PHYSICAL REVIEW B VOLUME 31, NUMBER 3 1 FEBRUARY 1985

Irreversibility in random-field ferromagnets and diluted antiferromagnets

Charles Ro
The James Franck Institute and the Department of Physics, The University of Chicago, Chicago, Illinois 60637

Gary S. Grest and C. M. Soukoulis*
Corporate Research Science Laboratory, Exxon Research and Engineering Company, Annandale, New Jersey 08801

K. Levin
The James Franck Institute, The University of Chicago, Chicago, Illinois 60637
(Received 9 October 1984)

Using the behavior of the free-energy surface, we compute the irreversibility phase diagram, history-dependent magnetizations, and specific heat for three-dimensional diluted antiferromagnets (AF), and random-field Ising ferromagnets (RFIM). Domain-wall hysteresis is studied and found to be qualitatively different for the AF and RFIM. This suggests that impurity pinning plays a significant role in the dynamics of the former system. The onset of time-dependent long-range order and magnetization "anomalies" are predicted for certain regions of the phase diagram.

In this paper we explore the differences and similarities of random-field ferromagnets, diluted antiferromagnets, and spin glasses in three dimensions (3D) by studying the evolution of the free-energy surface F, as the temperature T and the random or uniform external magnetic field are changed. The evolution of F forms the basis for a calculation of a number of irreversibility properties. For the random-field (Ising) ferromagnets[1] called RFIM and diluted Ising antiferromagnets called AF we determine the irreversibility phase diagram, the temperature and field dependence of the various history-dependent magnetizations and specific heats, and the magnetic field hysteresis of the domain-wall size. These are also compared with our earlier results[2] on spin-glass systems.

The approach we use is identical to that we used previously[2] to calculate irreversibility properties in spin glasses. This same numerical procedure has also been recently applied to diluted antiferromagnets by Yoshizawa and Belanger[3] whose work focused on establishing the relationship between the zero-field-cooled (zfc) process with long-range antiferromagnetic order (in 3D) and the field-cooled (fc) process with the formation of a multidomain state.[4] The essential physical assumption of our procedure is that on the time scale of a laboratory dc measurement the various random systems are trapped in a local minimum of the free-energy surface. Irreversibility on these laboratory time scales does not arise primarily from tunneling or thermal activation processes but rather from the disappearance of a given minimum as the field or temperature are changed. This disappearance in turn causes the system to reequilibrate (i.e., find the nearest minimum) and leads to irreversible behavior. Support for such a picture comes from the experimental observation that in these glassy magnets, cooling followed by heating procedures leads to reversibility whereas measurements made in the reverse order will *in general* show irreversible behavior.[2] The essential feature here is that both classes of experiments take place over the same time scale, so that the observed thermal hysteresis cannot derive primarily from time-dependent relaxation effects.

To understand this hysteresis we propose a highly nonequilibrium approach in which the evolution of a given minimum of F (prepared according to the appropriate experimental prescription) is followed. Because of numerical difficulties[2] deriving from the correction terms to the mean-field theoretic approximation to F, we will only consider the free-energy functional of mean-field theory. It should be stressed that this approximation is not basic to our *physical* picture. Moreover, it is equivalent to the Monte Carlo prescription for metastability both at low and high T where the Onsager corrections to mean-field theory are insignificant.

For all three random Ising spin-$\frac{1}{2}$ systems, we numerically solved the self-consistent equations $\partial F/\partial m_i = 0$. This implies

$$m_i = \frac{1}{2}\tanh\left[\beta/2\left(H + H_i + \sum_j J_{ij} m_j\right)\right] , \tag{1}$$

where m_i is the thermally average spin at site i, H_i a site random field with probability distribution $P(H_i)$ (which is zero except for the case of the RFIM). H is the applied field and J_{ij} the exchange interaction which is randomly distributed for spin glasses; for random-field ferromagnets $J_{ij} = J$ for all i, j and for diluted antiferromagnets $J_{ij} = -J\epsilon_j$. Here, $\epsilon_j = 1$ if the jth site has a spin and 0 otherwise. Equation (1) is solved iteratively following Ref. 2. We chose our systems to consist of up to $N = 2\times(30)^3$ sites on a bcc lattice for the AF case and up to $N = (30)^3$ spins on a sc lattice for the RFIM. A number of different random configurations were studied. In what follows temperature and field are measured in units of $J > 0$.

