} \int_0^\infty dX\, X^{1/\beta\delta} n^E(X) \int_0^X \frac{dX_1}{R^E(X_1)n^E(X_1)} \int_{X_1}^\infty dX_2\, X_2^{1/\beta\delta} \times n^E(X_2)\tilde{p}_{EE}(X_2, h|\epsilon|^{-\beta\delta}X_2, Y)\tilde{\chi}_{EE}(Y, h|\epsilon|^{-\beta\delta}) \,. \tag{4.20c}$$

Here U_d denotes the surface area of a d-dimensional unit sphere, and the autocorrelation exponents are [cf. also Eq. (2.20)]

$$\Delta^A_{\mu\mu} = \Delta_{\mu\mu} - 2\beta \,, \quad \Delta^A_{\mu E} = \Delta_{\mu\mu} - 1 + \alpha \,, \quad \Delta^A_{EE} = \Delta_{\mu\mu} - 2 + 2\alpha \,; \tag{4.21}$$

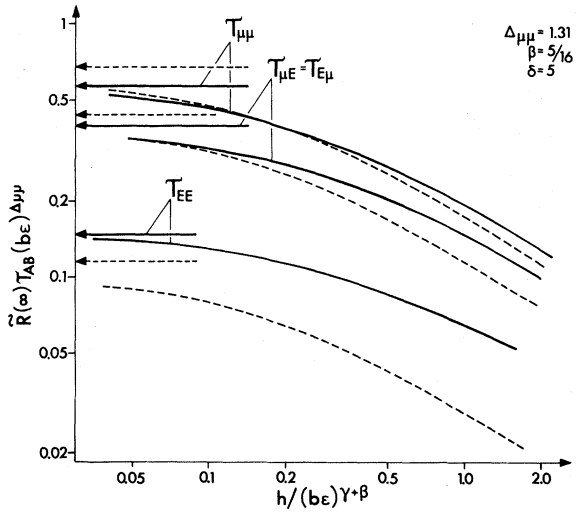

FIG. 5. Scaled relaxation times plotted vs scaled field for $d=3$, and the renormalization group estimate (Ref. 8) for $\Delta_{\mu\mu}$. Full curves were obtained from Eq. (4.17) by numerical integration, while the dashed curves are the result of the approximations Eqs. (5.10) and (5.17). The arrows indicate the asymptotes for $n \to 0$.

of course, Eq. (4.20c) makes sense only if $\Delta^A_{EE} > 0$. In (4.21) it was used that $\sum_q S_{\mu E}(\vec{q}) \propto |\epsilon|^\beta$, and static scaling was invoked. While the exponents Eq. (4.21) have been found previously,[35] Eqs. (4.20) provide amplitude relations similar to the case of the $\vec{q}$-dependent relaxation times.

The practical use of Eq. (4.20) is hampered by the fact, however, the full knowledge of p_{BC} is required in order to perform the $\vec{q}$ (i.e., Y) integration, which is not present in Eq. (4.17). Therefore it is interesting to construct an alternative approach, where the response of the cluster concentrations to local fields is considered. The corresponding analog of Eq. (4.6) is

$$\bar{n}_l(\vec{x}, t) = n_l[1 - \delta h^j e^{i\omega t} p^A_{\mu\mu}(l, \omega)] \,, \tag{4.22}$$

the definitions for $p^A_{\mu E}(l, \omega)$ and $p^A_{EE}(l, \omega)$ being analogous. The changes δh^j, δT^j are applied on one site j only. The further treatment is similar to the procedure from Eqs. (4.6)–(4.17), but we must take care to specify the static limit $p^A_{BC}(l)$ correctly. Applying the same field field δh^j (or δT^j) on all sites j of the cluster and summing up the individual contributions, the total effect should be given by $p_{BC}(l, \vec{q}=0)$ in linear response. Hence we have

$$p^A_{BC}(l) = p_{BC}(l, \vec{q}=0)/V_l \,, \tag{4.23}$$

since there are V_l sites in the cluster. Using Eq. (3.5b) we now get

$$\tau^A_{\mu\mu} = \frac{|\epsilon|^{-\Delta^A_{\mu\mu}}}{\mu_B^2 - \langle\mu\rangle^2} \frac{1}{\tilde{V}(\infty)} \int_0^\infty dX\, Xn(X) \int_0^X \frac{dX_1}{R(X_1)n(X_1)} \int_{X_1}^\infty dX_2\, X_2^{-1/\beta\delta} n(X_2) \tilde{p}_{\mu\mu}(X_2, h|\epsilon|^{-\beta\delta} X_2, 0) , \tag{4.24a}$$

$$\tau^A_{\mu E} = \frac{|\epsilon|^{-\Delta^A_{\mu E}}}{\mu_B B[J(0) - |E|]} \frac{1}{\tilde{V}(\infty)} \int_0^\infty dX\, Xn(X) \int_0^X \frac{dX_1}{R(X_1)n(X_1)} \int_{X_1}^\infty dX_2\, X_2^{1/\beta\delta - 1 - 1/\delta} n(X_2) \tilde{p}_{\mu E}(X_2, h|\epsilon|^{-\beta\delta} X_2, 0) , \tag{4.24b}$$

and

$$\tau^A_{EE} = \frac{|\epsilon|^{-\Delta^A_{EE}}}{\langle[\mathcal{H}(0,0)]^2\rangle - \langle\mathcal{H}\rangle^2} \frac{1}{\tilde{V}(\infty)} \int_0^\infty dX\, X^{1/\beta\delta} n^E(X) \int_0^X \frac{dX_1}{R^E(X_1)n^E(X_1)} \int_{X_1}^\infty dX_2\, X_2^{1/\beta\delta - 1 - 1/\delta} \times n^E(X_2) \tilde{p}_{EE}(X_2, h|\epsilon|^{-\beta\delta} X_2, 0) . \tag{4.24c}$$

The exponents in Eq. (4.24) are the same as before, but the amplitudes are now considerably simpler; apart from the common scale factor $\tilde{V}(\infty)$ only the same static quantities as in $\tau_{BC}(0)$ are involved. As an example we also evaluated Eq. (4.24) numerically, using again Eq. (3.6), where $\tilde{p}_{BC} = 1$. The results for $d = 2$ are shown in Fig. 6, where the relaxation times are plotted versus scaled field.

We conclude by remarking that any choice other than Eq. (3.5) for V_l in Eq. (4.23) inevitably leads to wrong Δ^A_{BC} exponents, if also Eq. (3.3) is assumed for the static-cluster distribution. This means in particular that Eq. (3.3) [with $\lim_{l\to\infty} \tilde{c} = 0$] cannot be valid, if the "contour-interpretation" is used, where $V_l = l$. Equation (3.5) was advanced on different grounds,[45] and we have introduced it here on the basis of percolation arguments[43] (Sec. III), but the above result is perhaps the most convincing argument that Eq. (3.5) is, in fact, the *only* possibility consistent with "hyperscaling" $d\nu = 2 - \alpha$. In addition, Eq. (3.5) is also indispensable to get the correct critical exponent for both the diffusion constant in the spin-exchange model,[18] and the width of the region of rounding phenomena in finite systems.[56]

V. EIGENFREQUENCIES AND SCALING FUNCTIONS

The relaxation times obtained in Sec. IV give some integral information on the relaxation spectrum of the system only. Although it was indicated [after Eq. (4.17)] that this approach can, in principle, be generalized to construct the dynamic susceptibilities systematically, this task seems difficult in practice. Therefore we give a different approach in this section, solving Eq. (2.33) directly in terms of an eigenfunction expansion. Of course, in this case n_l must be specified, and we give exact analytic solutions only in the case of the Fisher cluster model[32] [Eq. (3.6)] and for certain values of r. Even in this case the calculations are lengthy, and therefore we summarize here the main points only, giving more details in the Appendix. But it should be kept in mind that in the general case standard methods may be used to calculate approximate eigenfunctions and eigenvalues of Eq. (2.33) numerically to any desired degree of accuracy.[57]

A. Formulation of eigenvalue problem; rigorous bounds and symmetry relations

In all of the following, we refer to the case where the system is in thermal equilibrium specified by some external parameter $e + \Delta e$ (Δe may be inhomogeneous!), which is changed to e at $t = 0$ [cf. Eq. (2.9) and corresponding remarks]. (For example, a small magnetic field is switched off or diminished at time $t = 0$.) Then we have to solve Eq. (2.33) with $n_l^f \equiv n_l^{(e)}$ and the initial condition $\bar{n}_l(t=0) = n_l^{(e+\Delta e)}$. The convergence of the expansions Eqs. (3.1) and (3.4) and the structure of the coefficients Eq. (3.3), which are insensitive to changes of ϵ or h for $l \to 0$, suggest the following boundary conditions for $g_l(\vec{x}, t)$:

$$\lim_{l\to 0} g_l(\vec{x}, t) \equiv 0 , \qquad \lim_{l\to\infty} n_l g_l^2(\vec{x}, t) \equiv 0 . \tag{5.1}$$

As discussed in Sec. IV, [Eq. (4.10), etc.] the

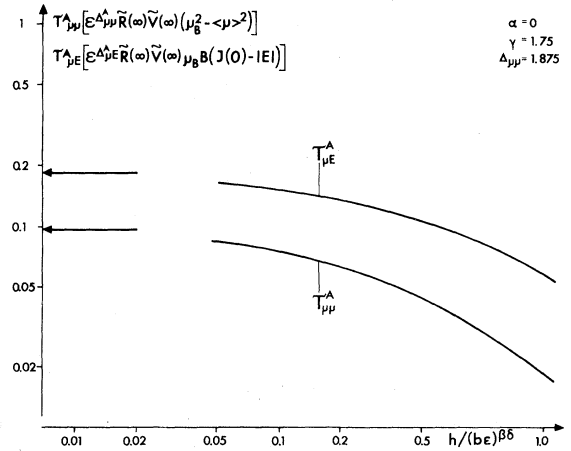

FIG. 6. Normalized scaled autocorrelation times plotted vs scaled field for $d = 2$ and $r = 1$. In this case τ^A_{EE} is finite at the critical point, and thus not shown. Full arrows indicate the values of $\tau^A_{\mu\mu}$ and $\tau^A_{\mu E}$ at $h = 0$.

last term in Eq. (2.33) is negligible in the critical region, and thus our "Sturm-Liouville problem"[57] is already specified completely. Any $\vec{x}$ dependence (or $\vec{q}$ dependence) enters in this calculation through the initial condition only.

