Field-Induced "Exchange Flips" in a Randomly Diluted Antiferromagnet

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High-field magnetization studies of the randomly diluted, anisotropic antiferromagnet $Fe_xZn_{1-x}F_2$ have confirmed the existence of novel single-spin "exchange flips," occurring at fields $H_0=\frac{1}{8}nH_E$ ($n=1,\ldots,5$), as predicted by a classical spin computer simulation. Field direction hysteresis of dM/dH_0 is shown to result from "crossover" behavior to states which have no long-range order.

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The effects of random dilution of an antiferromagnet (AF) on the static and dynamic properties ($\chi_\perp$, $C_{\rm mag}$, $\nu_{\rm AFMR}$, spin correlations, phase diagrams, critical exponents, etc.) are of current interest.[1] Our own susceptibility studies of these systems[2] were aided by $T=0$ K computer simulations. In calculating the spin-flop (SF) field $H_{\rm SF}$ of diluted FeF_2, we noticed that spins with $n=1$, ..., 5 exchange-coupled neighbors would have effective exchange fields $H_E'=(n/z)H_E$ less than $H_{\rm SF}$, where $z=8$ is the coordination number, and H_E the exchange field in FeF_2. When the external field H_0 just exceeds H_E', the total field acting on those spins *residing on the down sublattice* (moments antiparallel to $\vec{H}_0$) reverses direction. An abrupt magnetization reversal ("exchange flip") will occur for these spins at $H_0=\frac{1}{8}nH_E$, with $n=1$–5, or for which $H_E'<H_{\rm SF}(x)$. We have experimentally verified these predictions and found other interesting phenomena associated with the magnetic phase diagram of a randomly diluted, large-anisotropy AF.

The experiments were performed at the High Magnetic Field Laboratory, Osaka University, on ten crystals of $Fe_xZn_{1-x}F_2$. Pulsed fields up to 550 kOe were repetitively produced in a double-layer coil of maraging steel (maximum field: 600 kOe). Rod-shaped crystals ($2\times2\times15$ mm^3) were oriented with the long (c) axis parallel to $\vec{H}_0$ to within 0.5° and immersed in liquid helium for 1.3- and 4.2-K measurements. Both sample magnetization M and H_0 were measured with a set of calibrated, compensated pickup coils whose voltages were recorded by a dual-channel, digital transient recorder, and then processed to yield M and dM/dH_0 vs H_0. The $T=1.3$ K data on dM/dH_0 vs H_0 for two samples (F and G) are shown in Fig. 1. Sample F ($x=0.73$) shows peaks at values of $H_0=\frac{1}{8}nH_E$ for $n=2$, 3, 4, and 5. The peak which approximately coincides with the $n=5$ position probably arises from SF because (a) it occurs at the calculated value of $H_{\rm SF}$, (b) it is seen in both increasing and *decreasing* H_0, and (c) no higher "flips" are observed. Provided that $H_{\rm SF}$ is exceeded, the "exchange flips" ($n=2$–4) are observed only in *increasing* H_0. This irreversibility in dM/dH_0 with respect to the direction in which H_0 is swept will be shown to arise from the existence of several low-lying states of the system and to the absence of long-range order.

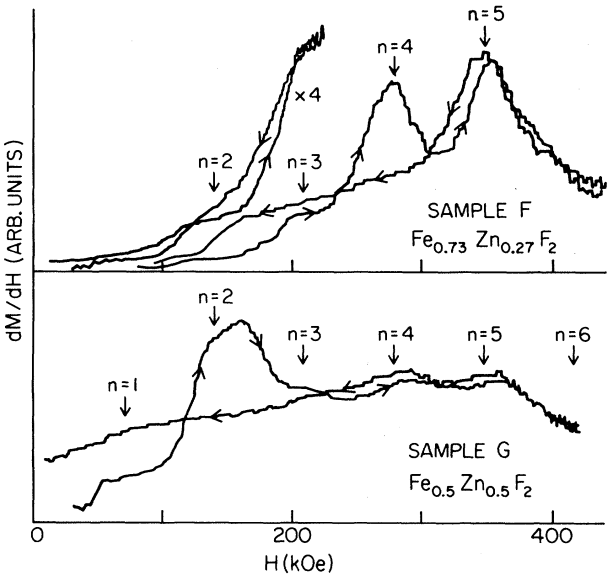

FIG. 1. dM/dH_0 vs H_0 for two samples. Arrows indicate the direction of field scan, and the "exchange-flip" fields are shown. The $n=5$ peak in sample F is actually SF, and the anomalous $n=2$ peak in sample G is due to ΔM at H_{cr}.

