

Role of random anisotropy in determining the phase diagram of spin glasses

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We use torque measurements to determine the field dependence of the spin-glass freezing temperature in $CuMn$ alloys doped with gold impurities. We find that by increasing the concentration of gold, and thus enhancing the random anisotropy forces, the transition line changes from Gabay-Toulouse-like to de Almeida-Thouless-like. This change to an Ising behavior is an agreement with the predictions of the mean-field model of Kotliar and Sompolinsky for a Sherrington-Kirkpatrick spin glass with random anisotropy.

A great number of experiments have been done to try to define the transition line(s) in the (H, T) plane predicted by the mean-field theories of spin glasses.¹ Most conventional experiments look for the onset of remanence, dissipation, time dependence, or the bifurcation between the field cooling and zero-field cooling magnetization curves. A de Almeida-Thouless²-like line [i.e., $T_g(H) - T_g(0) \sim H^{2/3}$] is generally found, whereas the mean-field theories for Heisenberg spin glasses predict an onset of irreversibility on the Gabay-Toulouse line of transverse freezing^{3,4} [i.e., $T_g(H) - T_g(0) \sim H^2$]. To explain this discrepancy it is often argued that these conventional experiments are weakly sensitive to transverse freezing and detect only a crossover to strong longitudinal and transverse irreversibility on a de Almeida-Thouless-like line. The onset of irreversibility has also been looked for by torque measurements,^{5,6} which are expected to be more sensitive to transverse freezing. It has actually appeared that the torque onset in $CuMn$ and $AgMn$ alloys is field independent and thus consistent with a transition line of the Gabay-Toulouse type.⁷ In this Rapid Communication we present new torque measurements on $CuMn$ alloys doped with Au impurities which are known to induce strong random anisotropy fields.⁸ We find that the irreversibility onset line progressively changes from Gabay-Toulouse-like to de Almeida-Thouless-like as the concentration of Au is increased. This is in agreement with recent mean-field calculations by Kotliar and Sompolinsky,⁹ which we refer to as KS, predicting that, in the presence of random anisotropy mixing the longitudinal and transverse spin components, an Ising-like phase transition on a de Almeida-Thouless (AT) line replaces the Gabay-Toulouse (GT) transition in the low-field limit.

We used a capacitance bridge technique to measure torque at temperatures down to 1.5 K and fields up to 9.15 kG on cylindrical samples of approximately 1 g. Our standard procedure consists in cooling the sample in a field H at cooling angle $\theta_H = 0$ from well above T_g to a temperature T . We then turn the field to $\theta_H = 5^\circ$ with a turn time of about 1 sec and back to $\theta_H = 0$ after a pause time of about 1 min. A signal proportional to the torque is exhibited as a function of time on a XY recorder. The sample is then warmed above T_g and the procedure is repeated for a different choice of T and H . The sensitivity of our torquemeter (about 10^{-3} dyne-cm in this work) restricts this choice to fields above 0.5 kG. Examples of raw torque recording

have been shown in Ref. 6.

The existence of nonzero torque is a criterion to establish the presence of irreversibility (or nonergodicity). For an isotropic paramagnetic sample \vec{M} and \vec{H} are parallel so there is no torque; if torque is observed in an isotropic system, the sample must necessarily be in a frozen state. The mechanisms giving rise to macroscopic anisotropy and torque have been studied in some detail for $CuMn$ spin glasses.^{8,10-12} However, for the purpose of this Rapid Communication, it is sufficient to assume the existence of a macroscopic anisotropy energy depending on the rotation angle of the remanent magnetization \vec{M}_r from the cooling field direction. In the small-angle limit, the anisotropy is written as

$$E_A = \frac{1}{2} K \theta^2 \quad (1)$$

in the usual notation. When the field \vec{H} is rotated by θ_H from the cooling direction, the equilibrium between the magnetic torque and the anisotropy restoring torque is expressed as

