Effect of Hydrostatic Pressure on the Magnetic Susceptibility of a Ag:Mn Spin-Glass

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The magnetic susceptibility of a Ag–3-at.%-Mn spin-glass has been measured under hydrostatic pressure to 15 kbar. The spin-glass freezing temperature T_0 increases with pressure at the rate $dT_0/dP \simeq +42 \pm 4$ mK/kbar. This pressure dependence is clear evidence that the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, and not the magnetic dipole interaction, is responsible for spin-glass freezing at T_0. The present results thus support those models of spin-glass freezing which are based solely on RKKY interactions between spins.

Spin-glasses as such have received wide experimental[1] and theoretical[2] attention for the better part of a decade. In spite of this effort, the precise nature of the spin-glass transition remains a mystery. There is, in fact, uncertainty whether or not there really is a *single* type of transition common to the multitude of metallic and insulating "spin-glasses" known today. Two basic but opposing views of the spin-glass transition seem to have emerged: (1) In the so-called *phase-transition picture*,[2] a single species of mutual spin-spin interactions [assumed to be Ruderman-Kittel-Kasuya-Yosida (RKKY) in metallic systems or superexchange in insulating systems] causes the spins to cooperatively freeze in place below a critical temperature T_0, leading to the characteristic ac susceptibility peak at T_0 and the time-dependent remanence effects for $T < T_0$. For metallic systems, we hereafter call this the "RKKY phase-transition model." (2) In the *cluster-blocking picture*,[3] on the other hand, the spin system is envisaged as being broken up into a multitude of noninteracting or weakly interacting superparamagnetic clusters of various sizes. These clusters are formed at temperatures well above T_0 by the mutual RKKY or other interactions between their spins. The actual spin-glass transition at the lower temperature T_0 is believed to originate from the progressive blocking of the clusters in certain fixed directions dictated by anisotropic energy barriers set up by a second type of interaction mechanism, usually assumed to be magnetic dipolar in origin. We hereafter call this the "dipolar cluster-blocking model."

Irrespective of which, if either, model is correct, the precise nature of the spin-glass transition in a given class of systems is certainly, at some level of understanding, sensitive to the details of the spin-spin interaction mechanism(s) which is (are) operative. To cite an example, in Monte Carlo computer simulations,[4] even such an overriding question as the occurrence or nonoccurrence of a phase transition appears to depend on whether a Gaussian or a $\pm J$ nearest-neighbor interaction distribution is assumed. It is also an interesting question whether or not the same interaction mechanism(s) responsible for the freezing at T_0 is (are) also responsible for the time-dependent remanence effects at lower temperatures. In the face of the present-day confusion as to the nature of the spin-glass transition, it would seem to be of value to "return to the basics" and attempt to establish experimentally which interaction mechanisms are actually important for the spin-glass behavior in a given class of systems.

In the case of metallic spin-glasses, the RKKY or dipolar interactions are expected to change in substantially different ways if the sample is subjected to high pressures.[5] A measurement of the pressure dependence $T_0(P)$ thus has the potential to reveal which of these two interaction mechanisms, if either, is responsible for spin-glass freezing. It should be noted that studies measuring $T_0(c)$ as a function of the impurity concentration c are not able to directly distinguish between dipolar or RKKY interactions, to the extent that both depend on R^{-3}, where R is the separation between two spins.

We report here the first high-pressure susceptibility measurements on a "classical" spin-glass. The spin-glass freezing temperature of Ag + 3 at.% Mn is found to increase with pressure at a rate which is in excellent agreement with that estimated[5] for the RKKY interaction, but nearly *four times faster* than that possible for a dipolar interaction. This allows the conclusion that in this system the RKKY interaction, and not the dipolar coupling, is the dominant interaction mechanism leading to spin-glass freezing at T_0. The present results thus support those models of spin-glass freezing, for example, the RKKY phase-transition model, which are based solely

on RKKY interactions between spins.

The Ag–3-at.%-Mn sample was melted inductively under high vacuum on a water-cooled copper boat and given a homogenizing anneal at 900°C for 15 h before being quenched in water. The 90-g binary Cu-Be pressure clamp[6] used to generate hydrostatic pressure[7] is suspended in the center of a massive oxygen-free high-conductivity copper tube, ensuring homogeneity and stability of the temperature to well within 0.05 K.[8] The dc magnetic susceptibility is determined in a Faraday magnetometer employing superconducting gradient (800 Oe/cm) and main (70 kOe) coils. Only the gradient coil with 400 Oe/cm is used for the measurement of the susceptibility peak at T_0. Fortunately, the contribution to the susceptibility from the pressure clamp alone is relatively small, its subtraction having no measurable effect on the value of T_0. Likewise, T_0 takes on the same value whether the small dc field is switched on above T_0 before cooling or below T_0 before warming, even though in the latter case the susceptibility peak is much more symmetric about T_0. Alternatively, to measure the time decay of the thermoremanent magnetization, a main coil field of 35 kOe is applied at a temperature 1 K above T_0 ($T_0 = 12$ K for $P = 0$), the temperature lowered to 7.5 K, and the main field then removed. Details of the high-pressure magnetometer used in these studies will be published elsewhere.[10]

