Dynamics of Dilute Magnets above T_c

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Nonperturbative contributions are shown to produce anomalously slow relaxation in dilute ferromagnets (or antiferromagnets), in both the paramagnetic and "Griffiths" phases. In the paramagnetic phase, the pseudoscaling form $C(t) \sim \exp\{-t^{d/(d+z)} f(t/\xi_p^{d+z})\}$ is predicted for the spin autocorrelation function, with ξ_p the correlation length of the *nondilute* system. In the Griffiths phase, $C(t) \sim \exp\{-(A \ln t)^{d/(d-1)}\}$ and $\exp\{-(Bt)^{1/2}\}$ are predicted for Ising and Heisenberg systems, respectively, for $t \gg \xi_p^{d+z}$.

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There has been recent interest[1-3] in the effects of Griffiths singularities[4,5] on the dynamics of random magnetic systems. In this Letter, the dynamics of *dilute* magnetic systems above the critical point are discussed, both in the paramagnetic phase $T \geq T_G$, where the "Griffiths temperature" T_G is the critical temperature of the *pure* (i.e., nondilute) system, and in the "Griffiths phase" $T_c < T < T_G$. Simple, intuitive ideas are employed, based on the existence of large, rare, quasiordered regions which relax very slowly. I find that the conventional picture, based on perturbative renormalization-group treatments near T_c, is inadequate in random systems. Instead I predict, independent of the details of the dynamics, a form of "critical slowing down" for $T \to T_G+$, with a nonexponential decay at T_G of the form $C(t) \sim \exp\{-t^{d/(d+z_p)}\}$, where $C(t)$ is the spin autocorrelation function, d is the spatial dimension, and z_p is the dynamic critical exponent of the *pure* system. Away from T_G, $C(t)$ has for large t and small $|T-T_G|/T_G$ a scaling form,

$$C(t) \sim \exp\{-t^{d/(d+z_p)} f(t/\xi_p^{d+z_p})\},$$

with ξ_p the pure-system correlation length. Note the unconventional form of the scaling variable, involving the exponent $d+z_p$ instead of simply z_p as in conventional dynamic scaling. For $T_c < T < T_G$, the relaxation is even slower than at T_G: Specializing to "model A" dynamics[6] (i.e., no conservation laws) I find, for $t \gg \xi_p^{d+z_p}$, $\ln C(t) \sim -A(\ln t)^{d/(d-1)}$ for Ising systems and $\ln C(t) \sim -(Bt)^{1/2}$ for Heisenberg systems. The amplitudes A and B *diverge* for $T \to T_G-$, while B *vanishes* for $T \to T_c+$.

For simplicity we will consider a dilute ferromagnet, with Hamiltonian

$$H = -\sum_{\langle ij\rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j.$$

The spins $\{\mathbf{S}_i\}$ are n-dimensional vectors. For bond dilution the nearest-neighbor interactions $\{J_{ij}\}$ are independent random variables taking the values J and 0 with probabilities p and $1-p$, respectively. For site dilution, $J_{ij} = Jc_ic_j$, where $c_i = 1$ or 0 with probabilities p and $1-p$, respectively. The spin autocorrelation function is $C(t) = [\langle \mathbf{S}_i(t) \cdot \mathbf{S}_i(0)\rangle]$, where $\langle \cdots \rangle$ and $[\cdots]$ indicate thermal and disorder averages, respectively.

The physical origin of anomalously slow relaxation in the dilute system is the presence, with nonzero probability per site, of arbitrarily large regions (clusters) which have a higher fraction of occupied bonds (or sites) than the system as a whole. These regions need not be, and in general will not be, isolated from the rest of the system. The basic assumption underlying the calculations is that the long-time dynamics is dominated by clusters of a particular shape, size, and mean concentration of occupied bonds or sites. These can be determined variationally to maximize $C(t)$. In a more formal theory, the "critical cluster" for a particular time t would presumably emerge as an "instanton solution" in an appropriate field-theoretic description. In the absence of such a rigorous theory, we will assume that the instanton would have spherical symmetry, i.e., that *compact* clusters dominate at long times. The size L, and the mean concentration p' ($>p$) of occupied bonds or sites which characterize the optimal clusters, will be determined variationally (i.e., by saddle-point methods). For $T < T_G$ clusters with p' such that $T_c(p') > T$ dominate the long-time dynamics, since they correspond to regions of "local order" whose temporal persistence is limited only by finite-size effects. For $T > T_G$, clusters with $p' = 1$ (i.e., clusters of fully occupied bonds or sites) dominate. The generic form for $C(t)$ within this approach is