$$M_r H (\theta_H - \theta) = K \theta \quad (2)$$

The torque acting on the sample, $\Gamma = K \theta$, is then given by

$$\Gamma = \left[\frac{1}{K} + \frac{1}{M_r H} \right]^{-1} \theta_H \quad (3)$$

Whether the main contribution to Γ arises from K or M_r has no importance as far as the detection of the onset of irreversibility is concerned. However, as it will appear later, $M_r H$ is much larger than K (in our experiments) as one approaches $T_g(H)$. Therefore, Γ/θ_H is actually a measurement of K . A last property to note is that the anisotropy K is a remanent quantity which, like the remanent magnetization, relaxes slowly with time.¹² In our experiments we record the torque Γ between $t_1 \approx 1$ s and $t_2 \approx 60$ s after the rotation and we observe some relaxation of Γ .⁶ To obtain the results presented in this Rapid Communication, we have chosen to pick the value of Γ at $t = 20$ s. We show typical curves of $\Gamma(t = 20 \text{ s})$ vs T at fixed field in Fig. 1. Γ decreases as T increases, and we define $T_g(H)$ as the temperature where Γ goes to zero.¹³ We have verified that choosing a different fixed t to define Γ hardly affects our results. Within our experimental accuracy, there is a unique value $T_g(H)$ above which $\Gamma = 0$, independently of the

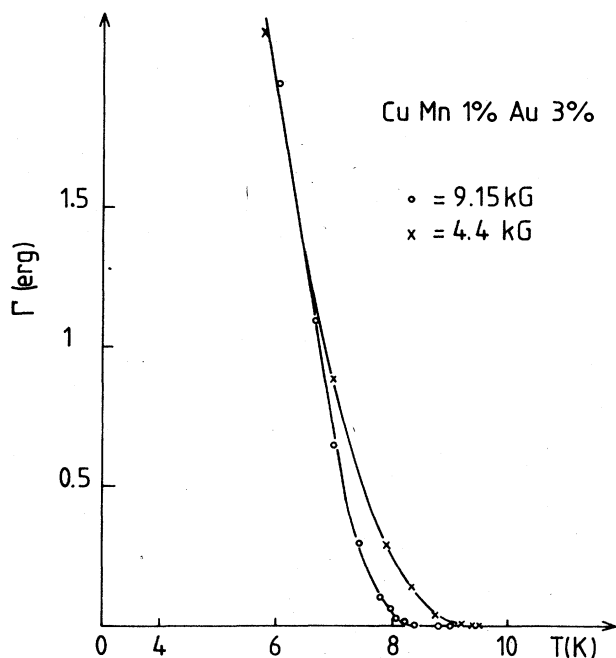


FIG. 1. Torque vs temperature in two different fields for CuMn 1 at.% Au 3 at.%.

choice of t (at least in our time window 1–60 s). We have also verified that the choice of t affects our results on the variation of $K(H, T)$ with T below $T_g(H)$ a little, but not significantly.

We show in Fig. 2, the phase diagrams derived from $T_g(H)$ for CuMn 1 at.% alloys containing 0.3 at.%, 1 at.%, and 3 at.% of Au, respectively. There is a very good agreement with what is expected by the KS mean-field model of spin glasses with Dzyaloshinsky-Moriya (DM) interactions.⁹ With the reduced units $h = \mu H / k_B T_g(0)$, $\tau = 1 - T / T_g(0)$, $d = D / k_B T_g(0)$, where D is the DM coupling constant and μ has to be identified to $g\mu_B \sqrt{S(S+1)}/3$, KS predict three regions of different behavior (see also inset of Fig. 2).

(i) $h \ll d^{3/2}$: Ising-like transition with transverse and longitudinal freezing at $\tau_c(h)$ given by

$$\tau_c(h) = \left(\frac{m+2}{4m} \right)^{1/3} h^{2/3} \quad (4)$$

for m component spins.

(ii) $d^{3/2} \ll h \ll d$: transition (transverse freezing) at a field independent τ_c ($\tau_c = d$).

(iii) $h \gg d$: crossover to a GT behavior with transverse freezing at $\tau_c \sim h^2$.

