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[15]It is interesting to note that for $p < p_c$ and $r_s \gg \xi$ one expects the large finite clusters to scale like "animals" (Ref. 1). The form (2) may then yield new diffusion laws. A new exponent θ_a may then replace θ in Eq. (1) when $\xi \ll r(t) \ll r_s$. In the present paper we ignore these (exponentially rare) cases.
[16]Following the present work, the authors of Ref. 10 looked again at their short-time data and found $\langle r^2(t)\rangle \sim t^{0.66}$ ($d=3$) and $\langle r^2(t)\rangle \sim t^{0.8}$ ($d=2$). Considering the error bars, and the fact that for very short times $\langle r^2(t)\rangle \propto t^2$, biasing the measured exponents to higher effective values, these preliminary results are probably not inconsistent with our predictions.
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[22]For $d=3$ gold samples, characterized by grains of size $a \sim 100$ Å, with $\xi \sim 50a$, at $T \sim 300$ K, we estimate $D(\xi) \sim 10^{-2}$ cm^2/sec. The characteristic crossover frequency is thus $\tilde{\omega} = (A/\xi)^{2+\theta} \sim 10^7$ sec^{-1}. The trivial upper limit is $1/\tau_0 \sim 10^{14}$ sec^{-1}, leaving a wide frequency range for experiments. Details will be published elsewhere.
[23]Quantum localization behavior is expected for $\omega \gtrsim 10^{12}$ sec^{-1}; see Ref. 14.

Absence of Irreversibility in Isotropic Heisenberg Spin-Glasses: Anisotropy Effects

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On the basis of a numerical study of a mean-field free-energy surface, the authors propose that an isotropic Heisenberg spin-glass has no irreversibility. The field-cooled and zero-field–cooled states are the same and magnetic hysteresis is absent. The authors show how Dzyaloshinsky-Moriya and uniaxial anisotropy effects lead to irreversibility. Hysteresis loops exhibit sharp jumps at constant field only when both microscopic anisotropy and a ferromagnetic tendency are present.

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There have recently been a number of theoretical studies of the Heisenberg spin-glass, both with[1] and without[2,3] anisotropy. Because there exist in the laboratory vector spin-glasses exhibiting a wide variety of anisotropy interactions, it is now possible to test theoretical predictions. Monte Carlo simulations[4] on Ruderman-Kittel-Kasuya-Yosida–interaction Heisenberg spin-glass suggest that microscopic anisotropy is necessary in order to obtain a cusp in the spin susceptibility. Theoretical studies of the infinite-range isotropic Heisenberg model find a spin-glass phase and the associated replica-symmetry breaking[3] (which is presumed to be related to irreversibility). By contrast, a working hypothesis of phenomenological approaches[5] is that macroscopic irreversibil-

ity exists only in the presence of microscopic anisotropy.

We demonstrated[6] that a study of the behavior of the minima of the free-energy surface as functions of temperature T and the external field H could explain the nature of reversible and irreversible behavior in an Ising spin-glass. It is the purpose of the present paper to extend this numerical study to the Heisenberg case with and without anisotropy effects and to report some observations which show that there are *qualitative* differences between isotropic Heisenberg and Ising spin-glasses. As discussed previously,[6] the reaction term in the free-energy surface $F\{\vec{m}_i\}$ leads to unphysical minima to which the system will readily flow. (Here $\vec{m}_i$ is the thermal average of the vector spin at the ith site.) Consequently, we can consider only the mean-field terms. Despite the clearly oversimplified approach, it represents a necessary first step in understanding the relationship between microscopic anisotropy and irreversibility.

