Antiferromagnetic spin-glass

James Shell, J. A. Cowen, and C. L. Foiles
Physics Department, Michigan State University, East Lansing, Michigan 48824
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We report data for a dilute magnetic alloy in which the average magnetic interaction is antiferromagnetic and a spin-glass transition occurs. Both this average interaction and the spin-glass temperature can be changed by altering the atomic order of the host. These data provide the first systematic confirmation of the validity of the left-hand portion of the Sherrington-Kirkpatrick phase diagram for dilute alloys.

Dilute alloys containing interacting magnetic moments continue to present challenging experimental and theoretical problems. One of the most interesting theoretical problems is the prediction of the phase diagram giving the boundaries between the paramagnetic, magnetically ordered (ferro- or antiferromagnetic) and spin-glass regimes. Several theoretical models, among them Sherrington and Kirkpatrick (SK),[1,2] self-consistent mean random field (MRF),[3] and Bethe-Peierls-Weiss (BPW)[3] give similar phase diagrams for spins with ferromagnetic interactions but appear to make no firm predictions for spins with antiferromagnetic interactions.

The SK model, for example, is formulated in terms of the average and the variance of a Gaussian distribution of interactions. Expressing these parameters in experimentally observed quantities Θ_c and T_{sg}, the Curie-Weiss temperature from high-temperature susceptibility data and the spin-glass temperature, respectively, the phase diagram takes the form shown in Fig. 1.[4]

When $\Theta_c/T_{sg} > 0$ ferromagnetism can occur and this portion of the diagram has been well documented experimentally in a variety of alloys. However,

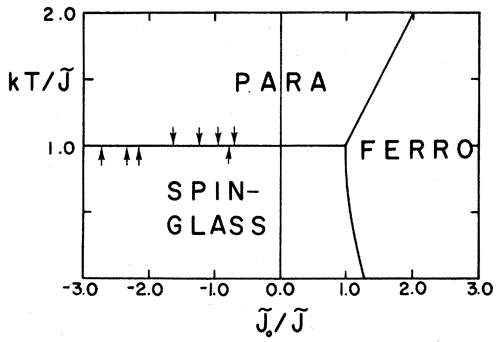

FIG. 1. The phase diagram predicted by the SK model with abscissa extended to −3.0. Upward pointing arrows denote ordered-state alloy data and downward pointing arrows denote disordered-state alloy data. Here $\tilde{J}_0/\tilde{J}$ is taken to be Θ_c/T_{sg}.

the status of the $\Theta_c/T_{sg} < 0$ portion is uncertain. Theoretical statements range from the initial implication that an antiferromagnetic structure analogous to the ferromagnetic structure occurs if the appropriate sublattice is chosen,[1,5] to a later specific claim that a spin-glass phase occurs at low temperature for all $\Theta_c < 0$[2], to an expressed doubt that spin-glass behavior actually occurs for $\Theta_c/T_{sg} < 0$.[6] In this paper we report magnetic data for a dilute magnetic alloy that indicates spin-glass behavior does occur as far as $\Theta_c/T_{sg} \cong -2.5$.

The alloy system $(Cu_3Pt)_{1-x}Mn_x$ has many properties which parallel those of the archetypical spin-glass sytem *Cu*Mn while it has the additional possibility of altering the Mn-Mn interactions at a *fixed concentration* by changing the degree of atomic order in the host Cu_3Pt. In its atomically disordered state Cu_3Pt has (i) an fcc crystal structure, (ii) an electrical resistivity comparable to that of Cu, (iii) an electronic specific heat within 25% of that for Cu, and (iv) a diamagnetic susceptibility.[7,8] In its atomically ordered state Cu_3Pt (i) retains an fcc array of lattice sites, but the chemical order among these sites causes a sc crystal structure with a basis of four atoms per unit cell, (ii) has an electrical resistivity twice that of Cu, but less than 25% that of Pt, and (iii) is even more diamagnetic.[7,8] All of these properties indicate that Cu_3Pt is an alloy host comparable to Cu. The remainder of this paper is concerned with the magnetic properties of $(Cu_3Pt)_{1-x}Mn_x$ and the changes in these properties as atomic order is changed while x is maintained constant.