Then the solutions of Eq. (3.30) which satisfy Eq. (5.1) can be represented as

$$g_l(\vec{x}, t) = \sum_{K=0}^{\infty} c_K \Psi_K(l) e^{-\lambda_K t} , \tag{5.2a}$$

with

$$L\Psi_K(l) \equiv \frac{d}{dl}\left(R_l n_l \frac{d}{dl}\Psi_K(l)\right) = -\lambda_K n_l \Psi_K(l) , \tag{5.2b}$$

i.e., the $\Psi_K(l)$ are the eigenfunctions and the λ_K the eigenvalues of the "Liouville operator" L. Using a vector notation $|\Psi_K\rangle$ corresponding to $\Psi_K(l)$, we define a scalar product

$$\langle f_1 | f_2 \rangle \equiv \int_0^{\infty} n_l f_1(l) f_2(l)\, dl = \langle f_2 | f_1 \rangle , \tag{5.3}$$

and then the orthonormality of the eigenfunctions is expressed as

$$\langle \Psi_K | \Psi_{K'} \rangle = \delta_{KK'} . \tag{5.4}$$

Although we use a discrete notation in Eqs. (5.2a) and (5.4), we do not exclude the possibility that the eigenvalue spectrum is continuous (this happens in our case right at the critical point; see Sec. V C). Because of physical reasons we assume that there always exist solutions to this eigenvalue problem [Eqs. (5.1)–(5.4)]. Rigorous mathematical-existence proofs have been given for a very restricted case only, namely, where Eq. (3.6) with $\epsilon = 0$, $h = 0$ is used in Eq. (5.2b).[58] We also assume, as usual, the set of eigenfunctions to be complete.

Expanding now the initial condition

$$g_l(\vec{x}, 0) = [n_l^{(e+\Delta e)}(\vec{x}) - n_l^{(e)}]/n_l^{(e)} \equiv f_l(\vec{q})$$

in terms of the complete set $\{|\Psi_K\rangle\}$,

$$f|(\vec{q})\rangle = \sum_K |\Psi_K\rangle \langle \Psi_K| f(\vec{q})\rangle , \tag{5.5a}$$

we find the coefficients c_K in Eq. (5.2a),

$$c_K = \langle \Psi_K | f(\vec{q})\rangle . \tag{5.5b}$$

Then the time-dependent change in magnetization

$$2\mu_B \Delta M^{\Delta e}(\vec{q}, t) = \langle \mu_q(t)\rangle - \langle \mu_q(0)\rangle$$

and energy

$$k_B T_c \Delta E^{\Delta e}(\vec{q}, t) = E_q(t) - E_q(0)$$

is given by [cf. Eqs. (5.3) and (3.4)]

$$\Delta M(\vec{q}, t) = \sum_{K=0}^{\infty} \langle l | \Psi_K\rangle \langle \Psi_K | f(\vec{q})\rangle e^{-\lambda_K t} , \tag{5.6a}$$

$$\Delta E(\vec{q}, t) = \sum_{K=0}^{\infty} \langle l^{1/\beta\delta} | \Psi_K\rangle \langle \Psi_K | f(\vec{q})\rangle e^{-\lambda_K t} , \tag{5.6b}$$

restricting our attention to the case of the Fisher cluster model.[59] In the case $\vec{q} = 0$, we find from Eqs. (3.9) and (3.10) and $p_{\mu E}(l, 0) = l^{1/\beta\delta}$, $p_{E\mu}(l, 0) = l$ that for linear response $\delta T \to 0$,

$$\Phi_{\mu}^{\delta T}(0, t) = \frac{\Delta M^{\delta T}(0, t)}{\Delta M^{\delta T}(0, 0)} = \frac{\sum_{K=1}^{\infty} \langle l | \Psi_K\rangle \langle \Psi_K | l^{1/\beta\delta}\rangle e^{-\lambda_K t}}{\sum_{K=1}^{\infty} \langle l | \Psi_K\rangle \langle \Psi_K | l^{1/\beta\delta}\rangle} , \tag{5.7a}$$

while

$$\Phi_{E}^{\delta H}(0, t) = \frac{\Delta E^{\delta H}(0, t)}{\Delta E^{\delta H}(0, t)} = \frac{\sum_{K=1}^{\infty} \langle l^{1/\beta\delta} | \Psi_K\rangle \langle \Psi_K | l\rangle e^{-\lambda_K T}}{\sum_{K=1}^{\infty} \langle l^{1/\beta\delta} | \Psi_K\rangle \langle \Psi_K | l\rangle} , \tag{5.7b}$$

which implies the desired symmetry relation (see Sec. IV A)

$$\Phi_{E}^{\delta H}(0, t) = \Phi_{\mu}^{\delta T}(0, t) . \tag{5.7c}$$

Next we mention an upper bound $\bar{\lambda}_0$ for the lowest eigenvalue λ_0, which follows from a variational principle.[57] Defining a functional $\mathcal{F}[f]$ by

$$\mathcal{F}[f] \equiv \langle R | f'^2\rangle \equiv \int_0^{\infty} n_l R_l \left(\frac{df}{dl}\right)^2 dl , \tag{5.8a}$$

it can be shown that

$$\lambda_0 \le \mathcal{F}[f]/\langle f | f\rangle \equiv \bar{\lambda}_0 \tag{5.8b}$$

holds for any function f satisfying the boundary conditions Eq. (5.1). A suitable trial function f is $f = l^{3+1/\delta-r}$, since it satisfies the boundary conditions, and is a solution of

$$Lf = R_l n_l \left[\frac{d^2 f}{dl^2} - \left(\frac{2 + 1/\delta - r}{l} + h + \frac{b}{\beta\delta}\,\epsilon l^{1/\beta\delta - 1}\right)\frac{df}{dl}\right] = -\lambda n_l f \tag{5.9}$$

asymptotically for $l \to 0$. The resulting upper bound can be found analytically both at the coexistence curve and at the critical isotherm,

$$\bar{\lambda}_0 = \left(1 + \frac{1}{\delta} + \frac{\Delta_{\mu\mu}}{\beta\delta}\right)\frac{\Gamma(2 + 1/\delta + \Delta_{\mu\mu}/\beta\delta)}{\Gamma(1 + 1/\delta + 2\Delta_{\mu\mu}/\beta\delta)} \times \tilde{R}(\infty) h^{\Delta_{\mu\mu}/\beta\delta} , \quad \epsilon = 0 \tag{5.10a}$$

$$\bar{\lambda}_0 = \frac{2 - \alpha + \Delta_{\mu\mu}}{(\beta\delta)^2}\,\frac{\Gamma(3 - \alpha + \Delta_{\mu\mu})}{\Gamma(2 - \alpha + 2\Delta_{\mu\mu})} \times \tilde{R}(\infty)(b\epsilon)^{\Delta_{\mu\mu}} , \quad h = 0 \tag{5.10b}$$

where $\Delta_{\mu\mu} = (2 - r)\beta\delta$ as before [Eq. (4.18a)].

B. Explicit eigenvalue spectra and eigenfunctions in special cases

Consider first the case of the critical isotherm $\epsilon = 0$ and $r = 1$ [note that the numerical estimates of Refs. 8 and 23 together with Eq. (4.18a) imply that $r = 0.93 \pm 0.03$ for $d = 2$ and $r = 1.14 \pm 0.03$ for $d = 3$; thus this choice approximates both $d = 2$ and $d = 3$ and would be strictly correct at about $d = 2.5$ dimensions]. Then Eq. (5.9) is rewritten in terms of $z = hl$ as

$$z\frac{d^2\Psi_K}{dz^2}-\left(1+\frac{1}{\delta}+z\right)\frac{d\Psi_K}{dz}+\Psi_K\frac{\lambda_K}{\tilde{R}(\infty)h}=0\,, \tag{5.11a}$$

and the eigenfunctions and eigenvalues of this hypergeometric differential equation[60] are (cf. Appendix)

$$\lambda_K=\tilde{R}(\infty)h(K+2+1/\delta)\,,\quad K=0,1,2,\ldots \tag{5.11b}$$

$$\Psi_K(z)=z^{2+1/\delta}L_K^{2+1/\delta}(z)\,, \tag{5.11c}$$

where the $L_K^{(\nu)}$ are generalized Laguerre polynomials.[60] With these results, Eqs. (5.6) can be evaluated explicitly (see the Appendix) and one obtains

$$\Phi_\mu^{\delta H}(0,t)=\frac{e^{-t\tilde{R}(\infty)h(2+1/\delta)}}{\Gamma(3+1/\delta)\Gamma(1-1/\delta)}\times{}_2F_1\left(1+\frac{1}{\delta},\ 1+\frac{1}{\delta},\ 3+\frac{1}{\delta},\ e^{-t\tilde{R}(\infty)h}\right), \tag{5.12a}$$

$$\Phi_E^{\delta H}(0,t)=\frac{e^{-t\tilde{R}(\infty)h(2+1/\delta)}\Gamma(1+1/\beta\delta)}{\Gamma(3+1/\delta)\Gamma(1/\beta\delta-1/\delta)}\times{}_2F_1\left(1+\frac{1}{\delta},\ 2+\frac{1}{\delta}-\frac{1}{\beta\delta},\ 3+\frac{1}{\delta},\ e^{-t\tilde{R}(\infty)h}\right), \tag{5.12b}$$

$$\Phi_\mu^{\delta T}(0,t)=\frac{e^{-t\tilde{R}(\infty)h(2+1/\delta)}\Gamma(1+1/\beta\delta)}{\Gamma(3+1/\delta)\Gamma(1/\beta\delta-1/\delta)}\times{}_2F_1\left(2+\frac{1}{\delta}-\frac{1}{\beta\delta},\ 1+\frac{1}{\delta},\ 3+\frac{1}{\delta},\ e^{-t\tilde{R}(\infty)h}\right), \tag{5.12c}$$

$$\Phi_E^{\delta T}(0,t)=\frac{e^{-t\tilde{R}(\infty)h(2+1/\delta)}\Gamma^2(1+1/\beta\delta)}{\Gamma(3+1/\delta)\Gamma(2/\beta\delta-1-1/\delta)}\times{}_2F_1\left(2+\frac{1}{\delta}-\frac{1}{\beta\delta},\ 2+\frac{1}{\delta}-\frac{1}{\beta\delta},\ 3+\frac{1}{\delta},\ e^{-t\tilde{R}(\infty)h}\right), \tag{5.12d}$$

where for Eq. (5.12d) we need $\alpha>0$. As an example we show $\Phi_\mu^{\delta H}(0,t)$ and $\Phi_\mu^{\delta T}(0,t)$ in Fig. 7 for the case of $d=2$, where $\beta=\frac{1}{8}$, $\delta=15$. As a check to our general proof Eq. (5.7c), we note the symmetry between $\Phi_\mu^{\delta T}(0,t)$ and $\Phi_E^{\delta H}(0,t)$, since[60] ${}_2F_1(a,b,c,z)={}_2F_1(b,a,c,z)$. Furthermore, we find for the small-t behavior that the region where the initial slowing down shows up [Eq. (2.15)] shrinks to zero in the scaling limit

