electric Power of Metals (Plenum, New York, 1976).
[11]G. Wedler and R. Chander, Thin Solid Films 65, 53 (1980).
[12]A. L. Efros and B. I. Shklorskii, J. Phys. C 8, L49 (1975); A. L. Efros, J. Phys. C 9, 2021 (1976); A. L. Efros, N. VanLien, and B. I. Shklorskii, Solid State Commun. 32, 851 (1979); B. Abeles, in *Applied Solid State Science*, edited by R. Wolfe (Academic, New York, 1976), Vol. 6, p. 1.
[13]R. C. Dynes and J. P. Garno, Phys. Rev. Lett. 46, 1371 (1981); W. L. McMillan and J. M. Mochel, Phys. Rev. Lett. 46, 556 (1981).

Monte Carlo Simulation of a Spin-Glass Transition

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Monte Carlo simulations show that for a system of Ruderman-Kittel-Kasuya-Yosida–coupled classical spins there exists an energy below which the system remains trapped near a single energy minimum. Provided that a small amount of anisotropy is introduced the system then exhibits, in the neighborhood of this energy, (a) a spin-glass-like peak in the susceptibility χ, (b) a well-marked maximum at $d^2\chi/dH^2$, and (c) evidence for spin freezing at lower energies. Without anisotropy these effects are absent.

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A satisfactory understanding of the spin-glass state in disordered magnetic systems with competing interactions remains elusive.[1] In this note we confine our attention to systems of isotropic, Ruderman-Kittel-Kasuya-Yosida (RKKY) coupled classical spins. For this case there exist a number of distinct spin configurations which minimize the energy locally.[2] These minima, almost degenerate in energy, are separated by energy barriers. At sufficiently low temperatures the thermodynamics of the system must be "broken" and controlled by the nature of the energy surface around a particular minimum. Such regions have very small curvature in certain directions, indicating that the ground-state configuration may be substantially distorted for a small cost in energy. If the barrier heights are widely distributed the various energy wells will merge progressively as the energy increases and the system will steadily migrate over wider regions of phase space. Alternatively, if the height distribution is narrow, there should be an abrupt effect upon the thermodynamics at a temperature where the average energy equals the barrier height. It is possible that this temperature marks the spin-glass "transition."

To examine these questions we have made Monte Carlo studies of a classical spin system with RKKY coupling for which ground-state configurations are known. The Hamiltonian is $\mathcal{H} = \sum J_{ij}\vec{S}_i\cdot\vec{S}_j$, where $J_{ij} = V_0\cos 2k_F r_{ij}/r_{ij}^3$. We find that there is a well-defined energy below which migration between energy wells ceases on the time scale of the simulation. The susceptibility in the neighborhood of this energy (temperature) passes through a maximum *provided that a small amount of anisotropic interaction is introduced,* but not otherwise. Spin freezing, as indicated by a time-independent value of the Edwards-Anderson parameter q,[3] is found under similar conditions. The temperature associated with this critical energy is lower than the transition temperature estimated from experiment by a factor of 3. This can be explained by the severe low-temperature distortion of the true energy-temperature relation by the use of Boltzmann statistics.

To examine the barriers separating energy minima we study the microcanonical low-field shattered susceptibilities χ_{sh}^{α} given by

$$\chi_{sh}^{\alpha}/\beta = \langle(\textstyle\sum_i \vec{n}_i\cdot\vec{n}_{i0}^{\alpha})^2\rangle_\mu, \tag{1}$$

where the unit vector spins $\vec{n}_i$ are a classical representation for a system of N spins $\vec{S}_i$ randomly sited on an fcc lattice.[4] The vectors $\vec{n}_{i0}^{\alpha}$, $i=1, \ldots, N$, represent the αth ground state of the system and $\langle\cdots\rangle_\mu$ denotes a microcanonical average. In Fig. 1 we show a plot of χ_{sh}^{α} values for all seven ground states of a system of $N=172$ spins[2] versus the internal energy ΔE of the system. The χ_{sh}^{α} values are seen to rise rapidly and quite uniformly as the energy is lowered. Below some point, however, the distribution of χ_{sh}^{α} values for different α's broadens rapidly. In this