We consider the atomically disordered samples first. Magnetization measurements for all these alloys at 4.2 K were linear in applied field up to 0.9 T. The dc susceptibility as a function of temperature was determined by measuring the magnetization at 0.9 or 0.5 T from 4.2 to 77 K. The resultant χ_{dc}^{-1} are given in Fig. 2(a). A Curie-Weiss form is obeyed over most of the temperature range and the resultant parameters are listed in Table I. Deviations do occur at lower temperatures and these deviations parallel those in *Cu*Mn which are indicative of spin-glass

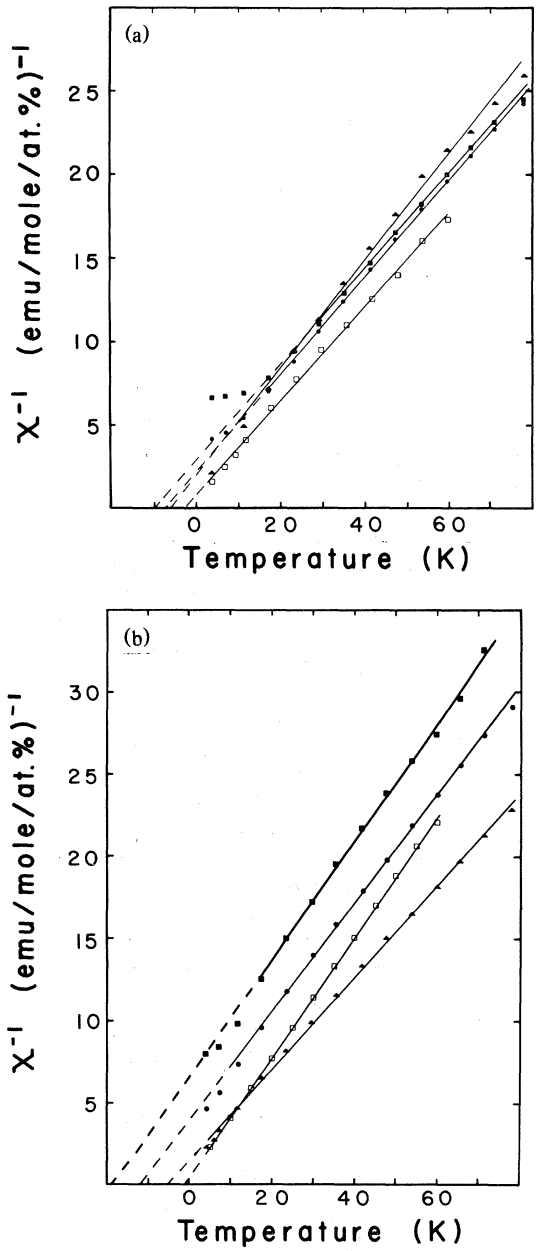

FIG. 2. High-field dc impurity susceptibiliy. Dark squares, circles, triangles, and open squares correspond to 3.03, 1.85, 0.81, and 0.5 at.% Mn, respectively. The solid line is a least-squares fit to data points taken with $T \geq 2T_{sg}$. The dashed portion is merely a straight-line extension. (a) Disordered host data. (b) Ordered host data.

behavior. Θ_c is a measure of the average Mn-Mn interaction. For our disordered $(Cu_3Pt)_{1-x}Mn_x$ samples Θ_c is negative, indicating that the average interaction is antiferromagnetic, and becomes more negative as a linear function of x within experimental error. This behavior should be contrasted with Θ_c for *Cu*Mn which is also linear in x but becomes more positive.

TABLE I. Experimentally determined parameters for ordered and disordered Cu_3PtMn alloys.

Alloy (at.% Mn)	p_{eff} (μ_B)	Θ_c (K)	T_{sg} (K)
Disordered			
0.5	5.3 ±0.3	−2.3 ±0.5	1.85 ±0.1
0.81	5.0 ±0.3	−5.4 ±0.9	3.3 ±0.2
1.85	5.2 ±0.3	−7.0 ±0.4	7.5 ±0.2
3.03	5.3 ±0.3	−9.7 ±0.4	13.6 ±0.2
Ordered			
0.5	4.7 ±0.3	−1.25 ±0.6	1.55 ±0.1
0.81	5.3 ±0.3	−5.0 ±0.5	2.3 ±0.2
1.85	4.9 ±0.3	−11.8 ±0.5	4.3 ±0.2
3.03	4.7 ±0.3	−18.3 ±0.5	7.8 ±0.2

The spin-glass temperature was measured using two different techniques. In the temperature range $1.25 < T < 4.2$ K, T_{sg} was determined by observing the cusp in the low-field ac susceptibility. For temperature above 4.2 K, T_{sg} was determined by observing the change in slope of χ_{dc} vs T measured on a sensitive superconducting quantum interference device (SQUID) susceptometer.[9] Using the ac system cusps in χ were found for the two more dilute samples (the resultant T_{sg} values are listed in Table I), while χ_{ac} for the more concentrated samples was temperature independent in this temperature range. Normalized χ_{ac} data proportional to χ per Mn atom are given in Fig. 3.