We first compute the irreversibility phase diagram for the diluted antiferromagnets which phase diagram has been a focal point for theoretical and experimental studies in spin glasses. We compared the fc states obtained upon cooling at constant H with the zfc states obtained by first cooling at $H = 0$, then applying H and reheating.[5] These states were distinct at the irreversibility phase boundary denoted by the solid line in Fig. 1(a). The concentration of magnetic sites is $c = 0.7$. Below the dashed line the zfc state [which also has long-range order[4] (LRO)] has the lower free energy. In

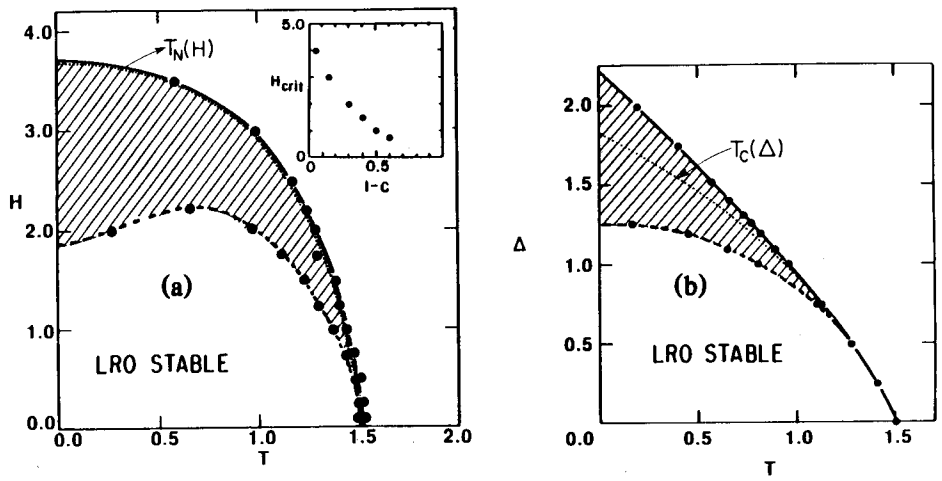

FIG. 1. Irreversibility phase diagrams for the (a) diluted antiferromagnet and (b) random field ferromagnet. Long-range order (LRO) is only metastable in the shaded region. The inset in (a) plots the $T=0$ intercept of the dashed line vs concentration.

the shaded region the fc state is more stable. The existence of this shaded region in the phase diagram corresponding to the lack of stability of LRO could be inferred from experimental considerations, provided one assumes that the free-energy surface evolves continuously with temperature. The inaccessibility of the LRO state upon cooling implies that it does not evolve directly out of the paramagnetic state free energy "well" as does the fc state. It is, therefore, quite natural that for some range of temperature the fc minimum will be deeper than that corresponding to LRO. If the system could access the stable LRO state upon cooling, it would follow that the multidomain state to LRO transition involves a finite jump in phase space. It is possible although it cannot be proved numerically that in thermodynamic equilibrium this would correspond to a first-order transition. Our numerical results support this schematic free-energy picture which has also been conjectured by Villain.[6] Furthermore, the existence of this "intermediate domain state[7]" [shaded region in Fig. 1(a)] should have a number of experimental signatures in the dc magnetization (discussed below) and leads to the onset of time dependence of the LRO state close to the Néel temperature or at sufficiently high fields.

In the inset of Fig. 1(a) is plotted the characteristic magnetic field H_{crit} above which the fc state is the more stable at $T=0$, as a function of c. Close to the percolation threshold this zero temperature critical field may be experimentally accessible. It should be noted that the dashed line in Fig. 1(a) is sensitive to finite-size corrections and that the relative size of the shaded region increases with the system size. By contrast, the irreversibility (solid) line in Fig. 1(a) is found to be relatively insensitive to system size. Within our numerical accuracy we could find no significant difference between the field-dependent Néel temperature $T_N(H)$ (obtained by extrapolating the finite field staggered magnetization to $N\rightarrow\infty$) and the irreversibility line. Note that $T_N(H)$ corresponds to the maximum temperature at which *meta*stable LRO can exist and is distinct from the *equilibrium* phase boundary, which is the dashed line in Fig. 1(a) [and its counterpart in Fig. 1(b)].