Our Hamiltonian consists of an isotropic random near-neighbor exchange interaction J_{ij} with Gaussian distribution of width $\bar{J}$ and mean J_0, a uniaxial anisotropy term $\sum_i D(S_i^z)^2$, and a Dzyaloshinsky-Moriya (DM) anisotropic interaction $\sum \vec{D}_{ij} \cdot \vec{S}_i \times \vec{S}_j$. Here $D_{ij}{}^{\mu} = -D_{ji}{}^{\mu}$ is given by a random delta-function distribution $\delta(D_{ij}{}^{\mu} \pm D')$ and μ, ν are Cartesian coordinates. When uniaxial anisotropy is absent, the variational condition $\partial F/\partial \vec{m}_i = 0$ requires

$$\vec{m}_i = (\vec{h}_i/|\vec{h}|) B_s(|\vec{h}_i|), \tag{1}$$

where B_s is the generalized Brillouin function for spin S and $h_i{}^{\mu} = \sum_{j\nu} J_{ij}{}^{\mu\nu} m_j{}^{\nu} + H\delta_{\mu z}$. The diagonal elements of $J_{ij}{}^{\mu\nu}$ are J_{ij} whereas the off-diagonal elements are simply related to $\vec{D}_{ij}$ within mean-field theory. All values of D', D, T, and the field H are in units of $\bar{J}$. When uniaxial anisotropy is included, Eq. (1) may be generalized for the quantum spin case $S=1$ (which value of S we use throughout this paper) by diagonalizing the matrix representation of the 3×3 mean-field Hamiltonian and using a numerical procedure to solve the resulting cubic equation. It should be noted that at zero temperature Eq. (1) implies that each $\vec{m}_i$ is parallel to $\vec{h}_i$. This criterion for metastability coincides with that derived on more general grounds by Ma,[7] and used in Monte Carlo simulations.[4] We solve Eq. (1) and its appropriate generalization ($D \neq 0$) iteratively for spins on a simple cubic lattice. For each new (T, H) we started our iterations at the $\{\vec{m}_i\}$ corresponding to the minimum of F evaluated at the limit values of the previous T or H. We then changed $\{\vec{m}_i\}$ by using an updated sequencing of the sites i until we got convergence at the nth iteration defined by[8]

$$\sum_i (\vec{m}_i{}^n - \vec{m}_i{}^{n-1})^2 / \sum_i (\vec{m}_i{}^n)^2 < 10^{-10}.$$

For convenience we also define $Q_{\parallel}$ and $Q_{\perp}$, the longitudinal and transverse order parameters (referred to the field direction): $\sum_i m_i{}^{\mu} m_i{}^{\mu} = Q^{\mu}$. The spin-glass transition temperature T_g is defined as the lowest temperature at which $Q_{\parallel}$ vanishes. It was necessary in all our calculations to apply a small ($\sim 10^{-4}\bar{J}$) transverse field in order to avoid supercooling into a longitudinally ordered state of higher free energy.

Our results for the Heisenberg case with and without anisotropy have many of the same features we reported earlier for the case of Ising spins. As we cool at constant H we always stay in the same minimum.[9] However, changing H generally leads to the disappearance of a minimum and thus to "minima hopping." The unique aspect of the isotropic Heisenberg system is that upon hopping the system settles into a minimum which is the field-cooled (FC) state at the appropriate H (arbitrarily positioned in the x-y plane). This is clearly seen by studying $M_z \equiv \sum_i m_i{}^z$ vs H which shows macroscopic reversibility but microscopic irreversibility (deriving from uniform spin rotations). *In the isotropic case, because of the ready accessibility (associated with rotational degeneracy) of the FC state, there is no measurable irreversibility.* These results are not necessarily in conflict with those of Ref. 3 because these authors[3] used the infinite-range model and only made a loose association between the zero-field–cooled (ZFC) susceptibility and that obtained in linear response theory. The contrast between Ising (which show clear hysteresis[6]) and Heisenberg spin-glasses mirrors the (mean-field) calculated hysteresis curves of *nondisordered* ferromagnets. In the second case, hysteresis is absent because Heisenberg spins can rotate freely to "follow" the field.

