Critical dynamics of random-field Ising systems with conserved order parameter

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(Received 18 May 1987)

Motivated by recent experiments on critical binary fluid mixtures in porous media, the dynamics of random-field Ising systems with conserved order parameter are considered. In the one-phase regime a new dynamic crossover length l_x is found: for $q \ll q_x = 2\pi/l_x$ the temporal decay of $S(q,t)$ is well approximated by a single exponential whose decay rate is determined by diffusive dynamics, while for $q \gtrsim q_x$ the temporal decay has a strongly nonexponential component, reflecting the activated dynamics of this system. As the ordering transition is approached, this dynamic length l_x diverges as $l_x \sim \exp(c\xi^\psi)$, where ξ is the static correlation length, and the activation free-energy barriers are of order $cT\xi^\psi$ (T being the temperature and c a nonuniversal constant).

A consistent picture of the dynamic critical behavior of random-field Ising systems has recently been put forward by Villain[1] and Fisher.[2] In these systems the critical point is found to be governed by a zero-temperature renormalization-group (RG) fixed point. This results in a new type of *activated* dynamic scaling,[2] different from the conventional dynamic scaling[3] found at critical points governed by finite-temperature RG fixed points. In conventional dynamic scaling,[3] the characteristic time scale τ diverges as the critical point is approached as a power of the correlation length ξ, i.e., $\tau \sim \xi^z$. For activated dynamic scaling,[2] free-energy barriers of height of order ξ^ψ, $\psi > 0$, and thus large compared to the temperature T have to be crossed to relax fluctuations on length scale ξ. This results in a relaxation time that diverges exponentially with ξ, i.e., $(\ln\tau) \sim \xi^\psi$.

However, only dynamics in the absence of conservation laws were explicitly treated by Villain[1] and Fisher.[2] Such dynamics, i.e., a random-field model A in the classification of Hohenberg and Halperin,[3] should be appropriate for dilute Ising antiferromagnets in a uniform field, which are random-field Ising systems that have attracted much experimental study.[4] Binary fluid mixtures in random porous media are another class of random-field Ising systems of experimental interest,[5,6] whose dynamics are constrained by order-parameter conservation. Recent light-scattering experiments by Dierker and Wiltzius[6] probing relaxation in such a system prompted the present theoretical study of the dynamics of random-field Ising systems with order-parameter conservation, i.e., model B.[3] I find that in the one-phase regime when the random-field is strong, the conservation law causes a qualitative change [e.g., in $S(q,t)$] from the equilibrium dynamics without the conservation law at momenta q satisfying $q\xi < 1$, where ξ is the static correlation length. There should be a crossover from nonexponential, activated dynamics at $q\xi = O(1)$ to standard diffusive exponential relaxation for $q \ll q_x$. The crossover momentum q_x is found to vanish exponentially with ξ as ξ diverges. For $q_x \lesssim q \ll \xi^{-1}$, $S(q,t)$ is found to consist of a sum of a diffusive part, whose temporal decay is well approximated by a single exponential, and an activated, strongly nonexponential part. The former part dominates for $q \ll q_x$, while the latter dominates for $q\xi = O(1)$. This picture is quite consistent with the behavior seen by Dierker and Wiltzius[6] near the phase-separation point of a critical mixture of lutidine and water in porous Vycor glass.

In many physical random-field systems there are also other crossovers occurring as ξ increases, and the full picture can be very complicated. For binary fluids in random porous media there is a crossover from a regime where momentum conservation is playing a role in the dynamics (a model H regime[3]) for ξ smaller than a length presumably of order the typical pore size, to a regime for larger ξ where hydrodynamic damping due to the random medium completely removes momentum conservation at length scales of order ξ.[7] If the random fields generated by the random medium are sufficiently weak, there will be a subsequent crossover from a regime of weak random field but strongly random geometry to a strong random-field regime as ξ increases further.[2,8] Otherwise, the random field is already strong for ξ of order the hydrodynamic crossover length. In this paper I focus only on the crossover from activated to diffusive dynamics, considering only length scales larger than the typical pore size and assuming ξ is large enough that we are in the strong random-field regime, where the activation free-energy barriers hindering the system's relaxation are large compared to the temperature T. Only three-dimensional systems will be explicitly considered; generalization to other dimensionalities is straightforward. The behavior discussed here will even occur in two-dimensional random-field systems if ξ gets large enough, though two-dimensional random-field systems do not appear to have a true phase transition at which ξ diverges.[1,2,9]