In Fig. 1 the dc-field-cooled magnetic susceptibility of Ag:Mn in a mean applied field of 140 Oe is plotted as a function of temperature for different pressures.[11] The T_0 value at a given pressure is the same within 0.03 K for three separate temperature runs. The susceptibility peak is seen to shift to higher temperatures with increasing pressure in a reversible manner at the rate $dT_0/dP \simeq +42 \pm 4$ mK/kbar which corresponds to $d\ln T_0/d\ln V \simeq -3.6 \pm 0.3$, where V is the atomic volume.[12] Identical results were attained in a second pressure run after reannealing the sample. If the spin-glass freezing were due solely to a magnetic dipole interaction, then one would expect, because this interaction is inversely proportional to the atomic volume V, that $d\ln T_0/d\ln V = -1$. The present measurements thus show that a dipole interaction alone definitely cannot be responsible for spin-glass freezing in dilute Ag:Mn. We estimate the anticipated pressure dependence of T_0 for the RKKY interaction from the relation $T_0 \propto J_{\mathrm{RKKY}}{}^2 N(E_F)$,[5,13] where J_{RKKY} is the RKKY-interaction parameter and $N(E_F)$ is the density of states at the Fermi surface. We thus obtain for the pressure (volume) dependence of T_0:

$$\frac{d\ln T_0}{d\ln V} = 2\frac{d\ln|J_{\mathrm{RKKY}}N(E_F)|}{d\ln V} - \frac{d\ln N(E_F)}{d\ln V}. \qquad (1)$$

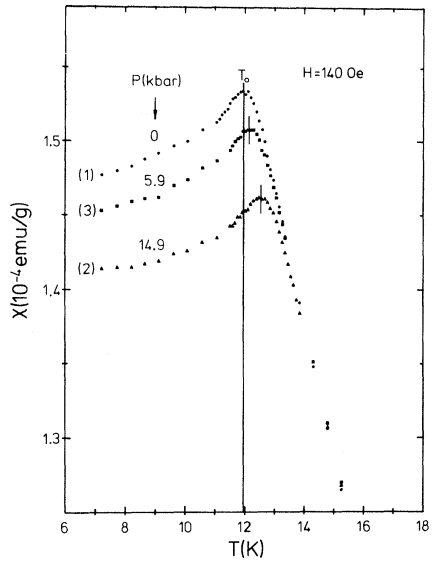

FIG. 1. Field-cooled dc magnetic susceptibility of Ag + 3 at.% Mn vs temperature for three pressures (see Ref. 11). Data points are averages at a given temperature over three separate runs. Numbers in parentheses give the order of measurement. A final measurement at ~ 1.9 kbar, omitted for clarity, has same value of T_0 and essentially identical temperature dependence as measurement (1). The spin-glass freezing temperature T_0 shifts *reversibly* to higher temperatures with pressure at the rate $dT_0/dP \simeq +42 \pm 4$ mK/kbar.

J_{RKKY} is given by the expression $J_{\mathrm{RKKY}} = \sum_l (2l+1) \times (-1)^l J_l$, with the assumption of scattering on a spherical Fermi surface.[5,13,14] For 3d impurities only the first three J_l angular momentum components are believed to be important,[5,14] i.e., $J_{\mathrm{RKKY}} \simeq J_0 - 3J_1 + 5J_2$. If we assume that the $5J_2$ term is dominant because of the presence of the large mixing-exchange contribution in the $l = 2$ partial wave for a 3d impurity,[5,14] which causes J_2 to be negative for Ag:Mn and leads to the well-known Kondo effect, then $J_{\mathrm{RKKY}} \simeq 5J_2$. The pressure (volume) dependence of $J_2 N(E_F)$ is known from previous work on Kondo alloys of extreme dilution (Ag + 10 ppm Mn),[5,15] which give $d\ln|J_2 N(E_F)|/d\ln V \simeq -1.3$. Thermal expansion studies[16] on pure Ag give $d\ln N(E_F)/d\ln V \simeq +1$, which differs slightly from the free-electron value of $+\frac{2}{3}$. Equation (1) can now be evaluated to give $d\ln T_0/d\ln V = 2(-1.3) - 1 = -3.6$. This projected value of