$$C(t) = \sum_{L,p'} P(L,p') \exp\{-t/\tau(L,p')\}, \tag{1}$$

where $P(L,p')$ is the probability that a given site belongs to a (compact) cluster of size L and mean concentration p', and $\tau(L,p')$ is the corresponding relaxation time. The temperature regimes $T \geq T_G$ and $T_c(p) < T < T_G$ will be discussed separately.

(1) $T \geq T_G$.—For this case clusters with $p' = 1$ dominate, since they are most nearly critical. The cluster

probability is

$$P(L,1)\sim p^{L^d}=\exp\{-cL^d\},\quad c=\ln(1/p), \tag{2}$$

while the relaxation time $\tau(L,1)$ is given by

$$\tau(L,1)=\xi_p^{z_p}f(L/\xi_p) \tag{3}$$

$$\sim\xi_p^{z_p},\quad L\gg\xi_p, \tag{4}$$

$$\sim L^{z_p},\quad L\ll\xi_p. \tag{5}$$

In Eq. (2), an algebraic prefactor has been dropped since it leads to only a power-law prefactor in $C(t)$. Other contributions to the latter prefactor come from corrections to the saddle-point evaluations of the sums on L and p', and (presumably) from fluctuations around the assumed spherical form for the dominant clusters. All such contributions are systematically neglected in what follows: We compute only the leading exponential factor in $C(t)$.

Substituting (2) and (3) into (1) and determining L variationally yields the scaling form

$$\ln C(t)\sim -t^{d/(d+z_p)}g(t/\xi_p^{d+z_p}), \tag{6}$$

with $g(x)$ a scaling function. For $t/\xi_p^{d+z_p}\gg 1$, the dominant clusters have $L\sim\xi_p$, and

$$\ln C(t)\sim -c\xi_p^d-t/\xi_p^{z_p},\quad t\gg\xi_p^{d+z_p}, \tag{7}$$

consistent with (6). In the opposite limit, Eq. (5) is required, and one obtains

$$\ln C(t)\sim -t^{d/d+z_p},\quad t\ll\xi_p^{d+z_p}. \tag{8}$$

In particular, Eq. (8) describes the dynamics at $T=T_{\rm G}$, when $\xi_p=\infty$.

The above results apply to both Ising and vector ($n\geq 2$) spin systems, for general dynamics—the value of n and the details of the dynamics only affect the value of the pure-system dynamical exponent z_p appearing in the general expressions.

(2) $T_c(p)<T<T_{\rm G}$.—In this regime the results depend on both the nature of the spins (Ising or Heisenberg) and the details of the dynamics. The latter we will take to be described by "model A," i.e., we assume no conservation laws.[6] The Ising and Heisenberg models will be separately discussed.

Ising systems: Again we find that $p'=1$ dominates at long times. This is because the relaxation time $\tau(L,p')$, which favors larger p', is exponentially sensitive in L (see below) to changes in p', while the "entropic term" $\ln P(L,p')$, which favors smaller p', is only algebraically sensitive to L. To estimate τ we note that the cluster relaxation is limited by the time taken for coherent reversals of the entire cluster. Since such a reversal requires creating, at an intermediate state, an interface separating regions of opposing magnetization, we have

$$\tau(L,1)\sim\tau_p\exp\{\sigma L^{d-1}\}, \tag{9}$$

where σL^{d-1} is the interface free energy, the factor $1/T$ expected in the Arrhenius form (9) has been absorbed into the surface tension σ, and τ_p is the characteristic relaxation time in the ordered phase of the pure system. Putting (2) and (9) into (1), with p' set equal to 1, yields

$$C(t)\sim\textstyle\sum_L\exp\{-cL^d-(t/\tau_p)e^{-\sigma L^{d-1}}\}.$$

For $t\to\infty$, the sum is dominated by values of L near that which maximizes the summand. This gives

$$L\sim\{(1/\sigma)\ln(t\sigma^{d/(d-1)}/\tau_p)\}^{1/(d-1)} \tag{10}$$

and

$$\ln C(t)\sim -\{(1/\sigma)\ln(t\sigma^{d/(d-1)}/\tau_p)\}^{d/(d-1)}. \tag{11}$$