When the correlation length is ξ, each correlation volume of linear size of order ξ has its own relaxation time due to thermally activated crossing of a free-energy barrier of height of order ξ^ψ.[1,2] The exponent ψ is probably equal to θ, where $-\theta$ is the renormalization-group eigenvalue of the temperature at the zero-temperature fixed point governing random-field Ising critical behav-

ior.[2] For two-dimensional systems $\theta = 1$, while $\theta \simeq 1.5$ for three-dimensional systems.[1,2,8–10] Because of these free-energy barriers, the local relaxation time for a fluctuation that changes the order parameter (the composition of a binary fluid) in a region of linear size ξ at $\mathbf{r}$ is

$$\tau(\mathbf{r}) = t_0 \exp[b(T,\mathbf{r})\xi^\psi/T] , \qquad (1)$$

where t_0 is a "microscopic" time (which may vary as a power of ξ) and the amplitude of the barriers $b(T,\mathbf{r}) > 0$ varies from correlation volume to correlation volume and is broadly distributed so the variations in b are comparable to its typical magnitude $\bar{b}$. In the absence of a conservation law (model A), this variation of b with $\mathbf{r}$ results in a spatially averaged relaxation function $S(q=0,t)$ whose decay occurs over a range in $(\ln t)$ proportional to ξ^ψ.[2] For large ξ, the scaling form expected is[2]

$$S(0,t) \approx S(0,0)f(T\ln(t/t_0)/\bar{b}\xi^\psi) , \qquad (2)$$

where $f(x)$ is a scaling function. Such behavior has been seen, at least qualitatively, in recent Monte Carlo simulations by Ogielski and Huse,[7] who fit their data to the empirical form $f(x) = \exp(-x^3)$.[11] If the random field is weak at microscopic scales and ξ is large compared to the length, l_0, at which the crossover from weak to strong random field regimes occurs, then the typical values of b will be of order T_c/l_0^ψ.[2] If the random field (RF) is strong, on the other hand, b will be of order E_{RF}/a^ψ, where a is a microscopic length and $E_{RF} \gtrsim T$ is a measure of the free-energy barriers at that length scale. For phase-separating binary fluids in random porous media, a is presumably of order the typical pore size, while E_{RF} is the typical free-energy barrier that must be crossed to change the composition in a pore (e.g., by moving a microscopic interface across the pore).

If we now have order parameter conservation (model B), a local fluctuation on length scale ξ will have somewhat slower relaxation, since its dynamics are constrained by the conservation law. The activation barriers that must be crossed are a little higher due to the constraint, but there should be no qualitative change at this length scale. The local relaxation time for such fluctuations is

$$\tau(\mathbf{r}) = t_0 \exp[\tilde{b}(T,\mathbf{r})\xi^\psi/T] , \qquad (3)$$

with $\tilde{b}$ again varying substantially from correlation volume to correlation volume, and taking on somewhat larger values than b (but $\tilde{b}/b$ is of order unity). A fluctuation of momentum q, with $q\xi \simeq 2\pi$ can relax by more-or-less independent, parallel relaxation of regions of linear size of order ξ, even with the conservation law. Thus for ξ large and $q\xi \simeq 2\pi$ we expect the time dependence of $S(q,t)$ to be qualitatively similar for both model A and model B random-field systems, both being qualitatively like the $S(q=0,t)$ with model A dynamics as discussed above and seen in the simulations of Ogielski and Huse.[7] In fact, the conservation law may actually be dangerously irrelevant at the RG fixed point governing random-field critical behavior, in which case the scaling function for $S(q,t)$ at $q\xi = O(1)$ in the limit $\xi \to \infty$, will be the same for models A and B.