The predictions of KS are clearly borne out by our experimental results. For CuMn 1 at.% with 0.3 at.% and 1 at.% of Au, τ_c varies as $h^{2/3}$ at low field and then takes an approximately constant value τ^* above a threshold field h^* ($\tau^* \approx 0.15$, $h^* \approx 0.065$ for 0.3 at.% Au, $\tau^* \approx 0.21$, $h^* \approx 0.112$ for 1 at.% Au). For CuMn 1 at.% without Au, $\tau_c(H)$ is constant (about 0.07) throughout our experimental field range, which means that our lowest field is above h^* (in the opposite limit our highest field is too low for the transition line to catch up with the GT line and vary as h^2). Similar results have been found in AgMn.⁶ For CuMn 1 at.% with 3 at.% Au, τ_c varies approximately as $h^{2/3}$

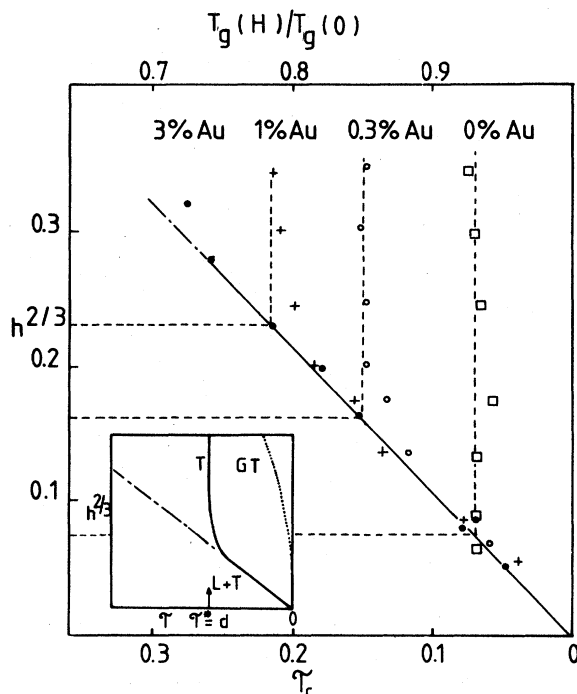


FIG. 2. $h^{2/3}$ vs τ_c for CuMn 1 at.% alloys containing the indicated concentrations of Au, with $h = g\mu_B[S(S+1)/3]^{1/2}H/k_B T_g(0)$, $g=2$, $S=5/2$, $\tau_c = [T_g(0) - T_g(H)]/k_B T_g(0)$. For 0 at.% Au $T_g(0)$ is derived from the cusp of the low-field susceptibility: $T_g(0) = 10.2$ K. For the other alloys $T_g(0)$ is derived by extrapolating the variation of $T_g(H)$ as $H^{2/3}$ to zero field: $T_g(0) = 10.2$ K for 0.3 at.% Au, $T_g(0) = 10.3$ for 1 at.% Au, $T_g(0) = 11.4$ for 3 at.% Au (weakly different values are derived from the susceptibility cusp). The oblique line corresponds to $\tau_c = 0.93h^{2/3}$. The vertical and horizontal dashed lines indicate τ^* and h^* , respectively. The uncertainty on $T_g(H)$ is about 0.1 K. The maximum field is 9 kG. Inset: schematic phase diagram of the KS model. Solid line: transition line. Dotted line: GT line (i.e., transition line in the absence of anisotropy).

throughout our field range, which indicates that h^* is above our highest field (the beginning of departure from $h^{2/3}$ at high field suggests that h^* is not very far above). For the three alloys exhibiting approximately a linear variation of τ_c with $h^{2/3}$, the slope of this variation is approximately the same, $\tau_c \approx 0.93h^{2/3}$, while $\tau_c \approx 0.75h^{2/3}$ is expected from Eq. (4) with $m=3$. However, because the field range is relatively small, our results cannot prove that the exponent is exactly $2/3$. As good fits could be obtained with, for example, $h^{0.5}$ or $h^{0.8}$.

