Dynamic Measurements in a Heisenberg Spin Glass: CuMn

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We have made extensive high field thermoremanent magnetization decay measurements on Cu:Mn(6%), in magnetic fields between 5 and 6 kG, over a wide range of temperatures and for three different waiting times to examine spin glass dynamics in the previously unexplored Heisenberg regime. We have found aging effects for both transverse and longitudinal freezing, with crossover behavior between the two regimes. This provides the first dynamical experimental evidence consistent with the Heisenberg spin glass *H-T* phase diagram derived by Gabay and Toulouse.

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Characterization of the universality class of the so-called canonical spin glasses (SG) Cu:Mn and Ag:Mn has proved elusive. Although this type of spin system is nominally Heisenberg, the presence of weak anisotropies induces an Ising-like state at sufficiently small magnetic fields [1]. At large fields, the Zeeman energy dominates over the anisotropic energy. The SG should regain its Heisenberg character, with, of course, anisotropy. This paper addresses the question of whether this high field state in Cu:Mn exhibits a phase transition at all, and whether the dynamical response is consistent with the prediction of Gabay and Toulouse (GT) within this state: Does a crossover from transverse to longitudinal freezing occur below the transition temperature for Heisenberg spin glasses? Our data exhibit behavior which is consistent with a crossover from a transverse freezing regime to a longitudinal freezing regime [2]. The two regimes are separated by crossover rather than a sharp phase transition, as stipulated by Gabay and Toulouse.

The existence of a phase transition in three dimensions for a purely Heisenberg spin glass is controversial. Numerical simulations of a Heisenberg spin system with random interactions [3] suggest that the lower critical dimension (LCD) of a Hiesenberg spin glass in the absence of anisotropy lies between three and four. Reger and Young [4] have considered RKKY interactions between Heisenberg coupled spins. They suggest that the LCD is around three, though they are unable to rule out the possibility of the LCD being less than three. Recently Matsubara and Iguchi [5] have concluded from simulations that a site disordered Heisenberg SG with RKKY coupling and zero anisotropy has a phase transition in three dimensions. To date, no numerical simulations have been made for a Hiesenberg spin glass with RKKY interactions and weak unidirectional anisotropy as a function of the magnetic field.

There is a growing body of experimental evidence that suggests that, in agreement with mean field predictions, a Heisenberg spin glass with weak unidirectional anisotropy behaves as an Ising spin glass in low magnetic fields. It then is supposed to cross over to a Heisenberg SG in larger magnetic fields. de Courtenay, Fert, and Campbell [6] have shown that the onset of irreversibility in the transverse component of the magnetization follows a d'Almeida-Thouless (AT)-like relation in low magnetic fields, crossing over to a Gabay-Toulouse (GT)-like relationship for magnetic fields greater than the anisotropy field. By measurement of the nonlinear magnetization, de Courtenay *et al.* [7] showed that the magnetic field critical exponent δ also exhibits crossover behavior from a low field "Ising" value to a high field "Heisenberg" value. The above authors have also shown, in agreement with mean field theory [1], that the Ising to Heisenberg crossover fields and temperatures are dependent on the strength of the anisotropy.

A previous study [8] of the zero field cooled (ZFC) and field cooled (FC) longitudinal magnetization over a large *H-T* range generated the phase diagram for CuMn (6%) in magnetic fields between 3 G and 30 kG in Fig. 1. In low magnetic fields, the data are consistent with the onset of an AT line, but with a field coefficient approximately 35 times larger than that which is predicted from mean field theories, in agreement with previously reported experimental observations of the AT line in Cu:Mn. However, above approximately 500 G the data are consistent with the mean field values of the coefficient predicted for the GT transition (transverse moment freezing) and at a lower temperature at the AT (now crossover) line (longitudinal moment freezing). The existence of a finite extrapolated zero-field transition temperature of the high field Heisenberg transition lines suggests a LCD less than three for a Heisenberg spin glass in the presence of anisotropy.