The more dilute sample G ($x = 0.50$) exhibits only "exchange flips" but no SF despite the virtual-crystal-approximation prediction that SF would persist until $x \lesssim 0.4$. Here $H_E(x) = H_A(x)$, the anisotropy field, and metamagnetism would occur. Note that the $n \geq 3$ "exchange flips" are seen in *both* increasing and decreasing H_0 while the $n \leq 2$ ones display the hysteretic behavior mentioned above. This hysteresis boundary decreases in field with decreasing x. It is accompanied by an apparently anomalous peak in dM/dH_0, occurring at $H_0 \approx \frac{2}{8} H_E$ for sample G. Further dilution moves this peak towards lower H_0, bringing it below the $n = 1$ value near the percolation limit $x_p = 0.24$ (samples H and I).

The "exchange flip" and SF data for all samples are collected in Fig. 2. The horizontal dashed lines are the predicted positions of the $n = 1, \ldots, 5$ "exchange flips" if one assumes the interaction between Fe pairs to be independent of x. It is most remarkable how well the data agree with these predictions. SF, which does vary with x, begins above the $n = 5$ "exchange-flip" field for $x \simeq 1$ but terminates fortuitously close to it, around $x = 0.5$. The hysteresis boundary between the high- and low-field "exchange-flip" behavior, characterized by the "anomalous" peak in dM/dH_0,

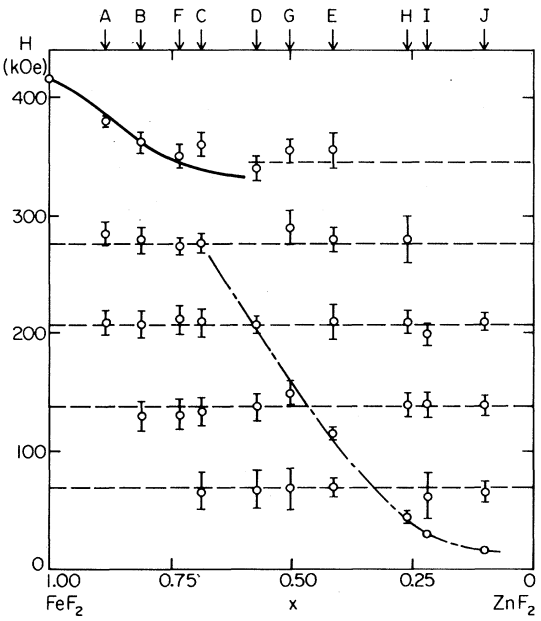

FIG. 2. dM/dH_0 peak fields for all samples. Horizontal dashed lines are predicted "exchange-flip" fields. The dot-dashed line marks the anomalous peak at H_{cr}, and the solid line is the predicted H_{SF}, ending at $x \approx 0.55$.

is shown by the broken line. It falls below the $n = 1$ field value at smaller x but remains finite even beyond x_p.

The energy E of the "flip" states in a field H_0, applied collinear with the easy axis, is simply the Zeeman energy in $H_0 + H_E'$. Except in the SF state, E is unaffected by H_A. E and M were calculated by a classical-spin computer simulation of a randomly diluted lattice of $10 \times 10 \times 20$ spins in which each spin was aligned parallel to the effective field at its site. Although only one spin was allowed to flip at a time, the process was iterated to ensure a self-consistent solution. A more general approach would also include the turning over of larger clusters with net moments and small net exchange.

As H_0 increases from zero, E and M may also be obtained from the same model and simple probabilistic considerations. The concentration of occupied sites with n neighbors is $C(n) = \binom{z}{n} \times x^{n+1}(1-x)^{z-n}$. In a field $H_0 = (n/z)H_E$, all "down" sublattice spins with n or fewer neighbors flip "up", and the energy E becomes (relative to the AF state)

$$E = 2g\mu_B S H_E N\left[\sum_{m=1}^{n} (m/z)C(m) - (n/z)\sum_{m=1}^{n} C(m)\right]. \quad (1)$$

Similarly, the change in M at $H_0 = (n/z)H_E$ is

$$\Delta M = 2g\mu_B S N C(n). \quad (2)$$

The predictions of Eqs. (1) and (2) were compared with the computer simulations, and were found to be in nearly exact agreement.