For long-wavelength $(q\xi \ll 1)$ order-parameter fluctuations, diffusive transport over distances of order the wavelength must occur in order to relax the fluctuation when we have order-parameter conservation. Thus we must consider both the activated relaxation of individual correlation volumes and diffusion between correlation volumes. The local relaxation times are broadly distributed on a logarithmic scale and this results in some spatial variation in local diffusion rates. However, the distribution of local diffusion rates is not broad on a logarithmic scale, as will be argued below, so we first consider a model in which the diffusion rates are assumed to be spatially uniform. Let us also restrict our attention to lengths large compared to the correlation length, so the individual correlation volumes may be treated as points.

Thus we consider the following model for the dynamic correlations at equilibrium: Each correlation volume i has its own relaxation time $\tau_i = \exp(\tilde{b}_i\xi^\psi/T)$, and the fluctuations of the total order parameter in i have intensity I_i. The variations in $\tilde{b}_i$ and I_i from correlation volume to correlation volume are comparable to their typical magnitudes. As the fluctuations in the order parameter in volume i occur, the conservation law on the order parameter tells us that diffusion into and out of the surrounding domains is also occurring. Let us consider the contribution of correlation volume i to the truncated dynamic correlation function

$$S(q,t) = \langle \hat{\rho}(\mathbf{q},0)\hat{\rho}(-\mathbf{q},t)\rangle - \langle \hat{\rho}(\mathbf{q})\rangle\langle \hat{\rho}(-\mathbf{q})\rangle , \qquad (4)$$

where $\hat{\rho}(\mathbf{q},t)$ is the Fourier transform of the local order parameter (the composition for a binary fluid) at momentum $\mathbf{q}$ and time t. If the diffusion constant is D, then the diffusive relaxation of $\hat{\rho}(\mathbf{q})$ will be at a rate Dq^2. The local relaxation of correlation volume i is at a rate τ_i^{-1}. Thus, in discussing $S(q,t)$ at a particular momentum q, we may divide the correlation volumes into "fast" modes with $\tau_i q^2 D < 1$ and "slow" modes with $\tau_i q^2 D > 1$. Note these "modes" are local relaxational modes within each correlation volume. Assuming we are in the strong random-field regime so that $\ln\tau_i$ is broadly distributed, there are very few modes near the borderline between "fast" and "slow" at $\tau_i q^2 D = 1$. The relaxation of the fast modes' contribution to $S(q,t)$ is diffusion limited, since the local relaxation rate τ_i^{-1} is so fast, while the relaxation of the slow modes' contribution is limited by the local relaxation. Dividing the modes this way we have

$$S(q,t) \approx \frac{1}{V}\left[\underset{\text{slow}}{\sum_i} I_i e^{-t/\tau_i} + e^{-tq^2 D}\underset{\text{fast}}{\sum_i} I_i\right] , \qquad (5)$$

where the sums run over all the slow and fast, respectively, modes in the system, and V is its volume. This expression exhibits the crossover from a simple exponential decay due to diffusive relaxation for small q, where there are essentially no slow modes, to a sum of a diffusive exponential decay and a nonexponential decay due to the broad distribution of relaxation rates of the slow modes at larger q. This crossover occurs at momentum q_x, where

$$|\ln(t_0 q_x^2 D)| \sim \bar{b}\xi^\psi / T \sim \langle \ln(\tau_i / t_0) \rangle \qquad (6)$$

for ξ in the strong random-field regime, where the angular brackets denote an average over correlation volumes i. This introduces a new dynamic crossover length, $l_x = 2\pi / q_x$, that diverges as $l_x \sim \exp(c\xi^\psi)$. The coefficient c is of order $\bar{b}/T$, but since the crossover is gradual, the precise value of c is somewhat arbitrary, depending on the details of the definition of q_x chosen. This crossover has apparently been observed experimentally by Dierker and Wiltzius[6] in a critical mixture of lutidine and water in porous Vycor glass. Note that $q_x \xi$ vanishes in the scaling limit $\xi \to \infty$. This means that the conservation law is acting as a dangerously irrelevant variable and the diffusive regime $q \ll q_x$ does not exist in the scaling limit where $\xi \to \infty$ with $q\xi$ fixed.