$$\Phi_\mu^{\delta H}(0,t)=1-[t\tilde{R}(\infty)h]^{1-1/\delta}\frac{\Gamma(1/\delta)}{\Gamma(2-1/\delta)\Gamma^2(1+1/\delta)}+\cdots\,, \tag{5.13a}$$

$$\Phi_\mu^{\delta T}(0,t)=1-[t\tilde{R}(\infty)h]^{1/\beta\delta-1/\delta}\frac{\Gamma(1+1/\beta\delta)}{\Gamma(1+1/\delta)\Gamma(1+1/\beta\delta-1/\delta)\,(1+1/\delta-1/\beta\delta)}+\cdots\,, \tag{5.13b}$$

and

$$\Phi_E^{\delta T}(0,t)=1+[t\tilde{R}(\infty)h]^{2/\beta\delta-1-1/\delta}\frac{\Gamma(1+1/\delta-2/\beta\delta)\Gamma^2(1+1/\beta\delta)}{\Gamma(2/\beta\delta-1-1/\delta)\Gamma^2(2+1/\delta-1/\beta\delta)}+\cdots\,, \tag{5.13c}$$

while for large t the decay is exponential since[60] ${}_2F_1(a,b,c,0)=1$. The dynamic susceptibilities can all be expressed in the form

$$\frac{\chi_{BC}(\omega)}{\chi_{BC}}=\sum_{K=0}^{\infty}C_K^{BC}\frac{1}{i\omega+\lambda_K}\,, \tag{5.14}$$

the expansion coefficients C_K^{BC} being derived in the Appendix. Instead of the simple form Eq. (2.13), we have a sum of Lorentzians; Eq. (5.14) shows the physical significance of the eigenfrequencies [Eq. (5.11b)].

As a second example, we consider the coexistence curve and $\Delta_{\mu\mu}=1$, i.e., we use the cluster reaction theory with mean-field exponents. Rewriting Eq. (5.9) using $z=b\epsilon l^{1/\beta\delta}$, we get the same differential equation as before [Eq. (5.11a)], only the constants are different,

$$z\frac{d^2\Psi_K}{dz^2}-(2-\alpha+z)\frac{d\Psi_K}{dz}+\Psi_K\frac{\lambda_K(\beta\delta)^2}{\tilde{R}(\infty)b\epsilon}=0\,, \tag{5.15a}$$

and thus

$$\lambda_K=[\tilde{R}(\infty)b/(\beta\delta)^2]\,\epsilon(K+3-\alpha)\,,\quad K=0,1,2,\ldots \tag{5.15b}$$

$$\Psi_K(z)=z^{3-\alpha}L_K^{(3-\alpha)}(z)\,. \tag{5.15c}$$

In full analogy to the previous treatment, we obtain

$$\Phi_\mu^{\delta H}(0,t)\equiv\frac{\exp(-t[\tilde{R}(\infty)b/(\beta\delta)^2]\epsilon(3-\alpha))}{\Gamma(\gamma)\Gamma(4-\alpha)}\Gamma^2(3-\alpha-\beta)\,{}_2F_1(3-\alpha-\beta\delta,\,3-\alpha-\beta\delta,\,4-\alpha,\,e^{-bt\tilde{R}(\infty)\epsilon/(\beta\delta)^2})\,, \tag{5.16}$$

the other functions being listed in the Appendix, and also Eq. (5.14) is found once more.

These results [Eqs. (5.12) and (5.16)] illustrate explicitly the universality and dynamic scaling struc-

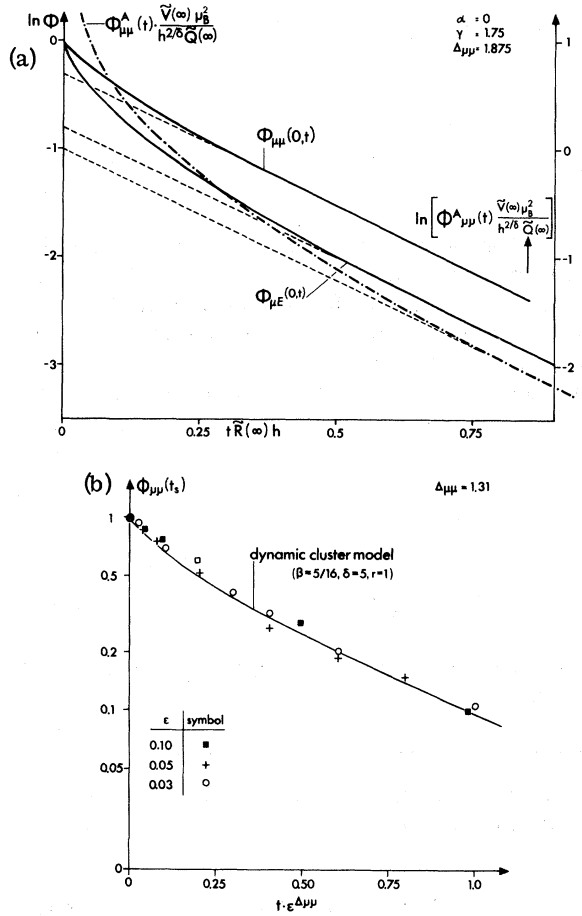

FIG. 7. (a) Relaxation functions $\Phi_{\mu\mu}(0,t)$ and $\Phi_{\mu E}(0,t)$ plotted vs scaled time at the critical isotherm, $d=2$ and $r=1$. The broken striaght lines indicate the asymptotic exponential behavior. The dash-dotted curves represent the (normalized) scaling function for the autocorrelation function $\Phi^A_{\mu\mu}(0,t)$. (b) $\Phi_{\mu\mu}(0,t)$ plotted vs scaled time $t_s = t\epsilon^{\Delta_{\mu\mu}}$ for $d=3$ and $r=1$. The points show Monte Carlo results, obtained for a simple cubic $20\times20\times20$ system with periodic boundary conditions, using Eq. (2.4).

ture of the relaxation functions $\Phi_{BC}(0,t)$,

$$\Phi_{BC}(0,t) = \tilde\Phi(t/|\epsilon|^{-\Delta_{\mu\mu}}\tilde\tau(h|\epsilon|^{-\beta\delta})) \xrightarrow[t\to\infty]{} \tilde\varphi^F_{BC} e^{-t/\tau_{\mu\mu}} , \tag{5.17a}$$

$$\Phi_{BC}(0,t) \xrightarrow[t\to 0]{} 1 - \frac{\tilde\varphi^I_{BC} t^{\gamma_{BC}/\Delta_{\mu\mu}} |\epsilon|^{\gamma_{BC}}}{[\tilde\tau(h|\epsilon|^{-\beta\delta})]^{\gamma_{BC}/\Delta_{\mu\mu}}} + \cdots . \tag{5.17b}$$

Here Eq. (5.17b) is related to the fact that all terms of the moment expansion (cf. Sec. II) vanish like $|\epsilon|^{\gamma_{BC}}$ (or $h^{\gamma_{BC}/\beta\delta}$, respectively) and this fact determines the ϵ or h dependence of $\Phi_{BC}(0,t)$ at $t=0$. Of course, any methods based on moment expansions would be very unsatisfactory, since in the scaling limit their radius of convergence is zero. The fact that $\tilde\varphi^F_{BC}$ does not depend on B alone, but on C as well, has the consequence that the generalized Ginzburg-Landau theory[33]

$$\frac{d\langle\mu(t)\rangle}{dt} = \mathcal{F}[\langle\mu(t)\rangle] , \tag{5.18}$$

where the functional $\mathcal{F}[z]$ need not be specified here, is not correct even for $t\to\infty$: The asymptotic decay depends on whether the initial state was produced by a field (switched off at $t=0$) or by temperature. Thus our present results cannot be described by any equation of motion which depends only on the magnetization at one time. Although the dynamics are Markoffian for the probability distribution of the spins [Eq. (2.2)] as well as for the cluster concentrations [Eq. (2.33)], it is basically non-Markoffian for the order parameter!

It is also important to note that the dynamic scaling structure [Eq. (5.17a)] which sometimes is described as "there is only one characteristic frequency $(\tau_{\mu\mu})^{-1}$ in the system," must not be misunderstood to conclude that $\chi(\omega)$ is a simple Lorentzian near T_c [cf. Eq. (5.14)].

Finally, the structure of Eqs. (5.12), (5.16), and (5.17) leads us to suggest the following approximation for $\Phi_{BC}(0,t)$ in the general case:

$$\Phi_{BC}(0,t) = \frac{\Gamma(c_1-b_1)\Gamma(c_1-b_2)}{\Gamma(c_1-b_1-b_2)\Gamma(c_1)} e^{-t\bar\lambda_0} \times {}_2F_1(b_1, b_2, c_1, e^{-t\bar\lambda_0/a_1}) , \tag{5.19}$$

with $a_1 = 1 + (2-\alpha)/\Delta_{\mu\mu}$; $c_1 = 2a_1 + (\gamma_{BC} - 2\beta\delta)/\Delta_{\mu\mu}$; $b_1 = a_1 - \beta\delta/\Delta_{\mu\mu}$ if $B=\mu$ and $b_1 = a_1 - 1/\Delta_{\mu\mu}$ if $B=E$; $b_2 = a_1 - \beta\delta/\Delta_{\mu\mu}$ if $C=\mu$ and $b_2 = a_1 - 1/\Delta_{\mu\mu}$ if $C=E$. Since in the two special cases treated here $\lambda_0 = \bar\lambda_0$, Eq. (5.19) is then exact; in the general case it has at least the correct structure of Eq. (5.17). The dashed curves in Fig. 5 are based on Eq. (5.19), and compared to the numerical evaluation of the (exact) equation (4.17). It is seen that for $\tau_{\mu\mu}$ and $\tau_{\mu E}$ the agreement is satisfactory. If higher precision is desired for $\Phi_{BC}(0,t)$, techniques for the numerical solution of Eq. (5.9) must be used.[57]

C. Local and wave-vector-dependent relaxation functions

The extension of the eigenfunction expansions to the response to local fields is simple, since we only need to insert initial conditions f_l based on Eq. (4.23). In the case where the field δh is switched off at one site at $t=0$, we have

$$f_l = [1/\tilde V(\infty)]\, l^{-1/\delta}\delta h , \tag{5.20a}$$

while in the case of a temperature change we have

$$f_l = [1/\tilde V(\infty)]\, l^{1/\beta\delta - 1 - 1/\delta} b(\delta T/T_c) . \tag{5.20b}$$