In Fig. 1(b) is plotted the irreversibility phase diagram for the RFIM for a field distribution $P(H_i)$ given by two delta functions at $H_i=\pm\Delta$. To obtain this irreversibility line we compared the two states prepared by cooling at constant Δ and by cooling to $T\sim 0$ at $\Delta=0$ and then applying Δ and heating.[5] The latter corresponds to long-range ferromagnetic order and the former yields a multidomain state. The region of stability of the domain state (shaded region) is less apparent[8] at low Δ than for the AF state at low H. Using a Gaussian distribution for $P(H_i)=1/(\sqrt{2\pi}\Delta)\exp(-H_i^2/2\Delta^2)$ we found led to a more pronounced intermediate domain state at small Δ. Furthermore, in this Gaussian case the irreversibility line appears to have a nearly vertical slope at low T. This reflects the fact that there are many distinct low-temperature domain states even for large Δ as a result of the weak field ($|H_i|<J$) sites in the alloy. Above the solid line the free energy has a single minimum as in the AF system. Metastable ferromagnetism persists up to temperatures T_c which are well inside the shaded region. At this $T_c(\Delta)$ line there appears to be a rather abrupt drop to zero magnetization particularly at $T=0$ (where our mean-field approach will coincide with Monte Carlo simulation results). As a consequence, the (meta)stable ferromagnetic state is reversible upon temperature cycling only up to $T_c(\Delta)$. Finally we note that for spin-glass[2] systems the field-dependent irreversibility line we compute using the same procedure coincides with the Almeida-Thouless prediction.[9]

In Fig. 2(a) are plotted the temperature-dependent fc (solid symbols) and zfc (open symbols) magnetizations for various H in the same AF system as in Fig. 1(a). The onset of irreversibility corresponds rather closely to the maximum in both magnetizations. This behavior is qualitatively similar to that measured by Ikeda and Kikuta[4] in $Mn_cZn_{1-c}F_2$, although the experiments were done on a considerably less anisotropic system. Two important features of the dc magnetization should be noted. Firstly, above the crossover field H_{crit} the low-temperature fc and zfc magnetizations invert so that $M^{zfc}>M^{fc}$; the opposite inequality holds below H_{crit}. This may be a useful experimental signature of the onset of metastability of LRO at low T. Secondly, we have found that M^{fc} is reversible at all T whereas M^{zfc} is reversible upon temperature cycling only up to roughly T_N. This reversibility in M^{fc} will be observable experimentally for sufficiently short times such that there are no appreciable relaxation effects. Close to T_N but inside the intermediate

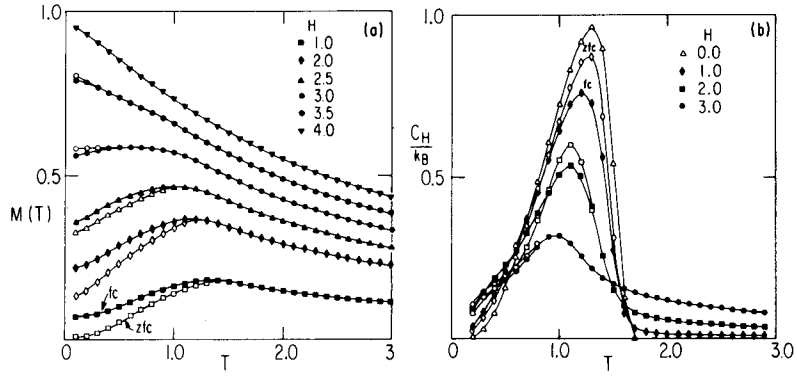

FIG. 2. Temperature dependence of the (a) magnetization and (b) specific heat for various fields in fc (solid symbols) and zfc (open symbols) processes.

domain state irreversibility in M^{zfc} sets in corresponding to the disappearance of this free-energy minimum, with increasing T. As a consequence, thermodynamic relations like the Maxwell equations should hold for all temperatures except close to T_N for the zfc state. Note that this reversibility will not be found upon heating an arbitrarily prepared low-temperature state. In fact, in spin glasses M^{zfc} is irreversible for all low T.

We have calculated the temperature dependence of the specific heat C_H for various H for the same AF system in the fc (solid symbols) and zfc (open symbols) states. Because of the Maxwell relations the field derivative of C_H is related to the temperature derivative of $M(T)$.[2] Thus, based on Fig. 2(a) some history dependence in C_H is expected; as shown in Fig. 2(b), the zfc specific heat is higher and has a slightly sharper peak than for the fc case. Both measurements are reversible upon temperature cycling below T_N. The peak in C_H seems to be somewhat below T_N for all but high fields. That we do not observe any sharpening of C_H upon increasing H, as is sometimes observed experimentally,[4] may be in part a consequence of our mean-field approximation. It should be noted that all the behavior shown in Fig. 2 has a counterpart in the RFIM system although there is no physically natural way of varying the Δ parameter. Our specific-heat curves are similar to experimental results obtained by Shapira and Oliveria.[4]