We have estimated the ratio anisotropy/exchange of our alloys by assuming the following standard expressions for the Ruderman-Kittel-Kasuya-Yosida (RKKY) constant and DM vector:^{8,10}

$$J_{ij} = V_0 \frac{\cos(2k_F R_{ij})}{R_{ij}^3}, \quad (5)$$

$$\vec{D}_{ij} = V_1 \frac{\sin[k_F(R_i + R_j + R_{ij}) + \phi] \hat{R}_i \cdot \hat{R}_j}{R_i R_j R_{ij}} (\hat{R}_j \times \hat{R}_i). \quad (6)$$

Equation (6) holds for magnetic impurities at \vec{R}_i and \vec{R}_j , and a spin-orbit scatterer at $\vec{R}=0$. In a mean-field ap-

proach, the coefficients J and D of the Hamiltonian of KS have to be identified to the following averages:

$$J = k_B T_g(0) = \sqrt{N} (\langle J_{ij}^2 \rangle)^{1/2}, \quad (7)$$

$$D = \sqrt{N} (\langle |\bar{D}_{ij}|^2 \rangle / 3)^{1/2}. \quad (8)$$

According to Bray and Moore¹⁴ and Levy Morgan-Pond, and Raghavan¹⁵ these lead to¹⁶

$$J = k T_g(0) \approx \frac{V_0}{a^3} c_{Mn}, \quad (9)$$

$$D \approx \frac{1}{a^3} \left[\ln \left(\frac{R_c}{r_0} \right) + \frac{1}{2} \right]^{1/2} [(V_1^{Mn} c_{Mn})^2 + (V_1^{Au})^2 c_{Mn} c_{Au}]^{1/2} \quad (10)$$

up to a dimensionless constant of order unity. V_1^{Mn} and V_1^{Au} are the DM coefficients for Mn and Au spin-orbit scatterers, respectively; c_{Mn} and c_{Au} are the concentrations of Mn and Au, $R_c \approx 42 \text{ \AA}$, $r_0 = 2.55 \text{ \AA}$. We have calculated $d = D/k_B T_g(0)$ from Eqs. (9) and (10) by using the values $V_1^{Mn}/V_0 \approx 2 \times 10^{-2}$ and $V_1^{Au}/V_0 \approx 8.4 \times 10^{-2}$ estimated from the anisotropy constant K of CuMnAu and CuMn alloys at $T \ll T_g$.¹⁰ We obtain $\tau^* = d = 0.036$, $\tau^* = d = 0.093$, and $\tau^* = d = 0.16$ for CuMn 1 at. % with 0%, 0.3 at. % and 1 at. % of Au, respectively. The agreement with the experimental values, $\tau^* = 0.07$, $\tau^* = 0.15$, and $\tau^* = 0.21$, respectively, is quite satisfying.

Besides the confirmation of the phase diagram of spin glasses with random anisotropy, our torque data also allow us to probe some predictions of KS on the critical behavior of K . In the low-field limit (i.e., $h < d^{3/2}$) KS predict

$$K \sim \tau^3 - \tau_c (h)^3 = \tau^3 - \frac{5}{12} h^2. \quad (11)$$

On the other hand, K is related to the torque Γ by Eq. (3) which, in the limit $MrH \gg K$ predicted by KS for $\tau \ll 1$,¹⁷ is written

$$K = \Gamma / \theta_H. \quad (12)$$

We have used Eq. (12) to derive $\Gamma(T, H)$ from our experimental results on Γ , and we have plotted K vs τ^3 for several values of H in Fig. 3. There is a remarkable agreement with Eq. (11). At fixed field, K varies linearly with τ^3 and increasing H shifts down this linear variation by an amount varying approximately as H^2 . In contrast, in the high-field regime (i.e., $h > d^{3/2}$), our results do not seem in agreement with the predictions of KS, i.e., $K \sim h(\tau - \tau_c)^2$. A more detailed discussion of this critical behavior will be presented in a further publication.