Subsequently we get at the critical isotherm with $r=1$ (see the Appendix),

$$\Phi^A_{\mu\mu}(t) = \frac{h^{2/\delta}\tilde{Q}(\infty)}{\tilde{V}(\infty)(\mu_B^2 - \langle\mu\rangle^2)} \frac{\Gamma(1-1/\delta)}{\Gamma(3+1/\delta)} \exp\left[-\tilde{R}(\infty)ht\left(2+\frac{1}{\delta}\right)\right] {}_2F_1\left(2+\frac{2}{\delta}, 1+\frac{1}{\delta}, 3+\frac{1}{\delta}, e^{-\tilde{R}(\infty)ht}\right), \tag{5.21a}$$

while at the coexistence curve for $d = 4^-$ the result is

$$\Phi^A_{\mu\mu}(t) = \frac{|\epsilon|^{2\beta}\tilde{Q}(\infty)}{\tilde{V}(\infty)(\mu_B^2 - \langle\mu\rangle^2)} \frac{\Gamma(\gamma)}{\Gamma(4-\alpha)} \exp\left(-t\frac{\tilde{R}(\infty)\epsilon b}{(\beta\delta)^2}(3-\alpha)\right) {}_2F_1(5-2\alpha-\beta\delta, 3-\alpha-\beta\delta, 4-\alpha, e^{-\tilde{R}(\infty)\epsilon bt/(\beta\delta)^2}). \tag{5.21b}$$

The results for $\Phi^A_{\mu E}(t)$ and $\Phi^A_{EE}(t)$ are similar. Of course, all these expressions are fully consistent with dynamic scaling. Note that the initial slowing down of these autocorrelation functions is not critical, and hence their scaling expressions [Eq. (5.21)] diverge as $t^{-(\Delta_{BC}-\Delta^A_{BC})}$ for small times. An example is given by the dash-dotted curve in Fig. 7. Equation (5.19) can be used also for the autocorrelation function $\Phi^A_{\mu\mu}(t)$, if b_1 is replaced by $b_1^A = b_1 + (2-\alpha)/\Delta_{\mu\mu}$, but b_2 and c_1 remain unchanged.

The calculation of the q-dependent relaxation function is somewhat more involved (see the Appendix). At the coexistence curve for $d = 4^-$ we use Eq. (3.13) to obtain

$$\Phi_{\mu\mu}(\vec{q}, t) = \exp\left(-\frac{\tilde{R}(\infty)b}{(\beta\delta)^2}\epsilon t(3-\alpha)\right) \frac{\varphi_{\mu\mu}(\vec{q}, t)}{\varphi_{\mu\mu}(\vec{q}, 0)}, \tag{5.22a}$$

with

$$\varphi_{\mu\mu}(\vec{q}, t) = \sum_{m=0}^{\infty} \left(\frac{-\vec{q}}{\kappa_0\epsilon^{\nu}}\right)^{2m} \frac{\Gamma(m+\beta\delta+1)}{\Gamma(m+1)} {}_2F_1(3-\alpha-\beta\delta-m, 3-\alpha-\beta\delta, 4-\alpha, e^{-b\tilde{R}(\infty)\epsilon t/(\beta\delta)^2}). \tag{5.22b}$$

Of course, the $m = 0$ terms in this expansion gives just the relaxation function $\Phi_{\mu\mu}(0, t)$ obtained previously [Eq. (5.16)]. $\Phi_{\mu E}(\vec{q}, t)$ and $\Phi_{EE}(\vec{q}, t)$ can be obtained similarly.

D. Relaxation at the critical point

If the state to which the system relaxes is the critical point ($h = 0$, $\epsilon = 0$), Eqs. (5.11b) and (5.15a) are replaced by ($r = 1$)

$$l\frac{d^2\Psi_K}{dl^2} - \left(1+\frac{1}{\delta}\right)\frac{d\Psi_K}{dl} + \frac{\lambda_K}{\tilde{R}(\infty)}\Psi_K = 0, \tag{5.23a}$$

which yields Bessel functions

$$\Psi_K \propto l^{1+1/2\delta} J_{2+1/\delta}\{2[\lambda_K l/\tilde{R}(\infty)]^{1/2}\}, \tag{5.23b}$$

the eigenvalue spectrum being continuous ($\lambda_K > 0$). As an example we give the relaxation functions after a switching off of the field,

$$\Phi_{\mu\mu}(0, t) = -\frac{[\tilde{R}(\infty)t]^{-1/\delta}}{(\delta h)^{1/\delta}\Gamma(1-1/\delta)\Gamma(2+1/\delta)} \times {}_2F_1\left(1, \frac{1}{\delta}, 2+\frac{1}{\delta}, -\frac{1}{(\delta h)\tilde{R}(\infty)t}\right), \tag{5.24a}$$

$$\Phi_{E\mu}(0, t) = \frac{[\tilde{R}(\infty)t]^{1/\beta\delta - 1 - 1/\delta}\Gamma(1+1/\beta\delta)}{(\delta h)^{1+1/\delta-1/\beta\delta}\Gamma(1/\beta\delta - 1/\delta)\Gamma(2+1/\delta)} \times {}_2F_1\left(1, 1+\frac{1}{\delta}-\frac{1}{\beta\delta}, 2+\frac{1}{\delta}, -\frac{1}{\delta h\tilde{R}(\infty)t}\right). \tag{5.24b}$$

As expected, the asymptotic decay is nonexponential, since for $t \to \infty$ the ${}_2F_1$ functions in Eq. (5.24) approach unity [for more details on the derivation of Eq. (5.24), see the Appendix]. This exponential decay can also be interpreted in terms of the dynamic scaling hypothesis, which we rewrite at the critical point as

$$\Phi_{\mu\mu}(0, t) = \langle\mu(t)\rangle / \langle\mu(t=0)\rangle = \tilde{\Phi}_{\mu\mu}(t[\langle\mu(t=0)\rangle]^{\Delta_{\mu\mu}/\beta}), \tag{5.25a}$$

$$\Phi_{E\mu}(0, t) = E_c(t)/E_c(0) = \tilde{\Phi}_{E\mu}(t[E_c(t=0)]^{\Delta_{\mu\mu}/1-\alpha}), \tag{5.25b}$$

where E_c is the "critical part" of the energy [$E_c = E - E(t=\infty)$]. It is reasonable to require that the asymptotic decay of $\langle\mu(t)\rangle$ or $E_c(t)$ should be independent of the initial state $\langle\mu(t=0)\rangle$ or $E_c(t=0)$, which gives a $t^{-\beta/\Delta_{\mu\mu}}$ or $t^{-(1-\alpha)/\Delta_{\mu\mu}}$ decay, respectively. Equations (5.23) and (5.24) are a special case where $\Delta_{\mu\mu} = \beta\delta$, since $r = 1$. The same asymptotic power-law behavior is also found from the generalized Ginzburg-Landau theory. Of course, these results are consistent with our previous findings, since Eqs. (5.11b) or (5.15b) imply that the distance between any two consecutive eigenvalues $\lambda_{K+1} - \lambda_K \to 0$ as $h \to 0$ or $\epsilon \to 0$, respectively.

The fact that the change of the field δh shows up in the relaxation functions [Eq. (5.24)], for arbitrarily small δh, shows that linear-response theory breaks down right at T_c. Of course, one can give a simple interpretation for this nonexponential decay in terms of our cluster picture: very large clusters always relax extremely slowly. However, only away from the critical point this does not matter since then clusters with $l \gg l_\xi$ have an ex-

ponentially small concentration, and thus do not contribute to the relaxation of the magnetization, energy, etc., significantly.

VI. RELAXATION FAR FROM THERMAL EQUILIBRIUM

A. Nonlinear relaxation in one-phase region

Here we consider the relaxation after a change of the field (ΔH) or temperature (ΔT), respectively, which is not infinitesimally small. We consider the limit

$$\mu_B \Delta H/k_B T_c \to 0\,, \quad \Delta T/T_c \to 0\,, \tag{6.1a}$$

but

$$\frac{\Delta h}{h}=\frac{\Delta H}{H}=c_1\,,$$

$$\frac{\mu_B \Delta H}{k_B T_c}\left|1-\frac{T}{T_c}\right|^{-\beta\delta}=c_2\,,$$

$$\frac{\Delta T}{T_c}\left(\frac{\mu_B H}{k_B T_c}\right)^{-1/\beta\delta}=c_3\,,$$

$$\frac{\Delta T}{T_c}\left|1-\frac{T}{T_c}\right|^{-1}=c_4\,, \tag{6.1b}$$

where the c_i are constants of order unity. In our previous treatment, the assumption $c_i \to 0$ was invoked, necessary for linear response. From Eq. (2.24) it is seen that the terms nonlinear in g or ΔS are important for small l' only. However, as long as Eq. (6.1a) is valid, we still have the boundary condition $g_l(t) \to 0$ when $l \to 0$, and hence the nonlinear terms make still a negligible contribution; thus Eq. (2.33) remains valid in the scaling limit defined by Eq. (6.1), which only requires that both the initial and the final state be within the critical region.

Straightforward extension of the eigenfunction-expansion method of Sec. V gives, for instance, if the final state is at the critical isotherm and $r=1$,

$$\Phi_\mu^{\Delta H}(0,t)=\frac{e^{-\tilde{R}(\infty)ht(2+1/\delta)}\sum_{m=1}^{\infty}(\Delta h/h)^m\,{}_2F_1(2+1/\delta-m,\,1+1/\delta,\,3+1/\delta,\,e^{-\tilde{R}(\infty)ht})}{\sum_{m=1}^{\infty}(\Delta h/h)^m\,{}_2F_1(2+1/\delta-m,\,1+1/\delta,\,3+1/\delta,\,1)}\,. \tag{6.2}$$

[For the derivation and other functions $\Phi_\mu^{\Delta T}(0,t)$, etc., see the Appendix.] In the linear-response regime $\Delta h/h \ll 1$, the $m=1$ terms in the summations are sufficient, and this yields Eq. (5.12a), of course. Equation (5.2) is consistent with the extension of the dynamic scaling hypothesis to nonequilibrium phenomena

$$\Phi_\mu^{\Delta H}(\vec{q},t)$$

$$=\tilde{\Phi}_\mu^{\Delta H}(h|\epsilon|^{-\beta\delta},\,\Delta h|\epsilon|^{-\beta\delta},\,\vec{q}\,|\epsilon|^{-\nu},\,t|\epsilon|^{\Delta_{\mu\mu}})\,, \tag{6.3}$$

which was suggested on the basis of Monte Carlo results.[15,61]

B. Relaxation across phase-boundary–nucleation processes

If we perform the change ΔH such that $H=H(t<0)+\Delta H<0$, but $H(t<0)\geq 0$ and consider temperatures $T<T_c$, the relaxation corresponds to a discontinuous phase transition across the coexistence curve, where the order parameter changes its sign. Now $g_l \neq 0$ for small l, and our previous treatment becomes invalid.

