# Irreversibility in diluted antiferromagnets

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We compute the irreversibility phase diagram, and history-dependent magnetizations for diluted Ising antiferromagnets in three dimensions. We use an iterative mean-field technique which has proved quite successful for treating spin glasses. Motion of the domain walls as the magnetic field is varied at low temperature is also discussed. The onset of time-dependent long-range order and magnetization anomalies are predicted for certain regions of the $H$-$T$ phase diagram from our studies of the free-energy surface. Our results are compared to those of the random field ferromagnet and spin glasses, which have the common feature that on the time scale of a physical measurement the system is trapped in a local minimum of the free-energy surface. Irreversibility then results primarily from the disappearance of a given minimum as the field or temperature are changed and not from tunneling or thermal activation processes.

It is now well established[1] that a moderately large magnetic field does not necessarily destroy long-range order (LRO) in three-dimensional diluted antiferromagnets above the critical concentration. However, whether LRO is, in fact, observed depends sensitively on the history of the sample. At sufficiently low fields $H$ and temperatures $T$, in the zero-field-cooled (ZFC) configuration LRO is obtained, whereas in the field-cooled (FC) state it is not.

The close similarity between this irreversible behavior and that observed in spin glasses has motivated us to apply a numerical scheme (we previously developed[2] for studying spin glasses) to this "random-field" problem. Different aspects of our work[3] and a related study by Yoshizawa and Belanger[4] have been reported elsewhere. Our central physical assumption is that the irreversibility observed on intermediate time scales in glassy systems is governed more strongly by the evolution of the free-energy surface with changing $T$ or $H$ than by the much slower relaxation processes over barriers. On these time scales thermal and magnetic hysteresis occur because small changes in $T$ or $H$ destroy minima on the free-energy surface. When this happens the system will quickly reequilibrate, i.e., find its way to the nearest minimum. Only on much longer time scales will it relax toward the state of lowest free energy.

In this paper we implement this physical picture with a numerical scheme and thereby study irreversibility in diluted antiferromagnets. We compute the irreversibility phase diagram for several concentrations as well as the various field and temperature dependent magnetizations.

To study the evolution of the free-energy surface $F[m_i]$ for diluted antiferromagnets we numerically solved the Ising spin 1/2 mean-field equations, $\partial F/\partial m_i = 0$. This implies

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

where the near-neighbor antiferromagnetic interaction $J_{ij} = J\epsilon_j$ and $\epsilon_j = 1$ (or 0) if the $j$th site is occupied (or unoccupied) by a magnetic atom and $J < 0$. Throughout this paper $T$ and $H$ will be measured in units of $|J|$. Calculations which attempted to improve upon mean field theory always led[5] to unphysical results and/or numerical difficulties. On bcc and sc lattices of $N = 54\,000$ and 125 000 sites, respectively, we numerically solved the $N$ simultaneous equations represented in Eq. (1) using an iterative technique.[2] In this way it was possible to follow a given minimum of $F$ prepared according to the appropriate experimental prescriptions.

In Fig. 1 we plot the spin configurations obtained at $H = 1.0$ after FC (a) and ZFC (b) processes in a $(50)^3$ system[6] with magnetic atom concentration $c = 0.75$. As can be seen, the FC state is permeated by domains of reversed "staggered spins" (solid circles) so that there is no LRO. By contrast the ZFC state consists of a single domain with a few "wrong" spins (solid circles). Note that the inverted spins or domains generally occur around vacancies.