Of particular interest is the limit $T\to T_{\rm G}-$. In this regime, $\sigma\sim\xi_p^{-(d-1)}$ and $\tau_p\sim\xi_p^{z_p}$, giving $L\sim\xi_p\ln(t/\xi_p^{d+z_p})$ and

$$\ln C(t)\sim -\xi_p^d\{\ln(t/\xi_p^{d+z_p})\}^{d/(d-1)},\qquad t\gg\xi_p^{d+z_p}. \tag{12}$$

Note that (12) is consistent with the general scaling form (6).[7]

Somewhat surprisingly, Eq. (12) contains no hint of the onset of long-range order at $T_c(p)$. Therefore it is not part of the dynamic scaling form that describes the dynamics near $T_c(p)$. This is a consequence of the fact that concentrated clusters ($p'=1$) dominate for all $T>T_c(p)$.[8]

Heisenberg systems: For $n\geq 2$, the relaxation times $\tau(L,p')$ is much shorter than for Ising spins, since there is no free-energy barrier hindering relaxation. Rather, relaxation occurs by "diffusion of the order parameter" over the surface of an n-dimensional sphere, driven by the thermal noise. The change in the cluster magnetization $\mathbf{M}$ due to the thermal noise acting in one "time step" is $\delta\mathbf{M}\sim L^{d/2}$, since the noise on different spins adds incoherently.[9] After t time steps one has $\delta\mathbf{M}(t)\sim L^{d/2}t^{1/2}$. Complete relaxation has occurred when $\delta\mathbf{M}\sim\mathbf{M}\sim L^d$, giving $\tau\sim L^d$. To make this more precise, we recognize that the basic time step is the relaxation time $\tau_r(p')$ of a bulk dilute system with concentration p', and that the length L should be measured in units of the correlation length $\xi_r(p')$ of the bulk dilute system. With these refinements we obtain

$$\tau(L,p')\sim\tau_r(p')[L/\xi_r(p')]^d. \tag{13}$$

Finally, the probability that a cluster of $N\equiv L^d$ bonds (or sites) contains Np' nonzero bonds (or sites) is

$$P(L,p')=\binom{N}{Np'}p^{Np'}(1-p)^{N(1-p')},$$

giving

$$\ln P(L,p') = -L^d\{p'\ln(p'/p) + (1-p')\ln[(1-p')/(1-p)]\} \equiv -L^d f(p'), \tag{14}$$

up to subextensive corrections.

Putting (13) and (14) into (1), and evaluating the sums by steepest descents, yields

$$\ln C(t) \sim -(Bt)^{1/2}, \tag{15}$$

$$B = \min_{p'} \xi_r(p')^d f(p')/\tau_r(p'). \tag{16}$$

Equation (15) agrees with the form previously derived in the $n \to \infty$ limit.[3]

Of particular interest are the limits $T \to T_G -$ and $T \to T_c(p)+$. For $T \to T_G -$, we must have $p' \to 1$ to ensure $T < T_c(p')$. In this limit, $f(p') \to c$, $\xi_r(p') \to \xi_p$, and $\tau_r(p') \to \tau_p \sim \xi_p^{z_p}$. Inserting these into (16) yields $B \sim \xi_p^{d-z_p}$ and

$$\ln C(t) \sim -(t\xi_p^{d-z_p})^{1/2}, \quad t \gg \xi_p^{d+z_p}, \tag{17}$$

consistent with the general scaling form (6).

