Effect of Gold Impurities on the Critical Properties of *Cu*Mn Spin-Glasses

Y. Yeshurun and H. Sompolinsky
Department of Physics, Bar-Ilan University, Ramat-Gan, Israel
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We present SQUID measurements of the nonlinear susceptibility χ_{nl} of "pure" *Cu*Mn and *Cu*Mn doped with gold impurities with concentrations c_{Au} of 1, 2, and 3 at.%. Gold impurities suppress the magnitude of χ_{nl}. Ordinary one-parameter scaling with respect to field and temperature is obeyed by χ_{nl} of the pure *Cu*Mn. However, strong deviations from this scaling behavior are observed in the gold-doped samples in the vicinity of the freezing temperature and in low fields, $H < H^*$ where H^* increases with c. This is interpreted as the onset of an anisotropy-induced crossover from a Heisenberg to an Ising spin-glass critical behavior.

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In this Letter we present the first experimental evidence that random-spin anisotropy strongly affects the nonlinear magnetic susceptibility χ_{nl} of spin-glasses at and above their freezing temperature T_g. Dilute metallic spin-glasses such as *Cu*Mn contain random anisotropic interactions which are, however, very weak compared to the Ruderman-Kittel-Kasuya-Yosida exchange.[1] The addition of nonmagnetic impurities such as Au, Pt, and Ni, in low concentrations, greatly enhances the random anisotropy without affecting significantly the exchange energy.[1–3] Theoretically it has been predicted that adding a weak random anisotropy to an (otherwise) isotropic spin-glass (SG) will have an important effect on the SG phase transition. In particular, the anisotropy induces a crossover from a Heisenberg to an Ising critical behavior.[4–6] A natural means for studying the critical properties of the spin-glass transition is the nonlinear magnetic susceptibility χ_{nl}, which plays a role analogous to the linear susceptibility in ferromagnetic transitions.[7–9] The study of χ_{nl} above T_g has the important advantage of being relatively free of the strong relaxation processes which set in below T_g.

In this work we have measured χ_{nl} as a function of temperature T and static magnetic field H of 1.2-at.% *Cu*Mn, doped with gold impurities of 1, 2, and 3 at.%. We have observed that the Au impurities strongly suppress the magnitude of χ_{nl} and modify its properties, whereas their effect on the total magnetization is negligible. Performing a scaling analysis of $\chi_{nl}(T,H)$ we find that (i) in high fields or temperatures the scaling properties of χ_{nl} are not affected by the Au impurities; (ii) in the regime of low fields and T close to T_g, χ_{nl} of the impurity-doped samples deviates significantly from a simple one-parameter scaling behavior. The deviations may be a manifestation of the abovementioned Heisenberg-Ising crossover.

Another critical property of the SG transition which is expected to be sensitive to Au impurities is the shape of the critical line $T_c(H)$ as $H \to 0$. Indeed, recent torque measurements[10] reveal that the addition of Au impurities changes significantly the shape of $T_c(H)$ in *Cu*Mn. Both the torque experiment and the present experiment yield the same order of magnitude of crossover fields below which anisotropy effects are seen. However, the observed scaling behavior of χ_{nl} is not entirely consistent with the experimental results for $T_c(H)$, as will be discussed below.

We used a SQUID susceptometer to measure the zero-field–cooled magnetization M of these alloys in the temperature range 6 K $\leq T \leq$ 40 K and in fields 20 Oe $\leq H \leq$ 44 kOe. The magnetization curves of these samples are not linear in H. To determine the linear susceptibility $\chi_0(T)$ we linearly extrapolate the measured M vs H values for $H \leq 50$ Oe down to zero field. We find that χ_0 is reduced by a few percent as the number of gold impurities increases. This reduction can be accounted for by the decrease in the number of Mn atoms per gram as a result of gold dilution. We use $\chi_0(T)$ to evaluate the nonlinear susceptibility $\chi_{nl} = \chi_0 - M/H$. The results of χ_{nl} as a function of T for $H = 5$ kOe are shown in Fig. 1(a). Clearly the Au impurities suppress considerably the nonlinear magnetization. This is observed in the whole range of fields of our experiment, but the effect becomes smaller with increase of H and T. It is interesting that the Au impurities do not change significantly the locus of the maximum of $\chi_{nl}(T)$ which is at $T = 11.2 \pm 0.2$ K for the field range $H \lesssim 20$ kOe [Fig. 1(b)].