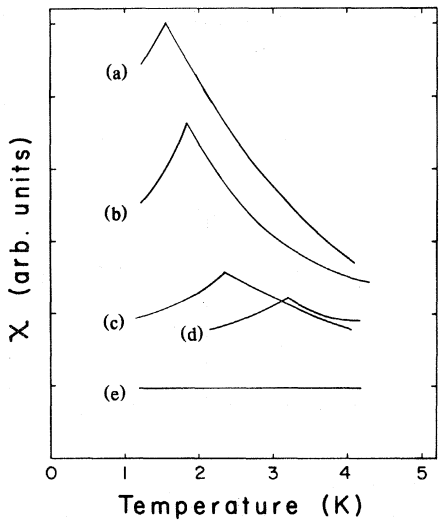

FIG. 3. Low-field ac susceptibility data. Curves (a) and (b) correspond to 0.5 at.% Mn ordered and disordered respectively. Curves (c) and (d) correspond to 0.81 at.% Mn ordered and disordered, respectively. Curve (e) corresponds to 1.85 at.% Mn ordered and disordered.

Careful heat treatment of these disordered alloys produces an atomic order which is confirmed by the appearance of additional Bragg peaks, called superlattice lines, in x-ray scattering from the alloys. A quantitative measure of the long-range order, which is denoted by S, can be obtained from the integrated intensities of these superlattice lines. S is one for a fcc unit cell with only Cu atoms on all face-centered sites and only Pt atoms on all corner sites, but is zero for a random distribution of Cu and Pt atoms in these sites. The long-range order of the present alloys has been measured[10] with the following results. The disordered alloys gave no indication of superlattice lines (and thus $S \cong 0$) and the ordered alloys had $S \gtrsim 0.85$. The variation of S with x was consistent with the Mn atoms being substitutional impurities on all sites or on all ordered Cu sites (75% of all sites) and proved that the Mn impurities were not restricted to ordered Pt sites. Thus, our present interpretation of magnetic data is based upon the assumption that these alloys are composed of an ordered Cu_3Pt host with random Mn impurities.

The introduction of atomic order causes significant changes in magnetic properties. Magnetization curves at 4.2 K for the atomically ordered samples are once again linear in applied field up to 0.9 T. The χ_{dc}^{-1} data obtained from $M(T,H=0.9$ or 0.5 T) are plotted in Fig. 2(b) and the associated parameters are given in Table I. Now the Curie-Weiss form is obeyed down to 4.2 K and low-temperature deviations are absent. Θ_c has increased in absolute value while remaining negative and essentially linear in x. The effective moment per Mn atom for the atomically ordered samples appears to be generally lower than that for the disordered samples. (Note that the $x=0.81$ at.% pair is the only exception to this pattern.) However, the similarity in p_{eff} values for the atomically disordered samples to those found in *Cu*Mn and the fact that the slightly lower p_{eff} values in the atomically ordered samples are consistent with each Mn atom having a spin of approximately 2 give no suggestion of unusual behavior.

Normalized χ_{ac} data for the atomically ordered samples are given in Fig. 3. The two more dilute samples exhibit a cusp with the associated T_{sg} values listed in Table I; for each ordered sample T_{sg} is lower than the value for the corresponding disordered sample. The more concentrated samples gave no evidence of spin-glass behavior in χ_{ac}, which was essentially temperature independent from 1.26 to 4.2 K.

Although there are unresolved low-field calibration problems with the SQUID system the temperature dependence of the relative χ's gave clear evidence of spin-glass behavior. Data for samples cooled in 0 ± 10 Oe and subsequently measured in 10 ± 10 Oe had clear discontinuities in temperature dependence which were comparable to the results found by Nagata *et al.*[11] for *Cu*Mn samples cooled in 5.09 Oe. The work of Nagata *et al.* proved that measurements in these weak fields gave reliable values for T_{sg} and thus we include in Table I the T_{sg} values found by this method.

No explicit calculation exists for the effect of a host order-disorder transition upon the magnetic properties of a spin-glass. In the present case the atomic order would be expected to introduce new zone gaps in the electron energy spectrum and thereby alter the behavior of conduction electrons. However, strong similarities in equilibrium and transport properties for Cu_3Pt and Cu were noted above and these similarities convince us that no fundamentally new considerations have been introduced. Thus, we will continue to use *Cu*Mn as an experimental prototype and we will use the theoretical SK model to interpret magnetic behavior.