Now let us examine the approximation of the uniform diffusion constant D that was made above. Since we are dealing with a random system that has slow dynamics, the local diffusion rate actually varies in both space and time. Thus we should replace D with $D_i(t)$ and divide the modes into "fast" and "slow" at $\tau_i q^2 D_i(t) = 1$. Because of the time- and space-dependence of $D_i(t)$, the Fourier transform of the diffusion away from volume i will not necessarily be of the simple Gaussian form $\exp[-tq^2 D_i(t)]$, but can be of a somewhat different shape. This changes the simple exponential decay in (5) due to the fast modes into a somewhat nonexponential decay, the magnitude of the effect depending on the time dependence and the breadth of the distribution of $D_i(t)$ at times t such that $tq^2 D_i(t) = O(1)$. The local time-dependent diffusion "coefficient" $D_i(t)$ is essentially the ratio of the suitably coarse-grained local-order-parameter conductivity coefficient $\lambda_i(t)$ and susceptibility $\chi_i(t)$. For diffusion away from i, the mean-square distance diffused in time t is $tD_i(t)$; this self-consistently sets the length scale over which the local transport coefficient and susceptibility are averaged to get $\lambda_i(t)$, $\chi_i(t)$, and thus $D_i(t)$. The local susceptibilities in each individual correlation volume i are broadly distributed, with relative variations of order unity. The local transport coefficients, on the other hand, do not have critical divergences for model B dynamics,[3] and are therefore not expected to be highly correlated at any distances larger than a microscopic length a (a pore size for fluids in random porous media). Thus for $\xi \gg a$ the relative variations in $\lambda_i(t)$ should be smaller than those of $\chi_i(t)$ by a factor of $(a/\xi)^{3/2}$. Now, $\chi_i(t)$ is coarse grained over $[tD_i(t)/\xi^2]^{3/2}$ correlation volumes so the relative breadth of its distribution, and thus that of $D_i(t)$ is

$$\frac{\Delta\chi_i(t)}{\chi_i(t)} \approx \frac{\Delta D_i(t)}{D_i(t)} \sim \left[\frac{\xi^2}{tD_i(t)}\right]^{3/4} . \qquad (7)$$

Therefore, as long as $tD_i(t) \gg \xi^2$, the distribution of $D_i(t)$ is quite narrow. This means that for $q\xi \ll 1$ it is a

good approximation to ignore the spatial dependence of $D_i(t)$, instead using $D(t) = \langle D_i(t) \rangle$.

The relative intensities of the activated and diffusive parts of $S(q,t)$ change with q. Let us define a time t_q such that $t_q q^2 D(t_q) = 1$. The fast modes are those that have relaxation times $\tau_i < t_q$. Thus they are the modes that contribute to the time-dependent susceptibility[12] $\chi(t)$ at $t = t_q$. The total intensity of the diffusive part of $S(q,t)$ therefore is

$$\frac{1}{V} \sum_{i \atop \text{fast}} I_i \approx T\chi(t_q) . \qquad (8)$$

Similarly, the total intensity of the slow, activated fluctuations is

$$\frac{1}{V} \sum_{i \atop \text{slow}} I_i \approx T[\chi_0 - \chi(t_q)] , \qquad (9)$$

where χ_0 is the equilibrium ($t \to \infty$) susceptibility. Note $\chi_0 = S(q \to 0, t=0)/T = \chi(t \to \infty)$. For large ξ, the time dependent susceptibility should scale as in (2) as