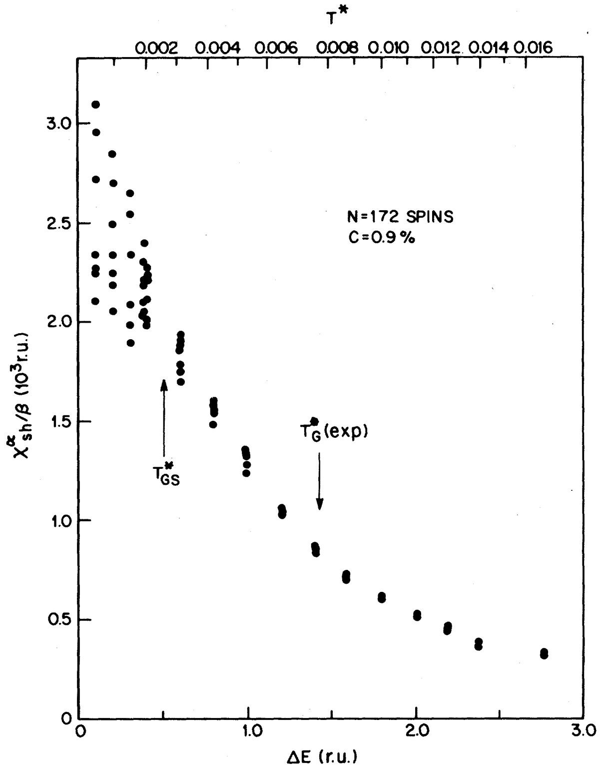

FIG. 1. Shattered susceptibilities $\chi_{sh}{}^{\alpha}/\beta$ evaluated with use of Eq. (1) (see Ref. 4) for all seven ground states of a distribution of 172 spins are plotted against internal energy ΔE, obtained from 5000-step microcanonical averages. The corresponding reduced temperature scale is shown at the top.

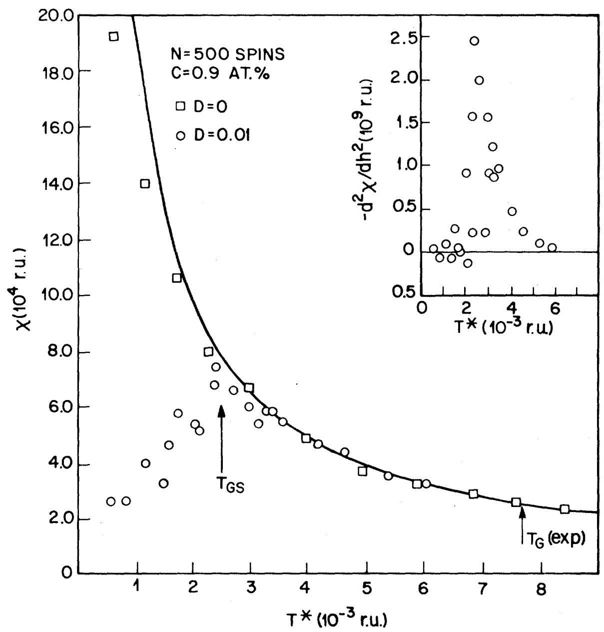

FIG. 2. Uniform susceptibility values obtained with Eq. (2) with use of 5000-step microcanonical averages are plotted against reduced temperature for a 500-spin sample both with and without anisotropy as shown. The solid curve shows Curie law behavior. Inset shows $d^2\chi/dH^2$ obtained concurrently with use of Eq. (3). D is in units of V_0.

region, examination of partial averages for the $\chi_{sh}{}^{\alpha}$'s during a 5000-step run shows that the system remains "trapped" near a single energy minimum, i.e., transitions between minima occur once at most. We refer to the point in energy or temperature below which this trapping occurs as the ground-state transition. This is indicated by $T_{GS}{}^*$ in Fig. 1, where the temperature scale is established by Boltzmann averages of the energy (top scale of the figure). Reduced temperatures are defined by $T^* = kTa^3/2\sqrt{2}\,V_0 S(S+1)$, where a is the fcc lattice constant. Also indicated in Fig. 1 is $T_G{}^*$,[5] showing $T_{GS}{}^* \sim \frac{1}{3}T_G{}^*$ as noted above.

If $T_{GS}{}^*$ is to be identified with the spin-glass transition, it is important to point out that the probable origin of the above discrepancy lies in the (unphysically) constant specific heat as $T^* \rightarrow 0$ yielded by Boltzmann statistics[6,7] in any classical treatment of the model described. In contrast, experimental values of the magnetic specific heat[8] drop sharply at low temperatures as required by the quantum-statistical behavior of the system near $T^*=0$.[2] The discrepancy between $T_{GS}{}^*$ and $T_G{}^*$ can be understood if we assume that the key variable is not the temperature, but the internal energy of the system. From our results for ΔE vs T^* we find that ΔE at $T_{GS}{}^*$ is about 90% of that of 0.88% Mn in Cu (Ref. 8) at $T_G{}^*$, in good accord with such an hypothesis.