In Figs. 3(a)–3(c) we illustrate the "field hysteresis" of domain walls by studying (for the purpose of illustration) a two-dimensional RFIM with a Gaussian distribution for $P(H_i)$. Figure 3(a) shows the domains of up spin at $T=0$ after cooling at $\Delta=1.0$. If the "field" is then decreased to $\Delta=0.1$ the domains grow in size as shown in Fig. 3(b). When Δ is then increased back up to the initial value of 1.0 the domains shrink somewhat but are clearly larger than in the initial configuration at $\Delta=1.0$, as shown in Fig. 3(c). For the RFIM these same effects have been discussed[6] using domain-wall energy arguments. This magnetic field cycling is studied for the AF in Fig. 4. In this case we found magnetic field hysteresis was nearly unobservable[10] for a wide range of the fields studied in 3D as well as 2D. It can be seen from the figure that there is virtually no difference between the initial and final states [(a) and (c), respectively]. Furthermore, the differences between the first and second panels are relatively subtle in contrast to Fig. 3. This difference arises because in the AF case, domains still persist after field cooling when the applied field is subsequently reduced, even to $H=0$. Thus, these domains, which are closely tied to the impurity sites, are not able to significantly rearrange themselves at low fields. Therefore, the state returns to its starting configuration when the field is then increased to the initial value. This suggests that, due to impurity pinning, there may be a significant differ-

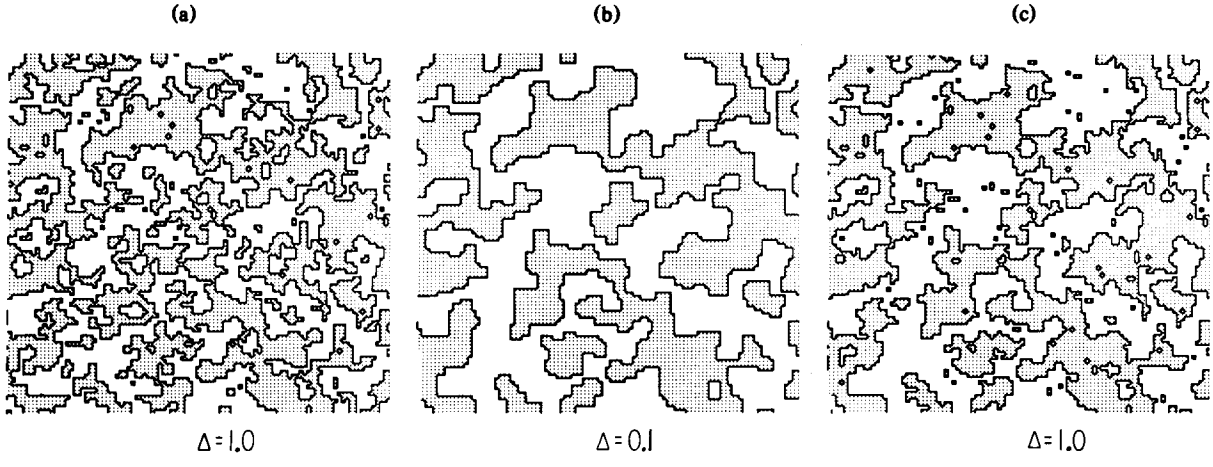

FIG. 3. $T=0$ domain configurations for a 100×100 RFIM system when (a) the system is cooled at $\Delta=1.0$, when (b) Δ is then lowered to 0.1, and when (c) Δ is then raised to the initial value 1.0.

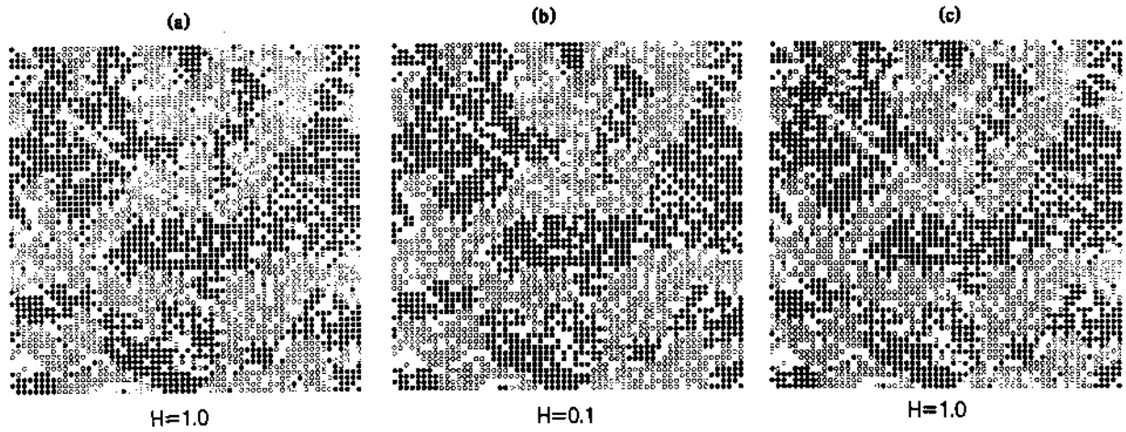

FIG. 4. $T=0$ domain configurations for an AF system with $c=0.75$ when (a) the system is fc at $H=1.0$, when (b) H is then lowered to 0.1, and when (c) H is then raised to the initial value.