If $H_0 > H_E$, the spins are fully aligned, and the original identities of the sublattices are lost. When H_0 is then decreased below $H_E'(n)$, a new, *decreasing field* (DF) state is entered, in which spins with n neighbors on *both* sublattices may flip "down" (providing, of course, that all n neighbors remain "up").[3] A simulation of this process reveals that the DF state has E slightly lower, and M slightly higher than the corresponding *increasing field* (IF) state, for H_0 above a certain threshold or crossover field, H_{cr}. Below H_{cr}, the DF state rapidly becomes much higher in energy than either the IF or the AF state. Since the DF state is nucleated from randomly placed spins, it possesses no long-range order, but is composed of many small regions of nominally "up" or "down" spins on what was a particular sublattice in the AF state. As H_0 is lowered, the real system tries to relax to the AF state

either by reversing some of these regions, or by boundary motion. Since the simulation allows only single spin flips, however, such processes are not considered, and the system finds itself trapped in a state nucleated at the $n=8$ field. Thus the difference between the IF and DF states should explain the hysteresis in M seen in the experiment. Moreover, the calculated values of H_{cr} agree with the observations shown in Fig. 2, when $H_{cr} < H_{SF}$ or no SF exists.

Compelling evidence for crossover and hysteretic behavior is given by the simulation of dM/dH_0 vs H_0, presented in Fig. 3 for samples F and G. The peaks in dM/dH_0 are plotted as superimposed Gaussians of the predicted amplitudes and an x-dependent width[4] δH, chosen to best fit the data. Note that the IF "exchange-flip" calculation agrees well with the IF data up to H_{SF} in sample F, in which the predicted $H_{cr} \approx H_{SF}$, while the DF data agrees better with the DF prediction. The δH in the latter case was increased to fit the lack of structure seen in the data, which fall below the DF calculation toward the IF one at lower H_0. These two effects suggest that *rate-dependent* effects may be important, giving both peak smearing and a gradual DF to IF crossover. The SF region in sample F is discussed below; note here that the lack of an $n=6$ "flip" is clear evidence the peak at the $n=5$ position is actually SF, and not the $n=5$ "flip". Further, the DF-type behavior below H_{SF} shows that SF nucleates a DF state similar to that nucleated by the fully aligned state.

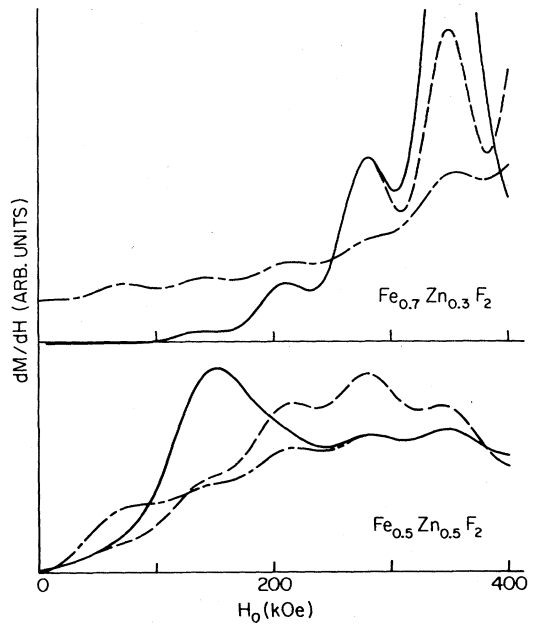

FIG. 3. Simulations of dM/dH_0 vs H_0. The $n=1-6$ IF "flips" (dashed) and DF "flips" (dot-dashed lines) are shown. For $x=0.7$, the solid line represents IF flips $n=1-4$ plus SF. For $x=0.5$ the solid line represents IF "flips" $n=1$ and 2, DF "flips" $n=3-6$, and ΔM at H_{cr}.

In sample G, dM/dH_0 closely follows the IF prediction in small H_0, but clearly crosses over to DF behavior above the $n=3$ "flip". Here it agrees very well with the DF calculation for *both* IF and DF. Further, the crossover idea explains the anomalous peak above the $n=2$ flip. Since M in the DF state exceeds that in the IF one by ΔM, one expects a peak at H_{cr} of this magnitude, in addition to the "flip" peaks. The IF crossover was simulated by the IF "flips" $n=1$ and 2, the DF "flips" $n=3-8$, and ΔM between the DF and IF states at $H_0=160$ kOe, with a single width. The resulting curve agrees quite well with the data; the sole disparity is that the crossover peak appears narrower than the "exchange-flip" peaks.

In decreasing H_0, the data follows the DF prediction all the way to $H_0=0$. The hysteresis in this case cannot be entirely due to rate-dependence effects, as evidenced by the IF crossover. We conjecture the following: In increasing H_0, the DF state may be reached, perhaps in a large number of ways, by flipping small clusters of spins, each cluster reversing itself *independently* of all others. The situation has similarities with the spin-glass problem,[5] in that there is a (perhaps large) set of nearly degenerate ground states in intermediate fields. In decreasing H_0, however, the long-range order of the IF state can be reestablished only by a *coordinated* reversal of *all* the previous flips, which can be done in only a single way. Thus the DF crossover is a cooperative, long-range phenomenon while the IF one is a set of independent, smaller-scale ones.