However, in nucleation theory[12,15,62] it is argued that it is most important to study the initial stages of these processes, where still $g_l \approx 0$. In fact, one considers a (fictitious) steady-state solution of Eq. (2.33) with the boundary condition $g_l \to 0$ when $l \to 0$ and a nonzero cluster current, the "nucleation rate" J.[12] A derivation very similar to Refs. 15 and 62 yields

$$J=\tilde{R}(\infty)\tilde{Q}(\infty)\left[\frac{\pi}{2}\left(1-\frac{1}{\beta\delta}\right)\right]^{1/2}\left(\frac{b}{\beta\delta}\right)^{-(\Delta_{\mu\mu}+\gamma/2+3\delta/2)/(\gamma+\beta-1)}\epsilon^{\Delta_{\mu\mu}+2-\alpha}(-h\epsilon^{-\beta\delta})^{(\Delta_{\mu\mu}+2-\alpha-1/2)/(\gamma+\beta-1)}$$

$$\times\exp\left[-(\gamma+\beta-1)(b/\beta\delta)^{(\gamma+\beta)/(\gamma+\beta-1)}(-h\epsilon^{-\beta\delta})^{-1/(\gamma+\beta-1)}\right]\,, \tag{6.4a}$$

which has the scaling structure

$$J=\epsilon^j \tilde{J}(h\epsilon^{-\beta\delta})\,, \quad j=\Delta_{\mu\mu}+2-\alpha\,. \tag{6.4b}$$

Contrary to the usual situation in nucleation theory,[12] where unknown prefactors make quantitative predictions difficult, we are thus able to express J entirely in terms of (static and dynamic) equilibrium properties, and in particular the scaling power j is determined unambiguously. Since the nucleation rate gives the number of "critical clusters" with size l^* formed per unit time,[12] and the number of clusters is related to the free energy [Eq. (3.1)] which has the exponent $2-\alpha$, the result for j in Eq. (5.4b) has a very simple and physically obvious

interpretation: The same critical slowing down which applies to equilibrium fluctuations also applies to the nucleation rate. Note that Eq. (5.4b) is a consequence of our general formulation [Eqs. (2.33), (2.35), and (3.3)] already, while the specific factors appearing in Eq. (5.4a) are based on the Fisher cluster model[32] [Eq. (3.6)]. Even within the framework of this model, Eq. (5.4a) is valid for $h \to 0$, $-h\epsilon^{-\beta\delta} \gtrsim 0.1$ only. In the limit $h \to 0$, the critical cluster size ultimately becomes very much larger than l_t, and then geometrical relations apply to the cluster properties (cf. Fig. 4) which give asymptotically a relation

$$J \propto \exp[-\text{const}(-h\epsilon^{-\beta\delta})^{-(d-1)}] , \tag{6.5}$$

consistent with conventional nucleation theory. For $d=2$, 3, the exponents of h in the exponential in Eq. (5.4a) are only slightly different from this geometric prediction.

A calculation of the nonequilibrium relaxation function which takes the nonlinearity of Eq. (2.24) approximately into account can be obtained as follows.[15] We disregard any time dependence of the nucleation rate J except for the fact that due to the growth of the nuclei to macroscopic domains the fraction $X(t)$ of the stable phase is increasing, and new nuclei originate only from the unstable phase. Thus the change dX at time t due to $J\,dt'$ nuclei per unit volume originating at time t' is

$$dX = (1-X)V(t,t')J\,dt' , \tag{6.6}$$

where $V(t,t')$ is the volume of a domain at time t, which grew from a critical nucleus originated at time t'. Calculating $V(t,t')$ we note that growth of the volume is caused by reactions within the surface region $\xi V^{1-1/d}$ of the volume only, and we have to use the reaction rate per spin in the region $l \gg l_t$ (see Fig. 4), i.e.,

$$\begin{aligned} dV &= \text{const}(R_l/l)(-h)(\xi V^{1-1/d})\,dt \\ &= \hat{R}(-h)\epsilon^{\Delta_\mu \mu - (\gamma+\beta)}\xi V^{1-1/d}\,dt , \end{aligned} \tag{6.7a}$$

where $\hat{R}$ is a constant of order unity, and obtain

$$\begin{aligned} V(t,t') - V_{l*} &\approx V(t,t') \\ &\approx [d\hat{R}\,\tilde{\xi}\epsilon^{\Delta_\mu \mu - \gamma - \beta - \nu}(-h)(t-t')]^d , \end{aligned} \tag{6.7b}$$

and finally from Eqs. (6.4b), (6.6), and (6.7b), we get the further dynamic scaling results[15,62]

$$X(t) = 1 - \exp[-(t\epsilon^{\Delta_\mu \mu}/\tau_R)^{d+1}] , \tag{6.8a}$$

$$\tau_R = \{[1/(d+1)](d\hat{R}\,\tilde{\xi})^d(-h\epsilon^{-\beta\delta})^d \tilde{J}\}^{-1/(d+1)} . \tag{6.8b}$$

Note that here we derive the scaling properties rather than assuming them; while the functions given by Eq. (6.8) are based on very simplified assumptions [Eqs. (6.6) and (6.7a)], the scaling property is more general and in good agreement with computer experiments.[15,62]

VII. CONCLUSIONS

In this paper we attempted to give a detailed account of the critical dynamics of kinetic Ising models without conservation laws. We list the main stages of our derivation in the following.

(i) The basic statement of our theory is the cluster reaction and diffusion equation (2.33), which we derive from the Ising-model master equation but which can be of more general validity: Although we start from a "contour description" of clusters (Fig. 1), we do not make any specific use of this very restrictive description. In fact, whatever the precise definition of clusters may be, the equation for the cluster concentrations $\bar{n}_l(t)$ will always be Eq. (2.21), α being suitably defined other cluster coordinates. Near T_c, only very large clusters are important, and since we also consider the dynamics on a coarse-grained time scale only, the factorization approximation Eq. (2.22b) seems well justified, in particular since it is still consistent with the exact static correlations (Sec. III), in contrast to the factorization approximation of mean-field theory.

(ii) While our treatment also constitutes the generalization of the static Fisher cluster model[32] to dynamic critical phenomena, we also give a far more general formulation [Eqs. (3.1)–(3.5)]. The various specific cluster models considered previously in the literature are special cases of this description, which is still consistent with the exact static correlation functions and equation of state. The main feature of this description is that two kinds of clusters (n_l and n_l^E) are introduced, for order parameter and energy fluctuations,[63] which happen to coincide in the Fisher model. However, since fluctuations of $\bar{\mu}(x) - \langle \mu \rangle$ of different sign (see Fig. 4) add up in n_l^E but are subtracted in n_l, the Fisher model is not extremely accurate, although it was shown to be a reasonable approximation at not too high temperatures by Monte Carlo methods.[15,43,47] Both the analysis of the cluster percolation problem[43] and the present results on local response (Sec. IV B) clearly show that for higher-dimensional systems any "contour description" of clusters is inadequate and a "fluctuation description" must be used [Fig. 4, Eq. (3.5)].

(iii) Relaxation times consistent with dynamic scaling are found to result from a power-law assumption for the cluster reaction rate [Eq. (2.35)]. The symmetry $\langle \delta\mu \delta\mathcal{K}(t) \rangle = \langle \delta\mathcal{K} \delta\mu(t) \rangle$ implies that the dynamics of the magnetization clusters n_l and the energy clusters n_l^E are closely related, therefore no second power law for R_l^E is needed. While we are able to understand the rough magnitude of the exponent r ($r \approx 1$), we are unable to calculate its precise value. In this respect, our method is clearly inferior to the renormalization-group approach.[8,9,64]

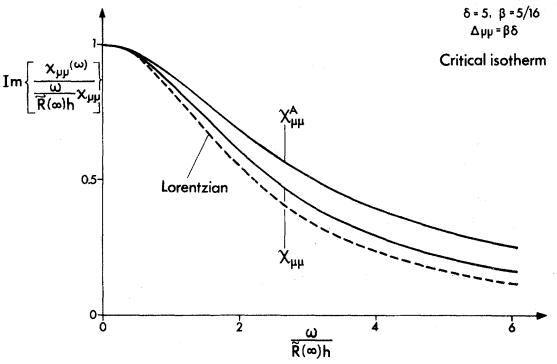

FIG. 8. Normalized frequency-dependent susceptibility $\chi_{\mu\mu}(\omega)$ plotted vs scaled frequency, and the corresponding autocorrelation function $\chi^A_{\mu\mu}(\omega)$. The dashed curve is the Lorentzian with the same curvature at $\omega=0$. This calculation refers to $d=3$ and $r=1$. Note that in experiments one would approximate these functions by Lorentzians with the same half-width instead of the same curvature, making the deviations more difficult to be detected.

This method as well as our present method are based on the same scaling and universality ideas, but our method makes no attempt to calculate critical exponents. On the other hand, we are able to obtain detailed numerical predictions for the linear- and nonlinear-response functions in two and three dimensions, and our method works easily also below T_c and in nonzero magnetic field.

(iv) Specifically, we obtain the exponents $\Delta_{\mu\mu} = \Delta_{\mu E} = \Delta_{EE} = (2-r)\beta\delta$ if $r \leq 1 + 1/\delta$, while otherwise the conventional theory ($\Delta_{\mu\mu} = \gamma$) holds. Here we have to use the behavior of the initial slowing down, which also implies the validity of the inequality[26] $\Delta_{\mu\mu} > \gamma$ within our model, but we do not require $\Delta_{\mu\mu} = \gamma$ as has been inferred[7] from the mode-mode coupling theories. In contrast to the numerical calculations[23,27] our method does not make use of any specific choice for the transition probability, and thus we get "universal" behavior also with respect to dynamic quantities [i.e., *different* systems have the *same* relaxation functions Φ apart from scale factors for the time, for $(T_c - T)/T_c$, for the magnetic field and (perhaps) the wave vector]. It is important to note that Eqs. (2.33), (2.35), and (3.3) may represent far more realistic systems than the kinetic Ising model as well. Of course, phenomenological derivations of these equations for other systems would be desirable.

(v) In Sec. V, we give some exact solutions for the model constituted by Eqs. (2.33), (2.35), and (3.6), which is well defined below T_c and exhibits nonlinear critical phenomena. We obtain the eigenfrequency spectrum and relaxation functions explicitly. We show that the dynamic susceptibility does *not* become a single Lorentzian even for wave vector $\vec{q} \to 0$. In principle, this deviation from Lorentzian line shape should be detectable in experiments. It has been suggested[8] that the critical behavior of uniaxial antiferromagnets such as FeF_2 is well represented by the model considered here, but neither the existing inelastic-neutron-scattering data[65] nor the NMR data[66] allow an accurate analysis of the line shape. Further experiments would be valuable. The numerical magnitude of the predicted deviations from Lorentzian shape are rather small, however (Fig. 8).