Susceptibility behavior in the vicinity of $T_{GS}{}^*$ is illustrated in Fig. 2 with microcanonical averages[9] from a system of 500 spins. These data are obtained with

$$\chi/\beta = \tfrac{1}{3}[\langle\vec{N}\cdot\vec{N}\rangle_\mu - \langle\vec{N}\rangle_\mu\cdot\langle\vec{N}\rangle_\mu] \qquad (2)$$

($\vec{N} = \sum_i \vec{n}_i$), taking averages $\langle\cdots\rangle_\mu$ over runs of 5000 steps. We first note that without anisotropic interactions there is no transition except, perhaps, as $T^* \rightarrow 0$. With a small dipolar anisotropy effect included, one finds a susceptibility peak near $T_{GS}{}^*$ similar to those obtained with use of ac methods.[10] The anisotropic interactions are taken to be

$$E_{\rm dip} = -D{\sum_{i>j}}'(\vec{n}_i\cdot\vec{r}_{ij})(\vec{n}_j\cdot\vec{r}_{ij})/r_{ij}{}^5,$$

and are restricted here to nearest-neighbor pairs.

It directly involves only 10% of the spins in this particular sample. In Fig. 2 its coefficient D is taken to be $0.01\,V_0$.[5] Thus, the resulting anisotropy energy $\langle E_{\text{dip}}\rangle_\mu$ at T_{GS}* is (5–10)% of ΔE. Apparently the anisotropy acts here simply to stiffen the ground-state-like configurations against low-energy distortions. The value assumed for D is ~20 times larger than that for classical dipolar interactions, which is not an unrealistic anisotropy level for spin-glasses.[11] In connection with these results it should be noted that susceptibility peaks for Heisenberg systems *without anisotropy* have been reported both for nearest-neighbor[6] and RKKY models.[7] We have also observed peaks in χ/β near T_G*, but only for spatial distributions with low mean squared spin moment in the ground states. In other cases with larger than average ground-state moments χ/β has been found to rise for T* less than T_G*. Furthermore, spin freezing near T_G* could only be achieved with anisotropy of a magnitude larger than that of typical exchange fields. We conclude that this behavior is an accidental feature of small sample size and is unrelated to spin-glass ordering.

Experimentally, the spin-glass transition is characterized by a large anomaly in the nonlinear portion of the susceptibility.[10] We have examined this effect using the relation

$$\frac{d^2\chi_z}{dh^2}=\frac{\beta^3}{3}[\langle N_z{}^4\rangle_\mu-3\langle N_z{}^2\rangle_\mu-4\langle N_z{}^3\rangle_\mu\langle N_z\rangle_\mu+12\langle N_z\rangle_\mu{}^2\langle N_z{}^2\rangle_\mu-6\langle N_z\rangle_\mu{}^4], \tag{3}$$

with the result shown in the inset in Fig. 2. Indeed, one finds a sharp maximum in $d^2\chi/dh^2$ at the same temperature as the peak in χ. There are a number of seemingly conflicting data points in this plot. This is not only because good statistics for $d^2\chi/dh^2$ are difficult to achieve, but also one finds that different runs in which the energy is lowered in successive steps give peaks shifted in temperature by ~±10% relative to one another. This effect can also be seen in the plot of χ vs T. Its origin is apparently that the transition point can vary somewhat depending on the ground state into which the system falls for samples as small as this.

The magnitude of the peak in $d^2\chi/dh^2$ can be gauged by comparison with the following simple model of correlated fluctuations. If a system of N classical spins of moment μ is divided into clusters of uniform size n with cluster moments of $n^{1/2}\mu$ (reflecting random spin ordering within the clusters) and if, further, these clusters are free to fluctuate independently, then one has

$$\chi(H)=\frac{N\mu^2}{3kT}\left[1-\frac{n}{5}\frac{\mu^2H^2}{k^2T^2}+O(H^4)\right]. \tag{4}$$

In Eq. (4) the leading term is unaffected by clustering but $d^2\chi/dH^2$ is enhanced by a factor n.[12] The enhancement of $d^2\chi/dH^2$ may therefore be roughly interpreted as the size of clusters which fluctuate coherently. In the present case this would be limited by the sample size. Indeed, one finds a peak enhancement in the inset in Fig. 2 of ~400, so that at the transition almost the entire sample is fluctuating coherently. Experimentally,[10] one finds for 1% Mn in Cu at a temperature 1 K above the susceptibility peak an enhancement $n \sim 10^4$.

It has been clearly demonstrated that spin-glass freezing does not occur for the case of Heisenberg spins.[6] With the addition of a small anisotropy energy as described above, however, we find that definite spin freezing does occur over the time scale of the 5000-step runs carried out. Defining

$$q=N^{-1}\textstyle\sum_i\langle\vec{n}_i\rangle_\mu\cdot\langle\vec{n}_i\rangle_\mu\,, \tag{5}$$

we plot q vs T* in Fig. 3 for four values of dipolar anisotropy coefficient, including zero. In the latter case there is no measurable freezing at any temperature, the lowest-temperature point simply having a longer decay time than the others.