A most important effect of the Au impurities is on the field dependence of χ_{nl}. In the pure *Cu*Mn, the $T = 11.2$ K isotherm follows a power law

$$\chi_{nl}(H) \propto H^{2/\delta}, \quad \delta \simeq 5.0 \pm 0.3, \tag{1}$$

as seen in Fig. 2. This behavior is consistent with the identification of $T_g = 11.2$ K as the SG critical temperature for the pure *Cu*Mn. On the other hand, $\chi_{nl}(H)$ of the doped samples does not obey a power law in the same range of fields, at any temperature. The smallest deviations from a power law are found in the $T \cong 11.2$ K isotherms which are displayed in Fig. 2. The results clearly show that in the high-field regime ($H \gtrsim 10$ kOe) the isotherms of the doped samples are very similar to each other and to that of the

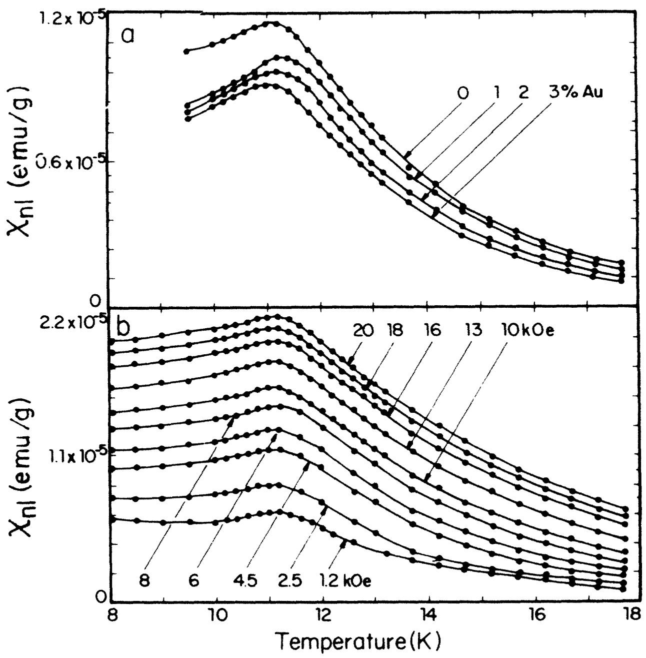

FIG. 1. Temperature dependence of the nonlinear susceptibility for $CuMnAu_x$ alloys. (a) $H = 5$ kOe and x as indicated. (b) $x = 0$ and fields as indicated.

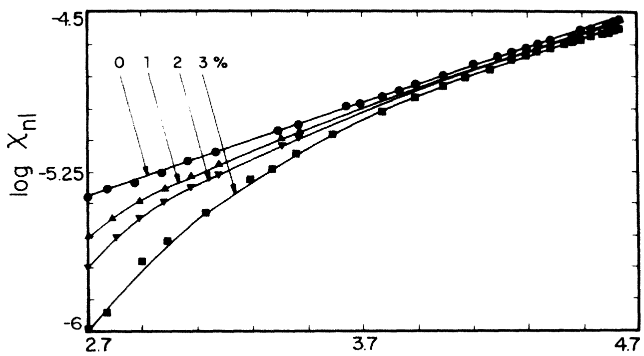

FIG. 2. Field dependence of the nonlinear susceptibility (shown on a log-log scale) at 11.2 K for the $CuMnAu_x$ alloys. The lines serve to guide the eye only. For clarity, high-field data are not exhibited for $x = 1$ and 2 at.% Au.

pure *Cu*Mn: They all follow the same power law as in Eq. (1). In low fields, the doped samples exhibit systematic deviations from a power law which increase with increasing concentration of gold. These deviations are also increased for isotherms above and below 11.2 K. In the absence of a power-law behavior at low fields it is impossible to identify the critical temperature of the doped samples. However, our results (including those of Fig. 2) suggest that T_g is close to 11 K in all samples; namely that the changes in T_g due to the Au impurities are relatively small. This conclusion is consistent with recent experimental results of Vier and Schulz.[11]

A more comprehensive analysis of the critical properties of $\chi_{nl}(T,H)$ should involve scaling. In the absence of anisotropy, the singular part of χ_{nl} is expected to have a single-parameter scaling form,[7-9]