Above the field-dependent Néel temperature $T_N(H)$ (defined as the maximum temperature at which the system can support LRO upon warming in a field) the FC and ZFC states are essentially indistinguishable. This line then closely

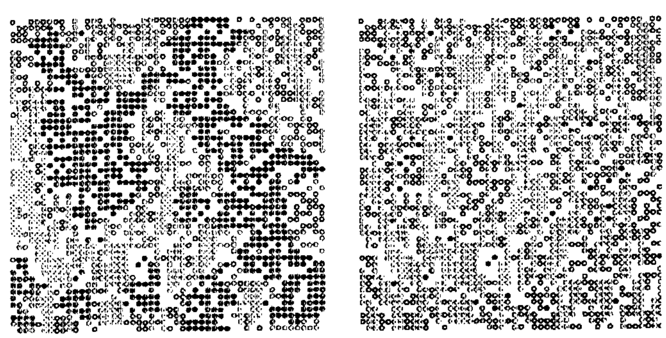


FIG. 1. Spin configuration for one layer of a sc lattice in the FC (a) and ZFC (b) states for $T = 0$ and $H = 1.0|J|$ with $c = 0.75$. The open and closed circles correspond to positive and negative staggered magnetizations, respectively.

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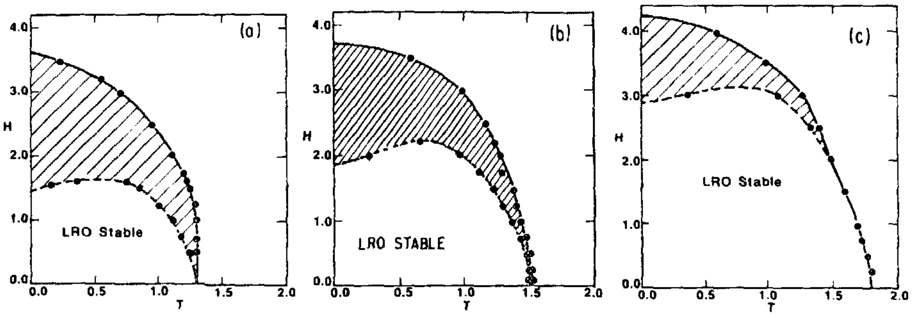


FIG. 2. Irreversibility phase diagrams for concentrations $c = 0.60$ (a) and $c = 0.70$ (b) and $c = 0.85$ (c) on a bcc lattice; long-range order (LRO) is only metastable in the shaded region.

coincides with the irreversibility temperature which is plotted in Figs. 2(a)–2(c) (solid lines). In this figure the concentrations shown correspond to $c = 0.6$ [Fig. 2(a)], $c = 0.7$ [Fig. 2(b)], and $c = 0.85$ [Fig. 2(c)]. The dashed line in the figures is the boundary of the shaded region in which the ZFC state is of higher free energy than the FC state; thus in this shaded region LRO is only metastable. By comparing Figs. 2(a)–2(c), one can see that the more magnetically dilute the alloy the larger the region of metastable LRO. While it is quite narrow for the less diluted alloy, it can be argued on quite general grounds that this shaded region must intercede between the paramagnetic and stable magnetically ordered region at all $H$. This is a direct manifestation of the fact that LRO is not accessed upon cooling at any nonzero field.[1] The free-energy minimum which evolves directly out of the high-temperature paramagnetic state is the multidomained FC state. While the ZFC minimum persists to all temperatures lower than the irreversibility temperature, warming the ZFC state *above* this temperature leads to irreversibility since the system then reequilibrates into the FC state. Because it appears only at much lower temperatures, the ZFC minimum is not as deep as that of the FC state for a narrow range of temperatures near the onset of irreversibility. However, at somewhat lower temperatures the long-range ordered state is the more stable. All of these expected features are found in our numerical calculations. Villain[7] has argued for a similar picture using a rather different viewpoint.