The calculation of the SF energy (E_{SF}) and M involved a computer simulation similar to the flip case. We let $Q_i = \partial H/\partial \varphi_i$,[6] where φ_i is the angle between the ith spin and the $\pm x$ axis, $\perp \vec{H}_0$. When SF is the ground state, $\{Q_i = 0\}$ can be solved by iteration, if we assume fixed values of all φ_j's with $j \neq i$ while solving $Q_i = 0$. When SF does not have the lowest E, this iteration scheme becomes unstable and converges on one of the "exchange-flip" states. In this case, the variance $\sigma^2 = \sum Q_i^2$ was minimized with respect to φ_i, fixing all φ_j's with $j \neq i$. φ_i was set to this value, and the process was iterated. Con-

vergence was considerably slower than before, but agreed with the previous SF state calculation. M was found to be linear in H_0, at least up to H_{SF}, with a slope slightly less than the virtual-crystal-approximation value. At higher H_0, dM/dH_0 decreases noticeably, becoming greatly reduced as M saturates.

H_{SF} is the field at which E_{SF} crosses below the IF exchange-flip state energy (E_{IF}); the results shown in Fig. 2 agree with the data. For $x \approx 0.55$, E_{SF} becomes equal to, but not less than E_{IF}; hence for $x \lesssim 0.55$, no SF occurs. This leveling off of H_{SF} with dilution is directly attributed to the existence of the "flip" states. Since E_{IF} lies below the AF energy, the H_0 required to bring E_{SF} below the former is greater than for the latter.

The calculated ΔM at H_{SF} in sample F is shown in Fig. 3, in the same manner as for the "exchange flips". Note that the calculated ΔM greatly exceeds the observed one, a result not understood but undoubtedly related to the greater complexity of the SF state than has been considered in this model. Further details of this work, including the temperature dependence of H_{SF} and the "exchange-flips" fields, will be published elsewhere.

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[1]Some recent work in this field and many earlier references are contained in R. A. Cowley *et al.*, Phys. Rev. B 22, 4412 (1980); R. A. Tahir-Kheli and A. R. McGurn, Phys. Rev. B 18, 503 (1978); C. B. de Araujo, Phys. Rev. B 22, 266 (1980); H. Rohrer *et al.*, J. Magn. Magn. Mater. 15–18, 396 (1980); D. Belanger *et al.*, J. Magn. Magn. Mater. 15–18, 807 (1980); F. G. Brady *et al.*, J. Appl. Phys. 50, 1726 (1979). An extensive review of the work before 1976 is found in R. A. Cowley, in *Magnetism and Magnetic Materials—1975*, edited by J. J. Becker, G. H. Lander, and J. J. Rhyne, AIP Conference Proceedings No. 29 (American Institute of Physics, New York, 1976), p. 243.

[2]A. R. King and V. Jaccarino, J. Appl. Phys. 52, 1785 (1981).

[3]No simple probability arguments can be applied to this case, because of the complexity of the implied conditional probabilities. While in the IF case, nearly all "up" sublattice spins remain "up", it is by no means obvious that the neighbors of an $n = 8$ spin (or any other n) will have $n < 8$ (or other n), and will therefore remain "up". This is especially clear for small dilution.

[4]The Gaussian half-widths δH used are 26 kOe (sample F) and 38 kOe (sample G), and for other samples (now shown) vary as $x(1-x)$ as expected for a strain-induced exchange broadening. If the variation of each exchange coupling were statistically independent then one might expect $\delta H \propto n^{1/2}$. For simplicity we have assumed δH not to depend on n.

[5]A. B. Harris and S. Kirkpatrick, Phys. Rev. B 16, 542 (1977).

[6]See, for example, A. Blandin, J. Phys. (Paris), Colloq. 39, C6-1499 (1978).

Dynamics of Vector Spin-Glasses

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A description of spin-glass dynamics at low temperatures, based on a "harmonic" theory, is presented. The Hamiltonian is approximated by a quadratic form in the spin deviations from a particular local minimum, and diagonalized. The dynamics is governed by the eigenvalue distribution $\rho(\lambda)$. The validity of this description is supported by computer simulations. These suggest that $\rho(0) \neq 0$ in two and three dimensions, implying a logarithmic decay in time of the spin autocorrelation function at low temperatures.

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The unusual low-temperature properties of spin-glasses are thought to be due to the very large number of metastable states which exist in these systems: the Hamiltonian has many local minima of comparable energy. Within the context of the Sherrington-Kirkpatrick model,[1] whose solution plays the role of a mean-field theory for spin-glasses, the number and properties of these minima may be calculated exactly,[2] but for more realistic models analytical progress is difficult. At low temperatures we will assume that at any instant of time the spin configuration is "close" to that characterizing one of the local minima. Dynamical processes are then of two