In Fig. 1(a) are plotted the FC and ZFC (indistinguishable) magnetizations (normalized to the maximum value of $S=1$) for the isotropic Heisenberg case as a function of temperature for $J_0 = 0$, $N = 10^3$, and various H. Once anisotropy of any kind is introduced the situation is radically changed; there is hysteresis, remanence, and, for small enough H, a splitting of the FC and ZFC magnetizations as a function temperature. The latter are plotted in Fig. 1(b) for $H = 0.5$ and

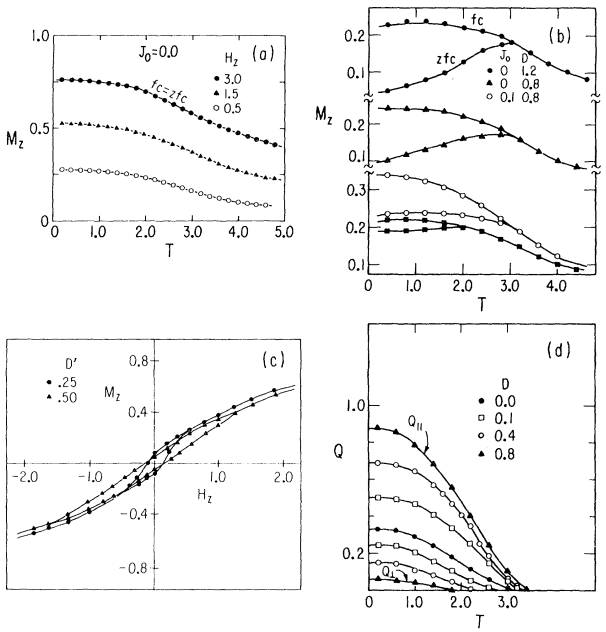

FIG. 1. Temperature T dependence of field-cooled and zero-field-cooled magnetization for (a) isotropic and (b) anisotropic interactions. Anisotropy constants D and ferromagnetic exchange J_0 are as indicated. Squares denote pure DM case $D'=0.25$, $J_0=0$. (c) Hysteresis loops for various Dzyaloshinsky-Moriya interactions, D', for $J_0=0$. (d) Temperature dependence of longitudinal and transverse order parameters for several (uniaxial) D, $J_0=0$.

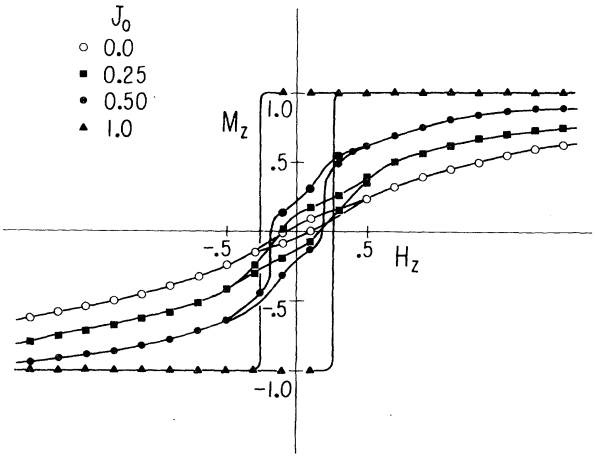

FIG. 2. Hysteresis loops for uniaxial anisotropy (at $D=0.4$) for various values of J_0 at low T ($T=0.2$).

different values of D and J_0. The last case (squares), $D'=0.25$, corresponds to a pure DM anisotropy and the first three to pure uniaxial anisotropy. For the uniaxial case with $D=0.25$, we get semiquantitatively similar behavior as in the DM case. Because of the numerical complexity (particularly in the uniaxial case), we were forced to study somewhat smaller systems than for Ising spins ($N=6^3$ and 8^3 for the uniaxial and DM cases, respectively). Consequently we had to consider larger values of H (to make the finite-size-induced zero-field magnetization insignificant). This in turn required somewhat larger values of D than may be physical, to obtain a significant splitting of the FC and ZFC magnetization. Despite these limitations, the results shown here illustrate clearly the systematic dependence of the macroscopic variables on the various parameters in the Hamiltonian.