$d\ln T_0/d\ln V$ is identical to the value -3.6 ± 0.3 obtained in the present experiment on a Ag–3-at.%-Mn spin-glass! This excellent agreement, while not proving the dipole interaction to be completely inoperative, is clear evidence for both the overriding importance of the RKKY interaction in spin-glass freezing and the dominance of the J_2 component in this interaction. This leads to the perhaps surprising conclusion that the same negative s-d interaction between impurity and conduction electron spins, which leads to the Kondo effect in extremely dilute Ag:Mn, is also responsible for spin-glass freezing in more concentrated Ag:Mn. This very assumption is, in fact, made in various calculations of the properties of the so-called Kondo lattice.[17] That $J_{RKKY} \simeq 5J_2$ is also inferred by Davidov *et al.*[18] for Ag:Mn, who estimate that the J_0 and J_1 terms nearly cancel out. Wu *et al.*[19] recently reported an increase of T_0 with pressure, $dT_0/dP \simeq +30$ mK/kbar, in a Pd:FeMn alloy exhibiting both spin-glass and ferromagnetic behavior. The similarity of this pressure dependence to that found here for Ag:Mn is probably coincidental, since in Pd:FeMn, in contrast to Ag:Mn, the density of states of the host matrix at E_F exhibits a very large pressure dependence.[20]

The time dependence of the thermoremanent magnetization σ_{TRM} of Ag:Mn was measured at a fixed temperature (7.5 K) as a function of pressure. Within experimental accuracy, no change in the slope α of the $\log\sigma_{TRM}$ versus log(time) curves with pressure could be observed, allowing only an estimate of the upper bound $|d\ln\alpha/d\ln V| \leq 4$. Monte Carlo calculations[21] using a Gaussian nearest-neighbor interaction distribution predict that $\alpha \propto T_0^{-1}$ which implies $d\ln\alpha/d\ln V = -d\ln T_0/d\ln V \simeq +3.6$, with use of the above pressure dependence of T_0. It would be expected that this relation would hold if a more realistic RKKY interaction were used in the Monte Carlo simulations. From the dipolar cluster-blocking model, on the other hand, one would expect $d\ln\alpha/d\ln V = +1$. The present thermoremance experiments are thus not able to distinguish between RKKY or dipolar models, being consistent with both. An appreciable extension of the pressure range should allow a critical test.

The present high-pressure susceptibility measurements on Ag:Mn are in excellent agreement with previous studies of the resistivity maximum under pressure on Ag:Mn (Refs. 5 and 15) and numerous other spin-glasses.[5] The temperature of the resistivity maximum T_M is shown in a theory by Larsen[22] to be a rather complicated function of both the rms interaction strength Δ_c and the Kondo temperature T_K. The high-pressure resistivity studies constitute a stringest test of this theoretical functional dependence and allow the prediction[5] that $d\ln T_0/d\ln V = d\ln\Delta_c/d\ln V \simeq -3.3$ to -4.2 for Ag:Mn, which is confirmed in the present experiment. In view of this excellent agreement, it would seem reasonable to assume that, as indicated by the resistivity studies, Ag:Mn behaves under pressure in a manner shared by a wide variety of other "classical" spin-glasses combining noble-metal hosts with transition-metal impurities. The conclusions reached here for Ag:Mn are thus expected to apply to systems such as Au:Mn, Cu:Mn, and Au:Fe, as well. It is also noteworthy that the present results are in remarkable agreement with thermal expansion studies on a Ag–1-at.%-Mn alloy[23] where, assuming that T_0 is proportional to the mean magnetic interaction energy, it can be estimated that $d\ln T_0/d\ln V \simeq -3.7$.

The present experiments give support to the RKKY-interaction models, for instance the RKKY-phase-transition model or Smith's giant-cluster percolation model,[24] of spin-glass freezing and show that a dipole cluster-blocking model, where the number of spins in a given cluster is assumed pressure independent, must be incorrect. One could, of course, argue that in the latter model the number of spins in a cluster should increase as the RKKY interaction increases with pressure. Such a behavior, however, would be directly opposed to the spirit of the cluster-blocking model itself where essentially independent noninteracting spin clusters are required. Certainly if a given spin can *enter* a cluster at high pressures then it is *strongly interacting* with, though outside, this cluster at ambient pressure! Recent zero-field NMR work on Cu:Mn (Ref. 25) and high frequency ac susceptibility studies on Ag:Mn (Ref. 26) also are inconsistent with the cluster-blocking model. Rather than artificially breaking up a spin-glass into strongly interacting clusters or clouds, which are supposed to form via RKKY interactions and freeze via dipole interactions, it would seem to be more reasonable to choose as a starting point a simpler model to describe spin-glass behavior, one which allows all spins to interact freely with each other via a single (RKKY) interaction mechanism. Only if this simple model is proven inadequate would it seem justified to consider a more complicated physical model requiring multiple interaction mechanisms.

Further theoretical work is badly needed to give a clear answer to the question of whether or not the simple RKKY models are adequate to describe spin-glass behavior.

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