For $T \to T_c(p)+$, optimization with respect to p' is nontrivial. Since p is close to (but less than) $p_c(T)$ [the inverse function of $T_c(p)$], we anticipate (and verify *a posteriori*) that p' will also be close to (but greater than) $p_c(T)$. For $p'-p$ small, $f(p') \sim (p'-p)^2$. Also $\tau_r \sim \xi_r^{z_r}$ and $\xi_r \sim (p'-p_c)^{-\nu_r}$ in this regime, where z_r and ν_r are the dynamical and correlation length exponents, respectively, for the *random* system. Inserting these forms into (16) yields

$$B \sim \min_{p'} \frac{(p'-p)^2}{(p'-p_c)^{(d-z_r)\nu r}}$$

$$\sim (p_c - p)^{\alpha_r + z_r\nu_r}$$

$$\sim (T - T_c)^{\alpha_r + z_r\nu_r},$$

where α_r is the specific-heat exponent of the random system, and the hyperscaling relation $\alpha_r = 2 - d\nu_r$ has been used. Hence (15) becomes, for $T \to T_c(p)$,

$$\ln C(t) \sim -\{(T-T_c)^{\alpha_r}(t/\xi_r^{z_r})\}^{1/2}. \tag{18}$$

It is interesting to note that, as was found for Ising spins, this nonperturbative contribution to $C(t)$ due to Griffiths singularities is not part of the scaling form near $T_c(p)$: For $t \to \infty$, $\xi_r \to \infty$, with $t/\xi_r^{z_r}$ fixed, the nonperturbative contribution (18) decreases as the exponential of a power with increasing t (since[10] $\alpha_r < 0$), whereas standard dynamic scaling predicts a power-law dependence on t in this regime.[11] Nevertheless, for $t \to \infty$ as fixed $T > T_c(p)$, the nonperturbative contribution (18) eventually dominates (for $t \gg \xi_r^{z_r - \alpha_r/\nu_r}$), since conventional critical dynamics implies $\ln C(t) \sim -t/\tau_r$ in this regime.

The above results for $C(t)$ can be extended[12] to the more general correlation function $C(r,t) = [\langle \mathbf{S}_i(t) \cdot \mathbf{S}_j(0)\rangle]$ with $r \equiv |\mathbf{r}_i - \mathbf{r}_j|$. For times long enough that the length scale $L(t)$ of the dominant spherical clusters satisfies $L(t) \gg r$, the spins at sites i and j will move together during coherent reversals (or rotations) of the cluster. Thus $C(r,t)$ will behave like $C(t)$, i.e., it will be essentially independent of r for $L(t) \gg r$. On longer length scales, elongated clusters, with length of order r and width $L(t) < r$, will dominate,[12] and $C(r,t)$ will decrease rapidly with r. Results for this regime, and for the crossover between the two regimes, will be presented elsewhere.[12]

In summary, the dynamics of dilute magnets exhibit nonexponential relaxation for all $T \le T_G$, i.e., *above* the critical temperature $T_c(p)$ which signals the onset of magnetic long-range order. Near T_G I predict a novel form of dynamic scaling, involving the scaling variable $t/\xi_p^{d+z_p}$. The weakest link in the argument is the assumption that clusters of a particular *shape*, i.e., close to spherical, dominate the dynamics at long times. For Ising systems in the Griffiths phase this is physically plausible since, for clusters of a given volume, the compact clusters are those with the longest relaxation times. For Heisenberg systems the result is less obvious and requires further justification. It is hoped that this will emerge from a steepest-descent calculation, valid for large t, in a more formal theory.

Finally, I note that while the results presented here have been derived specifically for dilute magnets, the Griffiths phase is a more general concept[2,3,5] and the results can be extended, with only minor modifications,[13] to general random magnets.

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[7]Equation (12) was first derived, without the factors of ξ_p and under more restrictive assumptions, by Dhar, Ref. 1, and generalized to Ising spin-glasses by Randeria, Sethna, and Palmer, Ref. 2.

[8]The author thanks D. A. Huse for an illuminating discussion on this point.

[9]Any interaction of the cluster with the rest of the system, occurring at the boundary of the cluster, merely generates additional thermal noise (since the neighboring spins typically relax much faster than those in the cluster) which, being a surface effect, is negligible for large L. For "model B" dynamics, however, corresponding to a conserved order parameter (see Ref. 6), such surface terms presumably dominate the cluster dynamics.

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