Hysteresis loops for the pure DM case at $T=0.2$ and $D'=0.25, 0.5$ are shown in Fig. 1(c), and in Fig. 1(d) are plotted (upper curves) $Q_\parallel$ and (lower curves) $Q_\perp$ for different values of the uniaxial anisotropy D at $H=0$ and $J_0=0$. From these can be determined the characteristic value of T_g. We have found a weak quadratic dependence of $Q_\perp$ on H^2 (as in previous theories[3]). It follows from this and Fig. 1(d) that for $D=0.8$ and $J_0=0$, $Q_\perp$ turns on at $T\approx 2.0$. For the case $D=1.2$, $J_0=0$, the corresponding temperature is $T\approx 1.2$. These temperatures do not correlate with any structure in the T dependence of the FC and ZFC magnetization. We find that irreversibility sets in (approximately) when longitudinal spin-glass order begins. The maximum value of D at which $Q_\perp\neq 0$ for all T is roughly 2.0, which is considerably larger than that found for the infinite-range model.[3]

An interesting feature which is seen in hysteresis experiments in the dilute spin-glasses containing Mn,[5] and in more concentrated AuFe,[10] is the sharp reversal of magnetization at fixed H and low T. These hysteresis loops are quite different from our previously published[6] Ising results, which resemble dilute AuFe. Our Fig. 1(c) shows that weak microscopic DM anisotropy alone does not yield the sharp magnetization reversals seen experimentally. Similar results hold for the uniaxial case which is plotted in Fig. 2 ($J_0=0$) at $T=0.2$. However, once $J_0>0$, we find sharp magnetization reversals. These are shown in the spin-glass phase for uniaxial anisotropy $D=0.4$ ($J_0=0.25, 0.50$) and in the ferromagnetic phase for $J_0=1.0$. Similar results are obtained for the DM case, although in this case the loops are found to be much narrower (by factors of 3 to 5). Furthermore, as expected, for the DM case we find that a larger value of J_0 is needed to obtain the same degree of sharpness of the magnetization reversal as in the uniaxial case. The notion

that some ferromagnetic interaction is necessary to explain hysteresis in CuMn has been previously invoked.[11] In AuFe a ferromagnetic tendency is unambiguously present and the change in the shape of the loop[10] as the Fe concentration increases corresponds roughly to the trends observed in Fig. 2 as J_0 increases. It should be noted, however, that the change in shape of the loop as a function of J_0 cannot be ascribed definitely to a change in this parameter only. It could derive from the *indirect* effect of J_0 through the value of the remanent magnetization, which was very large in the cases we considered.

A symmetrical field sweep, as shown in the figures, from large positive to equally large negative H values does not produce the so-called "displaced loop" characteristic of Mn-containing alloys.[4] Therefore, we have searched for displaced loops by starting at large positive H and reversing at a smaller negative value. Our results suggest that displaced loops are present *only* for the DM case. This is consistent with earlier claims[12] which suggested that displaced loops would occur for unidirectional anisotropy, as in the presence of DM interactions. While it was difficult to generate loops that looked similar to CuMn data, we did find, as expected, that in the second half of the cycle M changes sign before H.[5] Presumably the discrepancy between theory and experiment is due to the necessity of using large fields and to relaxation effects, some of which are not included here.

Phenomenological approaches[13] have proposed that the spin-glass free energy consists of a "macroscopic anisotropy" and a remanence term. Our conclusion here is that a necessary (but not sufficient) condition for obtaining remanence, and irreversibility in general, is that microscopic anisotropy be present; this can be tested for systems such as those discussed by Albrecht *et al.*[14] Therefore, it is unnatural for us to view the remanence and "macroscopic anisotropy" as independent mechanisms which separately contribute to hysteresis, etc. We note that our hysteresis curves can display either a smooth, gradual or a sharp boxlike shape. These two forms are often associated with the relative size of the macroscopic anisotropy and remanence term. A more relevant parameter, from our microscopic viewpoint, is the ratio of the microscopic anisotropy to the net ferromagnetic exchange interaction.

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