$$\chi_{nl}(T,H) = h^{2/\delta} f(t/h^{2/\phi}), \tag{2}$$

where h is the reduced field $h = \mu H/k_B T_g$ and t is the reduced temperature $t = 1 - T/T_g$. Equation (1) of course assumes the existence of a continuous phase transition at a finite temperature T_g. The exponents ϕ and δ are related to other critical exponents via the scaling relations $\phi = \gamma + \beta$, $\delta = 1 + \gamma/\beta$. In the case of zero anisotropy these exponents are the indices of the *Heisenberg* spin-glass critical point. The scaling function $f(x)$ approaches a constant as $x \to 0$, and $x^{-\gamma}$ as $x \to \infty$. The data of χ_{nl} of the pure sample has been fitted by the above scaling form, using the values $T_g = 11.2 \pm 0.2$ K and $\delta = 5.0 \pm 0.3$ deduced from the results of Fig. 2. We find a good scaling behavior for $\phi \simeq 5.8 \pm 0.5$ as shown in Fig. 3. Our estimate of δ is consistent with the results of Omari, Prejean, and Souletie[8] in CuMn(1%); however, their estimate $\gamma \simeq 3.2$ is significantly smaller than our result $\gamma = \phi(1 - 1/\delta) \simeq 4.6 \pm 0.5$. This discrepancy is probably due to the different procedure that has been used by Omari, Prejean, and Souletie[8] in the analysis of the data. For instance, they use as variables $1 - T_g/T$ and $\mu H/k_B T$ instead of our $t = 1 - T/T_g$ and $\mu H/k_B T_g$, respectively.

Figure 2 already implies that the results for the doped samples do not follow a single-parameter scaling in the range of fields of our experiment. Indeed, there does not exist a set of parameters T_g, ϕ, and δ for which the data of the doped samples collapse onto a single scaling curve. Figure 4 shows a scaling plot of the data of the 3% Au case using the same parameters which were deduced from the pure CuMn data. We

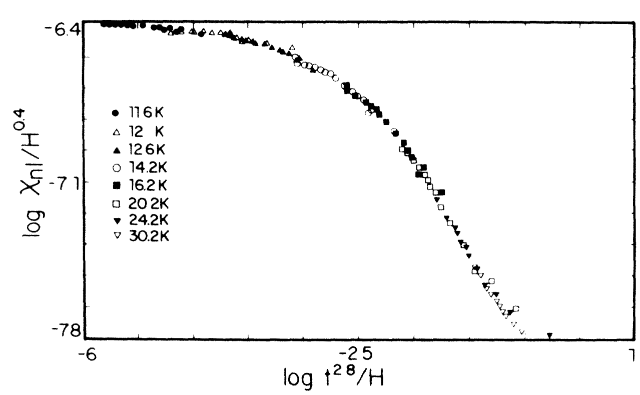

FIG. 3. One-parameter scaling fit [Eq. (2)] for the nonlinear susceptibility data of 1.2 at.% *Cu*Mn.

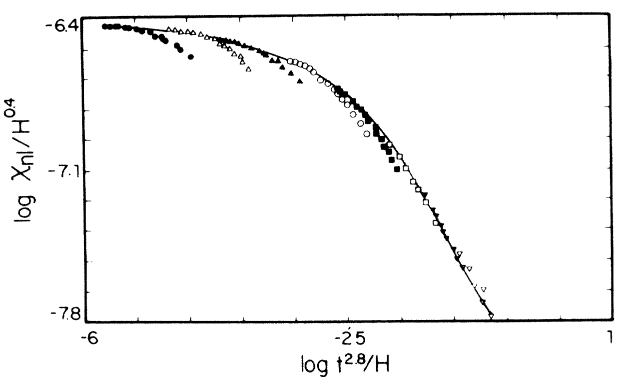

FIG. 4. One-parameter scaling fit [Eq. (2)] for the nonlinear susceptibility of 1.2 at.% *Cu*Mn doped with 3% gold for the same isotherms as in Fig. 3 and for the same scaling parameters. The solid line represents the scaling function obtained from the scaled data of Fig. 3. Note the deviation from this curve in the low-field regime and in the vicinity of T_g.

find that the high-field portions of all the isotherms fall on the same single curve as that of pure CuMn; see Fig. 3. However, systematic deviations from scaling are clearly seen in the low-field data. A similar behavior is observed in the other doped samples.

The failure of the one-parameter scaling fit in the Au-doped samples suggests that the gold impurities are a relevant perturbation which modifies the critical properties of the transition. Indeed, the addition of a random anisotropy induces a randomly distributed preferred orientation for the local spins. As a result, the spin-glass transition is expected to change from a Heisenberg to an Ising critical point. With the assumption that both Heisenberg and Ising SG three-dimensional systems undergo a phase transition at finite temperatures, the effect of a weak random anisotropy on χ_{nl} can be described by the following two-parameter scaling function:

$$\chi_{nl}(T,H,D) = h^{2/\delta} g(t/h^{2/\phi}, d^{2/\phi_A}/h^{2/\phi}). \tag{3}$$