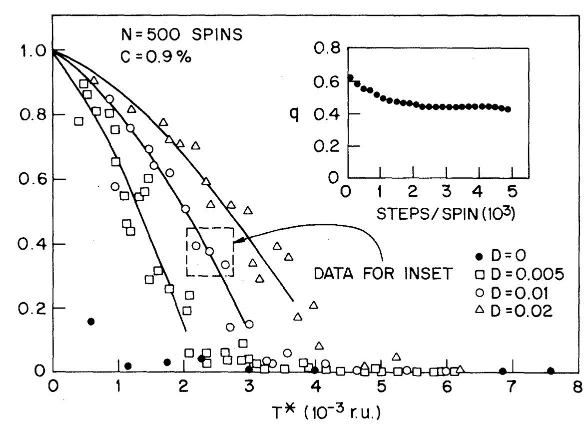

FIG. 3. The Edwards-Anderson spin freezing parameter q [Eq. (5)] is plotted against reduced temperature for a 500-spin sample with use of various levels of anisotropy as shown. Solid curves drawn are a guide for the eye. The variation of q over the course of a 5000-step run for typical data points is shown in the inset.

With nonzero anisotropy there is little decay of q over the run as illustrated in the inset in Fig. 3. Figure 3 shows, however, a significant increase in freezing temperature with added anisotropy, in contrast with the very slow increase observed for doped *Cu*Mn.[13] We believe this to be a consequence of our limited sample size, which requires greater anisotropy levels to produce freezing.

We conclude that a reasonable facsimile of the spin-glass transition is yielded by our numerical sample of 500 spins treated with Boltzmann and microcanonical averages. In spite of distortion of the low-temperature statistics resulting from the classical treatment, this approach appears to constitute an interesting laboratory for the examination of spin-glass phenomena. The importance of the inclusion of a small anisotropy to stabilize the spin-glass ordered state, a major conclusion, was anticipated by Anderson and Pond.[14] Further, it is evident that the transition is one of "trapping" the system in the vicinity of a particular energy minimum. While it may not be clear that such a trapping in one of many degenerate minima constitutes a phase transition, the observed enhancement of $d^2\chi/dh^2$ is evidence that the transition is a highly cooperative one. The observed coincidence of the susceptibility and ground-state transitions suggests that the basic mechanism involves trapping of the system within exchange-energy barriers. It would follow that quantum effects play a major role in determining the transition point, i.e., the temperature at which the internal energy reaches a value such that the system will escape this containment.

[1] For a discussion of numerical simulation studies of spin-glass models, see the review by K. Binder, in *Fundamental Problems in Statistical Mechanics V*, edited by E. G. D. Cohen (North-Holland, Amsterdam, 1980). In addition, recent theoretical progress on the infinite-range coupling model is summarized by G. Parisi and G. Toulouse, J. Phys. (Paris), Lett. 41, L361 (1980), and references therein.

[2] L. R. Walker and R. E. Walstedt, Phys. B 22, 3816 (1980), and Phys. Rev. Lett. 38, 514 (1977).

[3] S. F. Edwards and P. W. Anderson, J. Phys. F 5, 965 (1975).

[4] The response, $\Sigma_i \vec{n}_i \cdot \vec{n}_{ih}$, of a spin system to a set of unit magnetic fields, $\vec{n}_{ih}$, is referred to as a shattered susceptibility; it generalizes the familiar staggered susceptibility of a two-sublattice system. In actual calculations a rotationally averaged expression derived from Eq. (1) is used to avoid the possibility of incomplete Monte Carlo averaging over spin orientations.

[5] The reduced spin-glass temperature T_G* is taken from V. Cannella, in *Amorphous Magnetism*, edited by H. O. Harper and A. M. de Graaf (Plenum, New York, 1973), p. 195, with use of the value of V_0 obtained in Ref. 2.

[6] K. Binder, Z. Phys. B 26, 339 (1977).

[7] W. Y. Ching and D. L. Huber, J. Phys. F 8, L63 (1978).

[8] D. L. Martin, Phys. Rev. B 20, 368 (1979). T_G for this alloy has been estimated to be 11 ± 1 K from the data of Cannella, Ref. 4.

[9] Microcanonical averages have been found to give results similar to Boltzmann averages and are more economical to compute.

[10] Cannella, Ref. 4.

[11] F. Hippart, H. Alloul, and J. J. Préjean, Physica (Utrecht) 107B, 645 (1981); J. J. Préjean, M. J. Joliclerc, and P. Monod, J. Phys. 41, 1127 (1980).

[12] The effect of clustering on the field sensitivity of χ has been considered in Ref. 6.

[13] Adding 0.1 at.% Au to 0.3 at.% Mn in Cu increases T_G by ~ 5%. F. Milliken and S. Williamson, private communication.

[14] P. W. Anderson and C. M. Pond, Phys. Rev. Lett. 40, 903 (1978).