Our torque measurements on CuMnAu alloys support the theories predicting a phase transition and demonstrate the essential role of random anisotropy in determining the phase diagram of a Heisenberg spin glass in the (H, T)

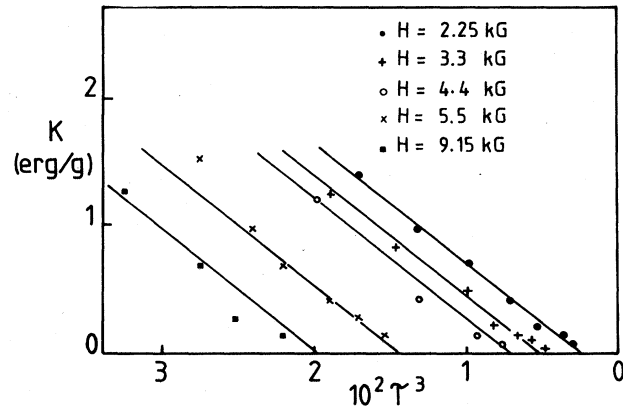


FIG. 3. Anisotropy coefficient K vs τ^3 in different fields for CuMn 1 at. % Au 3 at. %.

plane. They confirm the predictions of KS:⁹ the behavior is Ising-like and the irreversibility onset line is AT-like in the low-field strong anisotropy limit (i.e. $h < d^{3/2}$); as the ratio field/anisotropy is increased ($h > d^{3/2}$), there is a crossover to pure Heisenberg behavior with transverse only freezing on a GT-like line. CuMn alloys with small concentrations of Au have a small enough anisotropy to go from the first to the second behavior in our experimental field range. For 3 at. % of Au and above, only the strong anisotropy behavior can be observed in our fields. It is clear that most of the spin glasses have stronger anisotropy forces than CuMn and will generally exhibit the strong anisotropy behavior (AT-like line) in the usual experimental field ranges, as we have recently confirmed by torque measurements on AuFe alloys. It is likely that the pure Heisenberg behavior predicted by Gabay and Toulouse³ will be observed only in the few spin glasses having a very small anisotropy, such as CuMn, AgMn, NiMn, Moreover, as we have shown for CuMn, the GT transition line will be observed only in high enough fields and probably by only measurements sensitive to transverse freezing, such as torque or transverse susceptibility. Such measurements should be able to determine precisely the form of the transverse freezing line—in order to distinguish between the predictions of the mean-field models and those of more general pictures¹⁸—and to make clear the critical behavior of the anisotropy K in both the strong and weak anisotropy limits.

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⁷The results of the first torque measurements of this type appeared to be consistent with AT-like behavior (Ref. 5). It was later realized that the prefactor used in the expression of the AT line [taken from Ref. 19, R. V. Chamberlin *et al.*, Phys. Rev. B **25**, 6720 (1983)] was incorrect. By considering more accurate extended torque data on CuMn and AgMn alloys and using the correct prefactor, the same authors stated in Ref. 6 that the field dependence of T_g is much weaker than expected for an AT line [T_g is almost field independent with $T_g(H) \approx 0.9T_g(0)$ throughout the field range over which experiments could be performed].

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that of Fig. 3 and extrapolating to $K=0$. The results obtained by these different methods are slightly but not significantly different.

Figure 2 presents data obtained by the first method.

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¹⁶See Eqs. (18) and (19) of Ref. 14 and Eq. (5.10) of Ref. 15.

¹⁷A careful analysis of the experimental temperature dependence of Γ also shows that $K/M_r H \rightarrow 0$ as T approaches T_g . Well below T_g , although K is field independent (Ref. 10), Γ depends on H , which, according to Eq. (3) means that $K/M_r H$ is not very small. At higher T (e.g., 6 K in Fig. 1) Γ becomes nearly field independent. This means that K has decreased more rapidly than $M_r H$, so that $K/M_r H \ll 1$ and $\Gamma \approx K\theta_H$; as K is still field independent, Γ is field independent. Finally, as T approaches T_g , Γ again becomes H independent, this time because K has become field dependent; see Eq. (11).

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