Here D is the strength of the anisotropy energy, and $d = D/k_B T_g$. In the case of the Dzyaloshinsky-Moriya anisotropy in CuMnAu alloys, d^2 is of the form[1,2]

$$T_g d^2 = a c_{Mn}^2 + b c_{Mn} c_{Au}, \tag{4}$$

where c_{Mn} and c_{Au} are the concentrations of the Mn and Au atoms, respectively, and $b/a \approx 17$. The anisotropy crossover exponent ϕ_A as well as δ and ϕ in Eq. (3) are associated with the Heisenberg fixed point. The scaling function $g(x,y)$ reduces to the Heisenberg function $f(x)$, Eq. (2), as $y \to 0$. This is consistent with the observed scaling behavior for all samples in the high-field regime. According to this interpretation the values $\phi = 5.8 \pm 0.5$ and $\delta = 5.0 \pm 0.3$ derived above from our data correspond to Heisenberg critical indices. In the other limit, $y \to \infty$ (i.e., $h \to 0$), Eq. (3) reduces to the one-parameter scaling function of an Ising spin-glass. Deviation from the Heisenberg behavior should be observed in fields $h < h^* \sim d^{\phi/\phi_A}$. Indeed, we do find that the field H^* below which deviations from the one-parameter scaling behavior are observed increases with the Au concentrations. A rough estimate of H^* is 10, 12, and 15 kOe for the 1%, 2%, and 3% Au concentrations, respectively. On the other hand, H^* for the pure CuMn is less than 0.5 kOe. These estimates of H^* together with Eq. (4) suggest that ϕ/ϕ_A is less than 1, but since ϕ is large, ϕ_A *must be quite big*. Thus, the crossover exponent for random anisotropy in a Heisenberg spin-glass is significantly larger than the mean-field value $\phi_A = 1$. The big exponent found here is consistent with the mild shift δT_g of T_g observed in CuMnAu, since according to the above scaling $\delta T_g/T_g \sim d^{2/\phi_A} \sim c_{Au}^{1/\phi_A}$.[12] Nonetheless, we have not found a simple scaling behavior of the data in the low-field, strong-anisotropy regime (say, $H < 5$ kOe for $c_{Au} = 3\%$). This is consistent with Eq. (3) only if the crossover regime is quite wide. Thus, extension of the measurements to lower fields is needed in order to determine the validity of the above Ising-Heisenberg crossover hypothesis.

We now discuss the relation between the results for χ_{nl} at $T \geq T_g$ and the measurements of the field-dependent critical line $T_c(H)$ below $T_g = T_c(H=0)$, in various concentrations of Au. If we set $1 - T_c(H)/T_g \propto h^{2/\theta}$, it has been observed[10,13] that in small fields, the value of θ is close to 3, whereas in high fields, θ is significantly smaller ($\theta \sim 1$). The crossover fields for 1% and 3% Au concentrations were in the range of 5–10 kOe in rough agreement with the values of H^* observed here. Can one relate the exponent θ to the magnetic exponent ϕ of χ_{nl} [Eqs. (2) and (3)]? In mean-field theory, these exponents are unequal: θ equals 3 in an Ising system and 1 in a Heisenberg one, whereas $\phi = 2$ for both. However, a recent scaling analysis of the SG critical point showed that in short-range Ising as well as Heisenberg spin-glasses below six dimensions θ must be equal to ϕ.[6] Indeed, Malozemoff, Barnes, and Barbara[14] find that $\theta \sim \phi \sim 3.5$ in the GdAl spin-glass, in agreement with the scaling theory. However, our result $\phi \sim 5.8 \pm 0.5$ for the weak-anisotropy, high-field regime in CuMn indicates a significant discrepancy between ϕ and the estimate of $\theta \sim 1$ obtained from the torque measurements in the same range of fields and Mn concentrations. Does this apparent discrepancy between θ and ϕ indicate a breakdown of scaling in real SG's or is it an outcome of experimental imperfections? In our view, this is still an open question and more experimental work on both the nonlinear magnetic susceptibility and the critical lines is needed in order to uncover the scaling behavior of spin-glasses above and below their freezing temperature.

In conclusion, it should be noted that our estimates for the critical exponents for the pure CuMn are sensitive to details of the scaling analysis. A better experimental determination of the linear susceptibility χ_0 as well as limiting the range of fields and temperatures might yield more reliable exponents.[15] The main result of the present work is that random anisotropy induces strong crossover effects which modify the static critical properties of Ruderman-Kittel-Kasuya-Yosida spin-glasses. Finally, our results suggest that the observed scaling in pure CuMn corresponds to isotropic critical behavior. This would imply that the lowest critical dimension for the Heisenberg spin-glass transition is less than 3. The observed isotropic behavior in pure CuMn for fields above several hundred oersteds does not exclude the possibility of a crossover to Ising behavior at very low fields due to the weak (dipolar or Dzyaloshinsky-Moryia) anisotropy that is present in this system.

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