The magnetic behavior of both atomically disordered and ordered $(Cu_3Pt)_{1-x}Mn_x$ is consistent with a spin-glass whose average magnetic interaction is antiferromagnetic, $\Theta_c < 0$. To the best of our knowledge, these are the first data for a dilute magnetic alloy in which the absolute value of Θ_c and its variation with concentration are consistent with an average antiferromagnetic interaction.[12] Thus, these data both confirm the validity of and explore the nature of the left-hand portion of Fig. 1 for dilute magnetic alloys. At higher concentrations, samples of atomically ordered $(Cu_3Pt)_{1-x}Mn_x$ have $|\Theta_c|_{\text{ord}} > |\Theta_c|_{\text{disord}}$, have $(T_{sg})_{\text{ord}} < (T_{sg})_{\text{disord}}$ and thus they seem to be spin-glasses which extend this experimental confirmation to even more negative values of Θ_c/T_{sg}. We suggest that these data establish two features about the antiferromagnetic portion of the SK phase diagram:

(1) First and foremost, spin-glass behavior does occur in dilute magnetic alloys when the average interaction is antiferromagnetic.

(2) Unlike the ferromagnetic portion of the phase diagram, the condition $|\Theta_c| = T_{sg}$ need not be associated with the onset of magnetic order. Present data suggest that $|\Theta_c| \sim 2T_{sg}$ without loss of spin-glass behavior.

ACKNOWLEDGMENTS

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[1]D. Sherrington and S. Kirkpatrick, Phys. Rev. Lett. 35, 1792 (1975).

[2]S. Kirkpatrick and D. Sherrington, Phys. Rev. B 17, 4384 (1978).

[3]M. W. Klein, L. J. Schowalter, and P. Shukla, Phys. Rev. B 19, 1492 (1979).

[4]Formally the SK phase diagram was developed and presented in terms of the average and the variance of the interactions. However, it has become common practice to use T_c (or Θ_c) and T_{sg} as being synonymous, respectively, to the preceeding interaction parameters. This practice is followed in the present paper and was justified by D. Sherrington and B. W. Southern, J. Phys. F 5, L49 (1975), and by B. Southern, J. Phys. C 8, L213 (1975).

[5]No explicit consideration of the antiferromagnetic portion of Fig. 1 was given in Ref. 1 beyond the single sentence: "For interactions which are on the average antiferromagnetic or which include second-neighbor terms, analogous equations result in which m (an average magnetization) is replaced by the appropriate sublattice magnetization."

[6]S. Geschwind *et al.*, J. Phys. (Paris) 41, C5-105 (1980).

[7]C. L. Foiles, in *Thermal Conductivity,* edited by P. G. Klemens and T. K. Chu (Plenum, New York, 1976), Vol. 14, p. 97.

[8]C. L. Foiles, in *Magnetism and Magnetic Materials—1976,* edited by J. J. Becker and G. H. Lander, AIP Conf. Proc. No. 34 (AIP, New York, 1976), p. 17.

[9]Model 800 VTS susceptometer, S.H.E. Corporation, San Diego, Calif.

[10]C. L. Foiles, in *Magnetism and Magnetic Materials—1976,* edited by J. J. Becker and G. H. Lander, AIP Conf. Proc. No. 34 (AIP, New York, 1976), p. 358.

[11]S. Nagata, P. H. Keesom, and H. R. Harrison, Phys. Rev. B 19, 1633 (1979).

[12]*Cu*Mn alloys with Mn concentrations below $\frac{1}{2}$ at.% provide an interesting contrast in absolute value and variation of Θ_c. In *Cu*Mn Θ_c always becomes more positive with increasing Mn concentration, but the complication of $\Theta_c < 0$ for Mn $< \frac{1}{2}$ at.% has long been present and was recently discussed by C. A. M. Mulder *et al.*, Phys. Rev. B 23, 1384 (1981). In a later, related paper [A. F. J. Morgownik and J. A. Mydosh, Phys. Rev. B (in press)] it is suggested that $\Theta_c = -6.9 + 10.4x$, where Θ_c is in K and x is the Mn concentration in atomic percent. With this interpretation, all negative values of Θ_c arise from the negative constant. Both the origin and relation of this constant to magnetic interactions, and thus to the physical effect underlying the SK model, are unclear and require further study.