(vi) One of the main advantages of our treatment is that the generalization to the relaxation far from equilibrium is fairly straightforward. If both the initial and final state are on the same side of the coexistence curve, the nonlinear relaxation is still represented exactly in terms of the linear Eq. (2.33) in the scaling limit. In the case where the initial and final state are on different sides of the coexistence curve we are able to derive nucleation theory and give the scaling result for the nucleation rate, without any ambiguity with regard to its prefactor. Also, the extension of dynamic scaling to phenomena far from equilibrium[15] is confirmed. Of course, in the same way as the static-cluster model represents a physical "explanation" of static scaling,[32] our treatment represents a physical "explanation" of dynamic scaling. But clearly all these results would be far more useful if they could be extended to cases where the order parameter has more than one component, which may be not straightforward.[49]

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Note added. By numerical evaluation of the differential equation (2.31) it has been shown [R. Kretschmer, D. Stauffer, and K. Binder (unpublished)] that the scaled relaxation functions obtained here are changed only slightly if other values of r are used than are used here in the exactly soluble cases, or if other static droplet models are used, as, e.g., the model of Reatto and Rastelli.[49] In addition, it has been shown there that in the case of extremely nonlinear response (e.g., $\Delta h/h \to -\infty$) critical exponents occur which are different from $\Delta_{\mu\mu}$, and which agree with those found by Z. Racz (unpublished) and M. E. Fisher and Z. Racz (unpublished). The latter fact is seen most simply from Eq. (6.2), where for $t \to \infty$ the numerator approaches $(\Delta h/h)/(1 - \Delta h/h)$, which becomes -1 in

the limit of extremely nonlinear response.

APPENDIX: DERIVATION OF THE EIGENFUNCTION EXPANSIONS

The hypergeometric differential equation (5.11a) is considered first. It has two independent solutions,[60,67]

$$\Psi_k \propto {}_1F_1(-\lambda_k/\tilde{R}(\infty)h, -1-1/\delta, z)\,, \tag{A1a}$$

$$\Psi_k \propto z^{2+1/\delta}\,{}_1F_1(2+1/\delta-\lambda_k/\tilde{R}(\infty)h, 3+1/\delta, z)\,, \tag{A1b}$$

but we have to discard the first because of the boundary condition at $x=0$ [Eq. (5.1)]. We note from the series expansion of the general hypergeometric function

$${}_\nu F_\mu(b_1, b_2, \ldots, b_\nu, c_1, c_2, \ldots, c_\mu, z) = \sum_{m=0}^{\infty} \frac{(b_1)_m (b_2)_m \cdots (b_\nu)_m}{(c_1)_m (c_2)_m \cdots (c_\mu)_m} \frac{z^m}{m!}\,, \tag{A2}$$

where $(b)_m = \Gamma(b+m)/\Gamma(b)$, that ${}_1F_1$ behaves like e^z for large z, thus violating the boundary condition at large z. Similar to the treatment of bound states in quantum mechanics, it must be required that the series Eq. (A2) truncates to a polynomial. This happens if the first argument of ${}_1F_1$ in Eq. (A1b) is a negative integer, and thus one gets Eq. (5.11b). The time-dependent cluster concentrations are then given by

$$g_l(t) = \sum_{k=0}^{\infty} c_k \Psi_k(z) e^{-\lambda_k t} = \sum_{k=0}^{\infty} c_k z^{2+1/\delta} L_k^{2+1/\delta}(z) e^{-\lambda_k t}\,, \tag{A3a}$$

where the expansion coefficients c_k are [$c_k \propto \langle \Psi_k | f(\vec{q}) \rangle$; cf. Eqs. (5.6) and (5.7)]

$$c_k = \frac{k!\,\tilde{Q}(\infty)}{\Gamma(3+1/\delta+k)} \int_0^\infty e^{-z} f_z(\vec{q})\, L_k^{2+1/\delta}(z)\, dz\,. \tag{A3b}$$

We first consider a uniform change of the field,

$$f_z(\vec{q}) = \frac{n_l^{h-\Delta h} - n_l^h}{n_l^h} = e^{\Delta h l} - 1 = e^{(\Delta h/h)z} - 1 = \sum_{m=1}^{\infty} \left(\frac{\Delta h}{h}\right)^m \frac{z^m}{m!}\,. \tag{A4}$$

From the relation[60]

$$\int_0^\infty e^{-zt^c} L_k^{b_1}(z)\, dz = \frac{\Gamma(c+1)}{k!}\,(b_1 - c)_k\,, \tag{A5}$$

it then follows that

$$c_k = \frac{\tilde{Q}(\infty)}{\Gamma(3+1/\delta+k)} \sum_{m=1}^{\infty} \left(\frac{\Delta h}{h}\right)^m \left(2+\frac{1}{\delta}-m\right)_k\,. \tag{A6}$$

If a uniform temperature change is performed, we have similarly

$$f_z(\vec{q}) = \frac{n_l^{T+\Delta T} - n_l^T}{n_l^T} = e^{b(\Delta T/T_c) l^{1/\beta\delta}} - 1 = \exp\left(b\,\frac{\Delta T}{T_c}\, h^{-1/\beta\delta} z^{1/\beta\delta}\right) - 1\,, \tag{A7}$$

and thus

$$c_k = \frac{1}{\Gamma(3+1/\delta+k)} \sum_{m=1}^{\infty} \left(b\,\frac{\Delta T}{T_c}\, h^{-1/\beta\delta}\right)^m \frac{\Gamma(1+m/\beta\delta)}{\Gamma(1+m)} \left(2+\frac{1}{\delta}-\frac{m}{\beta\delta}\right)_k\,. \tag{A8}$$

In order to calculate $\Delta M(0, t)$ and $\Delta E(0, t)$ [cf. Eq. (5.6)], we also need

$$\langle l | \Psi_k \rangle = h^{1/\delta} \tilde{Q}(\infty) \int_0^\infty z e^{-z} L_k^{2+1/\delta}(z)\, dz = h^{1/\delta} \tilde{Q}(\infty)\, \frac{(1+1/\delta)_k}{k!}\,, \tag{A9a}$$

$$\langle l^{1/\beta\delta} | \Psi_k \rangle = h^{1+1/\delta-1/\beta\delta} \tilde{Q}(\infty) b \int_0^\infty z^{1/\beta\delta} e^{-z} L_k^{2+1/\delta}(z)\, dz = h^{(1-\alpha)/\beta\delta} \tilde{Q}(\infty) b\, \frac{(2+1/\delta-1/\beta\delta)_k}{\Gamma(1+1/\beta\delta) k!}\,. \tag{A9b}$$

From Eqs. (A9) and (A6), we then find, rearranging terms,

$$\Delta M(0, t) = \frac{h^{1/\delta} \tilde{Q}(\infty)}{\Gamma(3+1/\delta)} \exp\left[-th\tilde{R}(\infty)\left(2+\frac{1}{\delta}\right)\right] \sum_{m=1}^{\infty} \left(\frac{\Delta h}{h}\right)^m \sum_{k=0}^{\infty} \frac{(2+1/\delta-m)_k (1+1/\delta)_k}{k!(3+1/\delta)_k}\, e^{-th\tilde{R}(\infty)k}\,, \tag{A10a}$$

which leads to Eqs. (6.2) and (5.12a), noticing that the inner sum can be rewritten as a ${}_2F_1$ function [Eq. (A2)]. In the same way one finds that

$$\Phi_B^{\Delta e}(0, t) = e^{-th\tilde{R}(\infty)(2+1/\delta)}\, \varphi_B^{\Delta e}(t)/\varphi_B^{\Delta e}(0)\,, \tag{A10b}$$

with

$$\varphi_\mu^{\Delta T}(t) = \sum_{m=1}^{\infty} \left(b\,\frac{\Delta T}{T_c}\, h^{-1/\beta\delta}\right)^m \frac{\Gamma(1+m/\beta\delta)}{\Gamma(1+m)}\, {}_2F_1\left(2+\frac{1}{\delta}-\frac{m}{\beta\delta},\ 1+\frac{1}{\delta},\ 3+\frac{1}{\delta},\ e^{-th\tilde{R}(\infty)}\right)\,, \tag{A10c}$$

$$\varphi_E^{\Delta H}(t) = \sum_{m=1}^{\infty} \left(\frac{\Delta h}{h}\right)^m {}_2F_1\left(2+\frac{1}{\delta}-m,\ 2+\frac{1}{\delta}-\frac{1}{\beta\delta},\ 3+\frac{1}{\delta},\ e^{-th\tilde{R}(\infty)}\right)\,, \tag{A10d}$$

and

$$\varphi_E^{\Delta T}(t)=\sum_{m=1}^{\infty}\left(b\,\frac{\Delta T}{T_c}h^{-1/\beta\delta}\right)^m\frac{\Gamma(1+m/\beta\delta)}{\Gamma(1+m)}\,{}_2F_1\left(2+\frac{1}{\delta}-\frac{m}{\beta\delta},\,2+\frac{1}{\delta}-\frac{1}{\beta\delta},\,3+\frac{1}{\delta},\,e^{-th\tilde{R}(\infty)}\right). \tag{A10e}$$

From Eqs. (A10b)–(A10e) we get Eqs. (5.12b)–(5.12d), if we take the $m=1$ terms only and use the relation[60,67]

$${}_2F_1(b_1,\,b_2,\,c_1,\,1)=\Gamma(c_1)\Gamma(c_1-b_1-b_2)/\Gamma(c_1-b_1)\Gamma(c_1-b_2)\,. \tag{A11}$$

Treating local changes in linear response, we replace Eqs. (A4) and (A7) by Eq. (5.20) and find

$$c_k=\frac{\delta h}{\tilde{V}(\infty)}\langle l^{-1/\delta}|\Psi_k\rangle=\delta h\,\frac{1}{\tilde{V}(\infty)}h^{1/\delta}\,\frac{\Gamma(1-1/\delta)\,(2+2/\delta)_k}{\Gamma(3+1/\delta+k)}\,, \tag{A12a}$$

and

$$c_k=\frac{\delta T}{T_c}\,\frac{b}{\tilde{V}(\infty)}\langle l^{1/\beta\delta-1-1/\delta}|\Psi_k\rangle=\frac{\delta T}{T_c}\,\frac{b}{\tilde{V}(\infty)}\,h^{1+1/\delta-1/\beta\delta}\,\frac{\Gamma(1/\beta\delta-1/\delta)\,(3+2/\delta-1/\beta\delta)_k}{\Gamma(3+1/\delta+k)}\,. \tag{A12b}$$