$$\chi(t) \approx \chi_0 g(T \ln(t/t_0)/\bar{b}\xi^\psi) , \qquad (10)$$

where the scaling function $g(x) \to 1$ for $x \to \infty$. The susceptibility scales as $\chi_0 \sim \xi^{\gamma/\nu}$ for large ξ, so in order to have a sensible limit $[\chi(t)$ finite] for $\xi \to \infty$ at fixed large t we have $g(x) \sim x^{\gamma/\psi\nu}$ for $x \to 0$. Thus for $q \gg q_x$ (actually, $\ln q \gg \ln q_x$) the diffusive part of $S(q,t)$ represents a fraction

$$\frac{\chi(t_q)}{\chi_0} \approx g(x_q) \sim x_q^{\gamma/\psi\nu} = \left[\frac{T\ln(t_q/t_0)}{\bar{b}\xi^\psi}\right]^{\gamma/\psi\nu} \qquad (11)$$

of the total intensity of $S(q)$, where x_q is defined by this equation. Note that for $q = q_x$, $x_q = O(1)$, and the total intensities of the diffusive and activated parts are comparable, while for $q \ll q_x$ the activated part represents only a small fraction of the total intensity.

The time dependence of $D(t)$ for $t \gg t_0$ arises from that of $\chi(t)$, since the conductivity $\lambda(t)$ is set at microscopic scales and for $t \gg t_0$ should be time independent, $\lambda(t) \approx \lambda$, where λ remains finite and nonzero in the limit $\xi \to \infty$.[3] (The usual divergence of λ for binary fluids is suppressed by the random porous medium.) Thus we have $D(t) \approx \lambda/\chi(t)$ and, for $q\xi \ll 1$, we can approximate $S(q,t)$ as

$$S(q,t) \approx \left[\frac{1}{V} \sum_{i \atop \text{slow}} I_i e^{-t/\tau_i}\right] + T\chi(t_q) e^{-tq^2 \lambda/\chi(t)} . \qquad (12)$$

The second term here represents the diffusive part of the decay and has characteristic time t_q. In order to determine how far this term differs from a single exponential, let us expand about this time as

$$e^{-tq^2\lambda/2\chi(t)} = e^{-t/t_q}\left[1 + \frac{1}{2}t(t-t_q)q^2\lambda\left[\frac{1}{\chi^2(t)}\frac{d\chi(t)}{dt}\right]_{t=t_q}\right] + \cdots\right] . \qquad (13)$$

For times of order t_q, the correction to the single exponential decay is of relative order

$$\frac{T}{\bar{b}\xi^{\psi}g(x)}\frac{dg(x)}{dx}\bigg|_{x=x_q} . \tag{14}$$

Near the crossover at $q=q_x$, where the activated part of $S(q,t)$ becomes substantial, $g(x)$ and $dg(x)/dx$ are both of order unity so the relative correction is of order $T/\bar{b}\xi^{\psi}$ and thus small. For $q \ll q_x$, the correction is still smaller. For $q \gg q_x$, the correction is of relative order $T/(\bar{b}\xi^{\psi}x_q)$ and thus becomes substantial for sufficiently high momenta (low x_q). However, for $\bar{b}\xi^{\psi} \gg T$ this does not occur until, by (11), the intensity of the diffusive part is a very small fraction of the total intensity of $S(q)$. Thus for all momenta where the diffusive part represents a substantial fraction of the total intensity of $S(q,t)$ its temporal decay is well approximated by a single exponential with decay time t_q.