ence between the dynamics of the two systems. To probe this we performed Monte Carlo simulations for both cases during a field cycling. At low T the results essentially reproduced the mean field theoretic calculations shown in Figs. 3 and 4. After a very small number of MCS (~ 10) the systems rapidly converged to metastable states and thereafter the time dependence was negligble. At higher T the RFIM exhibited the expected[11] relaxation effects, whereas at the concentrations studied ($c \sim 0.7$) the AF showed no significant time dependence.

In summary, the simultaneous presence of a magnetic field and intrinsic disorder leads to a higher degree of rigidity associated with domain motion in the AF case as compared to the RFIM. This reflects itself in the lack of field hysteresis and a delayed onset of temperature hysteresis in the AF state. Temperature hysteresis of the LRO state appears considerably closer to the irreversibility boundary in the AF case than in the RFIM. A crucial test of our approach will be the observation of time-dependent LRO somewhat below the irreversibility boundary *but still within the shaded region* of Figs. 1(a) and 1(b). Possible time dependence of the fc state within the shaded region cannot be ruled out since we could not ascertain whether the fc state is the equilibrium domain state.

This work was supported by the National Science Foundation under Grant No. DMR 81-15618 and the Materials Research Laboratory under Grant No. NSF-DMR 16892. This work has been submitted in partial fulfillment of the Ph.D. requirement (for C.R.) of The University of Chicago.

---

*Present address: Ames Laboratory and Department of Physics, Iowa State University, Ames, Iowa 50010.

[1]For a review of theoretical developments see, for example, G. Grinstein, J. Appl. Phys. **55**, 2371 (1984), and references therein.

[2]C. M. Soukoulis, K. Levin, and Gary S. Grest, Phys. Rev. B **29**, 1495 (1983). A number of experimental results in spin glasses including the reversible and irreversible properties of the magnetizations and the validity of the Maxwell's relations for the fc specific heat are summarized here, and discussed in the references.

[3]H. Yoshizawa and D. P. Belanger, Phys. Rev. B **30**, 5220 (1984). The present calculations were begun independently of this work.

[4]For a summary of experimental data, see R. J. Birgeneau, R. A. Cowley, G. Shirane, and H. Yoshizawa, J. Stat. Phys. **34**, 817 (1984); D. P. Belanger, A. R. King, and V. Jaccarino, J. Appl. Phys. **55**, 2383 (1984); P. Z. Wong, J. W. Cable, and P. Dimon *ibid.* **55**, 2377 (1984); Hironobu Ikeda and Kuniko Kokuto, J. Phys. C **16**, L455 (1983); Y. Shapira and N. F. Oliveria, Phys. Rev. B **27**, 4336 (1983).

[5]Cooling in zero H or Δ sometimes led to numerical difficulties associated with a delayed onset of LRO. To avoid these, in the zfc case the system was forced into a perfectly ordered state at $T=0$ after which the field was applied and the temperature raised.

[6]J. Villain, Phys. Rev. Lett. **52**, 1543 (1984); see also R. Bruinsma and G. Aeppli, *ibid.* **52**, 1547 (1984).

[7]The existence of an intermediate domain state was conjectured by Po-Zen Wong and J. W. Cable, Phys. Rev. B **28**, 5361 (1983).

[8]For small systems it takes a minimum Δ or H to numerically observe irreversibility because the domain size increases with decreasing Δ or H (see Ref. 6). While our calculations are therefore not accurate for small Δ or H, we believe the intermediate domain state exists down to $\Delta=0$ ($H=0$).

[9]J. R. de Almeida and D. J. Thouless, J. Phys. A **11**, 983 (1978).

[10]The absence of field hysteresis is perplexing in view of recent neutron scattering experiments. This discrepancy may in part be due to the highly anisotropic nature of the systems studied here.

[11]G. Grinstein and J. Fernandez, Phys. Rev. B **29**, 6389 (1984); J. Fernandez and E. Pytte (unpublished).