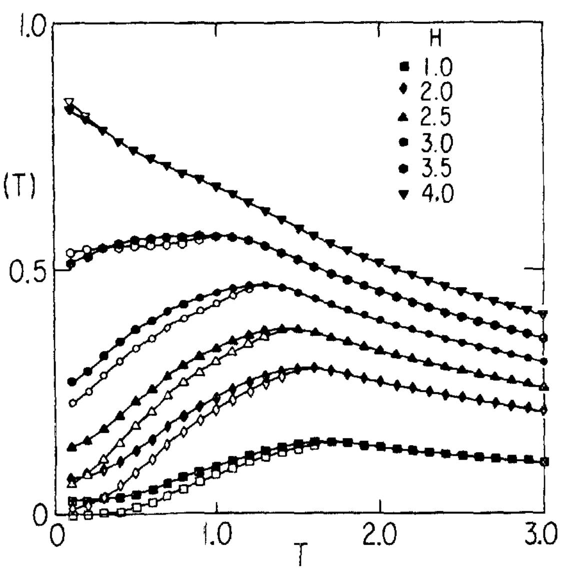


FIG. 3. Temperature dependence of the uniform magnetizations for various fields in FC (solid symbols) and ZFC (open symbols) processes for $c = 0.85$ on the bcc lattice of Fig. 2(c).

In Fig. 3 are plotted the temperature-dependent magnetizations for $c = 0.85$ on a bcc lattice for a range of magnetic fields. The open and solid symbols correspond to the ZFC and FC states, respectively. While the FC magnetization is generally higher than that of the ZFC state, near the border of the shaded region of Fig. 2(c) the relative magnitudes of the two magnetizations invert. Although this is a small effect, it was seen consistently for a range of concentrations and it presumably arises from topological constraints on the distribution of the spins which can be polarized by the field. This may be a useful signature of the onset of metastable LRO. An additional signature should be the onset of time dependence in the ZFC state close to $T_N$. We find that the irreversibility temperature coincides rather closely with the maximum in the two magnetizations. The $T$ dependent magnetization we compute appears to be qualitatively similar to measurements obtained by Ikeda and Kikuta,[8] although their systems were not truly Ising-like.

We also note that the magnetization in the FC state is reversible over the entire temperature region: this should be observed experimentally provided there is no relaxation over barriers out of the FC state. As noted above, in the ZFC state the magnetization is reversible only as long as the temperature does not exceed the characteristic irreversibility temperature. These features which derive from the behavior of the various free energy minima, appear[1] to be generally consistent with the data.

In Ref. 3 we presented results for the magnetic field hysteresis obtained by cycling $\Delta$ in a random-field Ising ferromagnet (RFIM). This system is presumed[9] to be closely related to a diluted antiferromagnet in an external field. In the RFIM the domains grow irreversibly when the random-field variable $\Delta$ is first decreased and then increased back to the initial value. By contrast in the diluted antiferromagnets we did not detect any appreciable magnetic hysteresis. This is one of the few examples of a qualitative difference between these two systems, and it is therefore of some importance. This effect derives from the fact that only the antiferromagnetic system contains intrinsic disorder (i.e., even at $H = 0$). This disorder "pins" the domains and prevents them from growing irreversibly when the field is cycled. The extent of this impurity "pinning" manifests itself in time dependent effects which will be addressed in a future publication.

In summary, we have found that the free-energy surface $F[m_i]$ for the diluted antiferromagnets is very similar to that of the spin glasses: once a minimum is formed it persists to all lower temperatures.[2] Changing the magnetic field $H$ or heating from an arbitrarily prepared ground state leads to the disappearance of a given minimum and hence to irreversibility. This picture which leads to qualitative agreement with experiment is believed to be general and not a consequence of the mean-field approximation. Furthermore, it should be stressed that at low $T$ this approximation becomes exact. This means that our calculated phase diagrams (whose behavior is fairly circumscribed by the low $T$ results) are of general validity and can also be verified by Monte Carlo simulations.

What is neglected in the present approach are the very slow relaxation processes which are presumed to lead[7] to domain radius growth $\propto \ln t$. This is reminiscent of the slow relaxation observed in spin glasses and arises in our free energy picture in much the same way, from transitions over barriers between different metastable states.

## ACKNOWLEDGMENTS

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