The activated part of the decay has a functional form that is undetermined. However, for $q\xi \ll 1$ and $t \gg t_q$, where the activated part dominates, $S(q,t)$ should be q independent. When $q\xi$ becomes of order unity, on the other hand, we have for $t \gg t_q$

$$S(q,t) \approx \frac{1}{V} \sum_{i \atop \text{slow}} I_i(q)e^{-t/\tau_i} , \tag{15}$$

where $I_i(q)$ is the structure factor of the ith mode. There is a tendency for the regions with longer-ranged than average local spatial correlations to have larger than average activation barriers and thus slower relaxation; this results in a stronger momentum dependence for $S(q,t)$ at long times than at short times. Thus, if we approximate with a Lorentzian, the momentum dependence of $S(q,t)$ for $t \gg t_q$ is of the form

$$S(q,t) \sim [1+q^2\xi^2(t)]^{-1} , \tag{16}$$

where the time-dependent correlation length $\xi(t)$ increases with increasing time. The modes with *very* long relaxation times will be rare locally ordered regions[13] of size L, with activation barriers proportional to their cross section L^2. This suggests $\xi^2(t) \sim (\ln t/t_0)$ for the longest times. [The static correlation length is $\xi = \xi(t=0)$.] Arguments similar to those in Refs. 13 ob-

tain a long-time decay of $S(q,t)$ of the form

$$S(q,t) \sim \exp[-(\xi/\xi_0)^{(d\nu-2)/\nu}x^{3/2}] , \tag{17}$$

where, as before, $x = T\ln(t/t_0)/\bar{b}\xi^{\psi}$ is the appropriate scaling variable, and ξ_0 is a length of order $(T/\bar{b})^{1/\psi}$. However, since $d\nu > 2$,[1,2,8−10,14] this form for $S(q,t)$ vanishes in the scaling limit $\xi \rightarrow \infty$ and, for finite ξ, is due to corrections to scaling and holds only at very long times.[15] It is not clear at this point what the long-time form of the decay of $S(q,t)$ is in the scaling limit. This requires a better understanding of the scaling distribution of barriers. If the fraction of the total intensity of $S(q)$ that is due to barriers with height greater than E is $h(E/\bar{b}\xi^{\psi})$, then for $E \gg T$ these modes will not have relaxed significantly at time $t=t_0e^{E/T}$ and we have, for $q\xi \ll 1$

$$S(q,t) \approx S(q \rightarrow 0,0)h(T\ln(t/t_0)/\bar{b}\xi^{\psi}) . \tag{18}$$

Thus the scaling function $h(x)$, which may be identical to the $f(x)$ in Eq. (2) for model A, is just the scaled and weighted integrated density of barriers. If, as one might expect, this density of barriers vanishes exponentially for $x \gg 1$ (large barriers) as $h(x) \sim \exp(-kx^p)$, where k is a number of order unity, then $S(q,t)$ will decay as

$$-\ln S(q,t) \sim [\ln(t/t_0)]^p \tag{19}$$

for $t \gg t_q$.

In conclusion, I have shown how a new dynamic correlation or crossover length l_x which diverges exponentially as a function of the static correlation length ξ enters in random-field Ising systems with conserved order parameter. For momenta $q \ll 1/l_x$, simple diffusive relaxation of $S(q,t)$ occurs, while for higher momenta, extreme variations in local relaxation times leads to more complicated behavior, with $S(q,t)$ being the sum of a diffusive term that is well approximated by a single exponential, and an activated part with a strongly nonexponential temporal decay. This crossover to activated dynamics appears to have been observed in recent light-scattering experiments on a critical mixture of lutidine and water in porous Vycor glass.[6]

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[11]Note this is an empirical form designed to give a reasonable fit to the bulk of the decay, and is not intended to be correct in either limit $x \ll 1$ or $x \gg 1$, although a decay of the form $\exp(-x^p)$ with some exponent p seems likely for $x \gg 1$.

[12]The time-dependent susceptibility $\chi(t) \approx \langle \chi_i(t) \rangle$ being discussed here is the linear response at time t to a field that has

been imposed since time zero. To give a precise definition, the field must have the momentum $q_{max}(t)$ that maximizes this response, since, due to the conservation law, there is no response to a uniform field.

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[15]The contribution of this type of rare region to the dynamic correlations appears to vanish in the scaling limit for any random critical point with $dv > 2$. This result is due to D. S. Fisher (unpublished).