We then find Eq. (5.21a) and the analogous expressions

$$\Phi_{\mu E}^A(t)=\frac{h^{1+2/\delta-1/\beta\delta}\tilde{Q}(\infty)b/T_c}{\tilde{V}(\infty)\mu_B D h^{1/\delta}[J(0)-|E|]}\,\frac{\Gamma(1/\beta\delta-1/\delta)}{\Gamma(3+1/\delta)}\exp\left[-\tilde{R}(\infty)ht\left(2+\frac{1}{\delta}\right)\right]$$
$$\times{}_2F_1\left(3+\frac{2}{\delta}-\frac{1}{\beta\delta},\,1+\frac{1}{\delta},\,3+\frac{1}{\delta},\,e^{-\tilde{R}(\infty)ht}\right), \tag{A13a}$$

and

$$\Phi_{EE}^A(t)=\frac{h^{2-2/\beta\delta}\tilde{Q}(\infty)b/T_c}{\tilde{V}(\infty)[\langle\mathcal{H}^2(0,0)\rangle-\langle\mathcal{H}\rangle^2]}\,\frac{\Gamma(1/\beta\delta-1/\delta)}{\Gamma(3+1/\delta)}\exp\left[-\tilde{R}(\infty)ht\left(2+\frac{1}{\delta}\right)\right]$$
$$\times{}_2F_1\left(3+\frac{2}{\delta}-\frac{1}{\beta\delta},\,2+\frac{1}{\delta}-\frac{1}{\beta\delta},\,3+\frac{1}{\delta},\,e^{-\tilde{R}(\infty)ht}\right). \tag{A13b}$$

In order to discuss the behavior of the relaxation functions at large times, one has to take the $k=0$ expansion coefficient [i.e., ${}_2F_1(b_1,\,b_2,\,c_1,\,0)=1$]. For discussing the behavior at short times, it is convenient to use the relation

$${}_2F_1(b_1,\,b_2,\,c_1,\,z)=\frac{\Gamma(c_1)\Gamma(c_1-b_1-b_2)}{\Gamma(c_1-b_1)\Gamma(c_1-b_2)}\,{}_2F_1(b_1,\,b_2,\,b_1+b_2-c_1+1,\,1-z)$$
$$+(1-z)^{c_1-b_1-b_2}\,\frac{\Gamma(c_1)\Gamma(b_1+b_2-c_1)}{\Gamma(b_1)\Gamma(b_2)}\,{}_2F_1(c_1-b_1,\,c_1-b_2,\,c_1-b_1-b_2+1,\,1-z)\,. \tag{A14}$$

From this relation we obtain Eq. (5.13), and the corresponding results for the autocorrelation functions.

The dynamic susceptibilities are obtained from the relaxation functions Eq. (5.12) by termwise integration

$$\int_0^\infty e^{-t\tilde{R}(\infty)h(2+1/\delta)}\,{}_2F_1(b_1,\,b_2,\,c_1,\,e^{-t\tilde{R}(\infty)h})\,e^{-i\omega t}\,dt=\sum_{k=0}^{\infty}\frac{(b_1)_k(b_2)_k}{(c_1)_k k!}\,\frac{1}{i\omega+\tilde{R}(\infty)h(2+1/\delta+k)}$$
$$={}_3F_2\left(b_1,\,b_2,\,2+1/\delta+\frac{i\omega}{\tilde{R}(\infty)h},\,3+\frac{1}{\delta}+\frac{i\omega}{\tilde{R}(\infty)h},\,1\right)\frac{1}{i\omega+(2+1/\delta)\tilde{R}(\infty)h}, \tag{A15}$$

and thus the coefficients c_k^{BC} in Eq. (5.14) are

$$c_k^{\mu\mu}=\frac{[(1+1/\delta)_k]^2\tilde{R}(\infty)h}{k!(3+1/\delta)_k}\Big/\sum_{m=0}^{\infty}\frac{[(1+1/\delta)_m]^2}{m!(3+1/\delta)_m(2+1/\delta+m)}\,, \tag{A16a}$$

$$c_k^{\mu E}=\frac{(1+1/\delta)_k(2+1/\delta-1/\beta\delta)_k\tilde{R}(\infty)h}{k!(3+1/\delta)_k}\Big/\sum_{m=0}^{\infty}\frac{(1+1/\delta)_m(2+1/\delta-1/\beta\delta)_m}{m!(3+1/\delta)_m(2+1/\delta+m)}\,, \tag{A16b}$$

$$c_k^{EE}=\frac{[(2+1/\delta-1/\beta\delta)_k]^2\tilde{R}(\infty)h}{k!(3+1/\delta)_k}\Big/\sum_{m=0}^{\infty}\frac{[(2+1/\delta-1/\beta\delta)_m]^2}{m!(3+1/\delta)_m(2+1/\delta+m)}\,. \tag{A16c}$$

The expansion coefficients of $\chi_{BC}^A(\omega)$ and $\chi_{BC}(\omega)$ at the coexistence curve $d=4^-$ can be obtained analogously, but will not be reproduced here.

Since Eq. (5.15a) has the same structure as Eq. (5.11a), the calculation is rather similar. But note that z has now a different meaning, and hence Eq. (A4) is replaced by

$$f_z(\vec{q}) = e^{\Delta h l} - 1 = \exp\left(\frac{\Delta h}{(b\epsilon)^{\beta\delta}} z^{\beta\delta} - 1\right) = \sum_{m=1}^{\infty} \left(\frac{\Delta h}{(b\epsilon)^{\beta\delta}}\right)^m \frac{z^{m\beta\delta}}{m!} , \tag{A17a}$$

while Eq. (A7a) is replaced by

$$f_z(\vec{q}) = \exp\left(b \frac{\Delta T}{T_c} l^{1/\beta\delta}\right) - 1 = e^{(\Delta T/\epsilon T_c) z} - 1 = \sum_{m=1}^{\infty} \left(\frac{\Delta T}{\epsilon T_c}\right)^m \frac{z^m}{m!} . \tag{A17b}$$

From Eqs. (A2), (A3), (A5), and (A17), we then obtain after some algebra

$$\Phi_B^{\Delta e}(0, t) = \exp\left(-\frac{t\tilde{R}(\infty) b}{(\beta\delta)^2} \epsilon(3 - \alpha)\right) \frac{\varphi_B^{\Delta e}(t)}{\varphi_B^{\Delta e}(0)} , \tag{A18a}$$

with

$$\varphi_\mu^{\Delta H}(t) = \sum_{m=1}^{\infty} \left(\frac{\Delta h}{(b\epsilon)^{\beta\delta}}\right)^m \frac{\Gamma(1 + m\beta\delta)}{\Gamma(1 + m)} \, {}_2F_1\left(1 + \beta, 3 - \alpha - m\beta\delta, 4 - \alpha, e^{-t[\tilde{R}(\infty) b\epsilon/(\beta\delta)^2]}\right) , \tag{A18b}$$

$$\varphi_E^{\Delta H}(t) = \sum_{m=1}^{\infty} \left(\frac{\Delta h}{(b\epsilon)^{\beta\delta}}\right)^m \frac{\Gamma(1 + m\beta\delta)}{\Gamma(1 + m)} \, {}_2F_1\left(2 - \alpha, 3 - \alpha - m\beta\delta, 4 - \alpha, e^{-t[b\tilde{R}(\infty)\epsilon/(\beta\delta)^2]}\right) , \tag{A18c}$$

$$\varphi_\mu^{\Delta T}(t) = \sum_{m=1}^{\infty} \left(\frac{\Delta T}{\epsilon T_c}\right)^m \, {}_2F_1\left(3 - \alpha - m, 1 + \beta, 4 - \alpha, e^{-t[b\tilde{R}(\infty)\epsilon/(\beta\delta)^2]}\right) , \tag{A18d}$$

and

$$\varphi_E^{\Delta T}(t) = \sum_{m=1}^{\infty} \left(\frac{\Delta T}{\epsilon T_c}\right)^m \, {}_2F_1\left(3 - \alpha - m, 2 - \alpha, 4 - \alpha, e^{-t[b\tilde{R}(\infty)\epsilon/(\beta\delta)^2]}\right) . \tag{A18e}$$

The linear response contribution of Eq. (A18a), i.e., the terms with $m > 1$, reduce to Eq. (5.16), if one uses Eq. (A11).

Next, we turn to the wave-vector-dependent linear response for $d = 4^-$. Instead of Eq. (A17) we have to use Eq. (3.13),

$$f_z(\vec{q}) = \delta h l \frac{b\tilde{\chi}}{\tilde{Q}(\infty)} \exp\left[-l^{1/\beta\delta}\left(\frac{\vec{q}}{\kappa_0}\right)^2 b\right] = \frac{\delta h}{(b\epsilon)^{\beta\delta}} \frac{b\tilde{\chi}}{\tilde{Q}(\infty)} \sum_{m=1}^{\infty} \frac{z^{m+\beta\delta}}{m!} \left(\frac{-\vec{q}^2}{\epsilon\kappa_0^2}\right)^m , \tag{A19}$$

which leads to Eq. (5.22).

Then we discuss the eigenfunction expansions in the case of Eq. (5.23a). Using the Hankel integral formula

$$G(z) = \int_0^\infty J_{2+1/\delta}(yz) C(y) y \, dy , \tag{A20}$$

$$C(y) = \int_0^\infty J_{2+1/\delta}(yz) G(z) z \, dz , \tag{A21}$$

we find for $G(z)$ with $z = l^{1/2}$ [cf. Eq. (5.23b)] that

$$G(z) = f_z(\vec{q})/l^{1+1/2\delta} = l^{-1-1/2\delta}(e^{\delta h l} - 1) = z^{-2-1/\delta}(e^{\delta h z^2} - 1) . \tag{A22}$$

From Eqs. (5.2a), (5.23b), and (A20)–(A22), we first obtain

$$g_l(t) = \int_0^\infty dy \, y e^{-\tilde{R}(\infty) t y^2/4} J_{2+1/\delta}(y l^{1/2}) \int_0^\infty J_{2+1/\delta}(yz) z^{-1-1/\delta}(e^{\delta h z^2} - 1) \, dz , \tag{A23a}$$

which by expansion of $e^{\delta h z^2}$ reduces to

$$g_l(t) = -\frac{1}{\Gamma(3 + 1/\delta)} \left(\frac{l}{\tilde{R}(\infty) t}\right)^{2+1/\delta} \sum_{k=0}^{\infty} (\delta h \tilde{R}(\infty) t)^{-k} \, {}_1F_1\left(k + 2 + \frac{1}{\delta}, 3 + \frac{1}{\delta}, -\frac{l}{\tilde{R}(\infty) t}\right) . \tag{A23b}$$

Using

$$\Phi_{\mu\mu}(0, t) = \int_0^\infty l^{-1-1/\delta} g_l(t) \, dl \Big/ \int_0^\infty l^{-1-1/\delta} g_l(0) \, dl$$

and

$$\Phi_{E\mu}(0, t) = \int_0^\infty l^{1/\beta\delta - 2 - 1/\delta} g_l(t) \, dl \Big/ \int_0^\infty l^{1/\beta\delta - 2 - 1/\delta} g_l(0) \, dl ,$$

one obtains Eq. (5.24). Discussing the asymptotic properties of Eq. (5.24) it is convenient to use one

more relation for the ${}_2F_1$ functions[60]

$$ {}_2F_1(b_1, b_2, c_1, z) = (1-z)^{-b_1}\, {}_2F_1(b_1, c_1-b_2, c_1, z/(z-1)) . \quad \text{(A24)} $$

Finally we consider the treatment of the diffusive term in Eq. (2.33) using perturbation theory; this serves as a consistency check to our treatment in Sec. IV, where we argued that this contribution is negligible. Writing

$$ g_l(\vec{x}, t) = G_l(\vec{q}, t) e^{i\vec{q}\cdot\vec{x}} , $$

we have to solve the equation

$$ \frac{\partial G_l(\vec{q}, t)}{\partial t} = L G_l(\vec{q}, t) - a^2 q^2 c (R_l/l^2) G_l(\vec{q}, t) \quad \text{(A25)} $$

or the corresponding eigenvalue problem, writing

$$ G_l(\vec{q}, t) = \Psi_k(\vec{q}) e^{-\lambda_k(\vec{q})t} , $$

$$ L\Psi_k(\vec{q}) - a^2 q^2 c (R_l/l^2)\Psi_k(\vec{q}) n_l + \lambda_k(\vec{q})\Psi_k(\vec{q}) n_l = 0 . \quad \text{(A25a)} $$

Analogously to standard perturbation theory,[57] we get

$$ \lambda_k(\vec{q}) = \lambda_k + q^2 a^2 c \langle \Psi_k | (R_l/l^2)\Psi_k \rangle + O(q^4) , \quad \text{(A26a)} $$

$$ |\Psi_k(\vec{q})\rangle = |\Psi_k\rangle + q^2 a^2 c \times \sum_{k1 \neq k} \frac{|\Psi_{k1}\rangle\langle\Psi_{k1}|(R_l/l^2)\Psi_k\rangle}{\lambda_k - \lambda_{k1}} + O(q^4) , \quad \text{(A26b)} $$

where the λ_k and $|\Psi_k\rangle$ are the (unperturbed) eigenvalues and eigenfunctions of our previous treatment. Evaluation of the matrix elements in Eq. (A16b) shows then that

$$ \lambda_k(\vec{q}) = \lambda_k [1 + \text{const}(a^2 q^2)] , $$

i.e., one obtains only irrelevant corrections in the scaling limit. A similar treatment applies to the last term of Eq. (2.25) also.

As a last point, we derive lower bounds on the lowest eigenvalue λ_0. We consider the problem [cf. Eqs. (5.1) and (5.2b)]

$$ L\Psi_k(l) \equiv \frac{d}{dl}\left(R_l n_l \frac{d}{dl}\Psi_k(l)\right) = -\lambda_k n_l \Psi_k(l) , \quad \text{(A27a)} $$

with the boundary conditions

$$ \Psi_k(0) = 0 , \quad l n_l \Psi_k^2(l) \xrightarrow[l\to\infty]{} 0 . \quad \text{(A27b)} $$

For this eigenvalue problem we can make use of the theorem[57]

$$ \lambda_0 > \min \phi(l) , \quad 0 \le l < \infty ; \qquad \phi(l) = -Ly/n_l y , \quad \text{(A28)} $$

where y is any function which satisfies the boundary conditions [Eq. (A27b)] and does not change sign. A suitable trial function y is

$$ y = l^A (e^{Bl} + Cl^{1/\beta\delta}) , \qquad 0 < A < (\Delta_{\mu\mu} + 2 - \alpha)/\beta\delta , \qquad B = \tfrac{1}{3}h , \quad C = \tfrac{1}{3}b\epsilon , \quad \text{(A29)} $$

which gives

$$ \phi(l) = \tilde{R}(\infty) l^{r-2} \left[\left((h-B)l + \frac{b\epsilon - C}{\beta\delta} l^{1/\beta\delta} + 3 - r - A + 1/\delta\right) \times \left(A + Bl + \frac{Cl^{1/\beta\delta}}{\beta\delta}\right) - Bl - \frac{C}{(\beta\delta)^2} l^{1/\beta\delta}\right] . \quad \text{(A30)} $$

From Eq. (A30) it is seen that λ_0 is larger than a constant, if either $\epsilon \neq 0$ or $h \neq 0$ (or both), and if $\Delta_{\mu\mu} \le 2$, because then we have $\phi(l) > 0$ and

$$ \lim_{l\to 0} \phi(l) = \lim_{l\to\infty} \phi(l) = +\infty . $$

This consideration shows that the decay is exponential for large times in all cases of interest if the eigenfunctions can be normalized (apart from the decay at T_c where $\epsilon = 0$ and $h = 0$).

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[34]Note that the "proof" for the equality $\Delta_{\mu\mu}=\Delta_{EE}=\Delta_{\mu E}$ derived in Ref. 28 from the homogeneity ansatz Eq. (2.18) requires a further assumption concerning the correction exponents x_ϵ, x_H, i.e.,

$$x_\epsilon/2\beta\delta > (\beta\delta x_H - x_\epsilon)/(\alpha+\Delta_{EE}-\beta\delta x_H),$$

where x_ϵ and x_H are defined by

$$\chi_{EE}\tau_{EE}=\tilde{C}\tilde{\tau}_{EE}\,|\epsilon|^{-\alpha-\Delta_{EE}}(1+C_1|\epsilon|^{x_\epsilon}+\cdots), \quad H=0$$

$$\chi_{EE}\tau_{EE}=\hat{C}\hat{\tau}_{EE}H^{-(\alpha+\Delta_{EE})/\beta\delta}(1+\hat{C}_1H^{x_H}+\cdots), \quad \epsilon=0.$$

According to renormalization-group treatments [F. J. Wegner, Phys. Rev. B 5, 4529 (1972); and private communication] one has $x_\epsilon=\beta\delta x_H$ and then the above inequality is always fulfilled.

[35]In some special cases this result was found by Stoll *et al.*, Ref. 27.

[36]This rigorous characterization of "clusters" is particularly valuable for discussion low-temperature properties of the Ising model; see, e.g., G. Gallavotti, A. Martin Löf, and F. Miracle Solé, in *Battelle Seattle 1971 Recontres* (Springer, Berlin, 1971).

[37]For large enough l, l should be simply this excess number. The precise meaning of l will be specified in Sec. III. The "contour picture" is a special case where the excess number equals the total number of reversed spins. In general, the total number V_l of spins in a cluster (cluster volume) may exceed l.

[38]This fact can be seen as well by putting $\Delta S_{l,l'}(t)\approx 0$, $\Delta C_{l,l'}(t)=g_{l'}(t)$, and calculating the contribution of this term by perturbation theory (see Sec. V and the Appendix). The higher-order terms neglected in the expansion Eq. (2.27) would also yield such irrelevant correction terms.

[39]Equation (2.21) remains valid in the general case, if we associate a mass $m_i=1$ only with the l excess spins.

[40]The D_l defined in Ref. 11 is denoted by R_l in the present paper to distinguish between cluster reactions (i.e., diffusion in space $\{l\}$) and cluster diffusion (in space $\{\vec{x}\}$). In Ref. 15 an extra factor $\tilde{b}(\vec{l})^2$ was used in this reaction rate in order to have the same notation as in the related work of C. S. Kiang *et al.* [J. Atmosph. Sci. 28, 1112 (1971)].

[41]Of course, the asymptotic l dependence of the reaction rate could be more complicated, containing, e.g., $\ln l$ factors, which would give rise to $\ln(1-T/T_c)$ corrections to the dominating critical behavior.

[42]For an introduction to the general background of this problem, see J. W. Essam, in Ref. 13, Vol. 2.

[43]H. Müller-Krumbhaar, Phys. Lett. A 48, 459 (1974). According to Monte Carlo simulations described in this reference the percolation temperature T_p is even slightly below T_c in three dimensions.

[44]Again we assume that a cluster definition can be found where no extra $\ln l$ factors occur, as in Eq. (2.32) (Ref. 41).

[45]In order that one picks up the large l contribution of $c(l,\epsilon,hl)$, K may be any integer ≥ 2.

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[47]The Monte Carlo counting of clusters was made using the contour definition of clusters only. However, since these calculations were made for ϵ not so small, where $\langle\mu\rangle \gtrsim 0.5$, both the contour picture and the fluctuation picture are essentially equivalent (the coarse graining has to be made over a distance smaller than ξ, which then is also a few lattice spacings only).

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49 (1972)], where also some indirect evidence for Eq. (3.5b) is given. In this paper the "contour picture" was denoted as "Fisher picture" and the "fluctuation picture" as "Kadanoff picture" of clusters.

[51]In the case of the Fisher cluster model the relation $n_l^E = b n_l$ of course implies that $p_{\mu\mu} = p_{E\mu}$ and $p_{\mu E} = p_{EE}$.

[52]In these equations we disregard any time dependence of the background terms $-\partial F_0/\partial H$ and $\partial(F_0/k_BT)/\partial(1/k_BT)$. Of course, this is strictly true within the framework of the "contour interpretation," where $-\partial F_0/\partial H = 2\mu_B$ in Eq. (4.1a), and $\partial(F_0/k_BT)/\partial(1/k_BT) = -\frac{1}{2}J(0)$, $J(\vec{k})$ being the Fourier transform of the exchange. Within the framework of the "fluctuation interpretation," however, we must assume that the dominant singularities are due to the fluctuation contributions, which is correct for dimensionality $d < 4$ and finite range of interaction. Our whole treatment is restricted to this case.

[53]In our preliminary communication (Ref. 11), $p_{\mu\mu}(l, 0, \omega)$ was denoted by $g_l^{\mu}(\omega)$, etc. We do not use this notation here to avoid confusion with the $g_l(t)$ of Eq. (2.28).

[54]Within the framework of the Fisher cluster model, where $p_{\mu\mu} = p_{E\mu}$, this relation can be proven very simply; see Sec. V. In the general case it follows from our basic model, Eqs. (2.1)–(2.3).

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[59]In the general case, Eq. (5.2b) has to be supplemented by $L_E \Psi_k^E(l) = -\lambda_k^E n_l^E \Psi_k^E(l)$ and a further type of scalar product must be defined, $\langle f_1 | f_2 \rangle_E = \int_0^{\infty} n_l^E f_1(l) f_2(l)\, dl$, and Eq. (5.6b) is appropriately changed. However, the symmetry relation Eq. (4.8) then requires that $\Psi_k^E = \Psi_k$ and $\lambda_k^E = \lambda_k$. This is not surprising, of course, since we could reformulate the cluster-reaction theory in terms of the $c(l, \epsilon, hl)$ of Eq. (3.1) instead of using the two derivatives n_l and n_l^E.

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