We measure the long time decay of the high field thermoremanent magnetization (HFTRM) in this study to explore the dynamical behavior in the Heisenberg regime, inclusive of both the longitudinal and transverse freezing regimes. All measurements are made in static fields be-

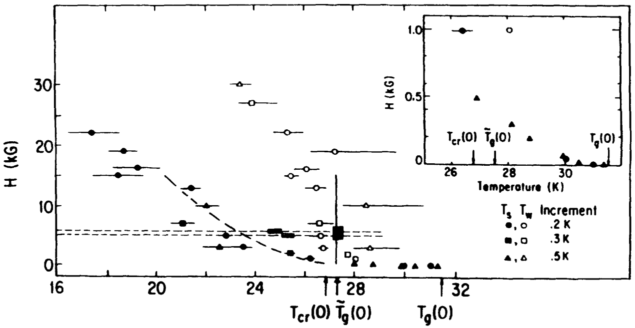

FIG. 1. A plot of H vs T phase diagram for Cu:Mn 6% in fields between 2 G and 30 kG. The solid line is the best fit of the data to the mean field Gabay-Toulouse line while the dashed line is the theoretical high field d'Almeida-Thouless crossover line to longitudinal freezing. The low field "Ising" regime is blown up in the inset. The thick solid area indicates the temperature region where magnetization decays and aging effects are no longer discernible. The shaded areas reflect the temperature regions where crossover behavior occurs for the stretched exponential n for $H = 5.8$ and 5.1 kG.

tween 5 and 6 kG to ensure that the sample is well within the Heisenberg spin glass regime (Fig. 1). Similarities between the HFTRM and the low field TRM decay measurements which have been used extensively to study the low field Ising behavior of these systems allow for a comparison of both the form of the TRM decays and aging effects in the Heisenberg and Ising regimes.

The sample used in this study was a $Cu_{0.94}Mn_{0.06}$ alloy of approximate dimensions 2 mm×2 mm×3 mm. It was prepared by melting the proper stoichiometric ratios of $5^{9's}$ Cu and $4^{9's}$ Mn in an arc furnace. The sample was then cut from the master alloy to the proper size with a string saw, and etched in nitric acid. The sample was vacuum annealed for 48 h at 600 °C and then quenched to 77 K.

The magnetization decay measurements were made with a homemade high field SQUID magnetometer meriting a detailed description: The sample sits in the top loop of epoxy-embedded pickup coils in a second order gradiometer configuration. Concentric to the pickup coils is a small copper solenoid with which fields of up to 120 G can be turned on or off. Concentric to the small solenoid is a large superconducting solenoid. All of the components inside the magnetic field are bolted to each other and to the superconducting solenoid to reduce field dependent vibrational noise.

The large magnetic field was produced by a Cryomagnetics model 190 superconducting solenoid. It required approximately two weeks after the initialization of the persistent current for measured drifts in the system to become small enough to allow measurements. The magnet is in a 90 liter He Dewar which was topped off every 60 h. The process of filling the He Dewar induces a strong drift which, after about 10 h, decays to a level (drift measuring time of 1 h) on the order of the noise attributable to sample temperature fluctuation.

The requirement of extreme magnetic field stability required continuous operation for periods of 3 to 6 months. The data presented in this study arise from three different periods of continuous operation. Between these periods of operation attempts were made to improve temperature resolution, reduce the signal-to-noise ratio, and obtain a more accurate value of the magnetic field with the introduction of an *in situ* Hall probe at the beginning of the third period.

The HFTRM decay measurements were performed using the following protocol. The sample was first heated to 35 K (well above the SG transition temperature) in a field of 5 to 6 kG induced in the superconducting solenoid (5.8 kG in the first period of operation and 5.1 kG in subsequent periods). During the first period of operation a magnetic field of 70 G was applied using the small solenoid (120 G for subsequent periods of operation). The sample was then rapidly cooled to the measuring temperature T_m. After waiting a time t_w at T_m, the small 70 G field was cut to zero. The waiting times employed were 480, 1000, and 3600 s. Immediately after the small magnetic field was cut to zero, a rapid initial drop in the magnetization occurs which we are unable to measure. After, we measure the longitudinal magnetization decay from 5 to 3600 s using a BTI SQUID

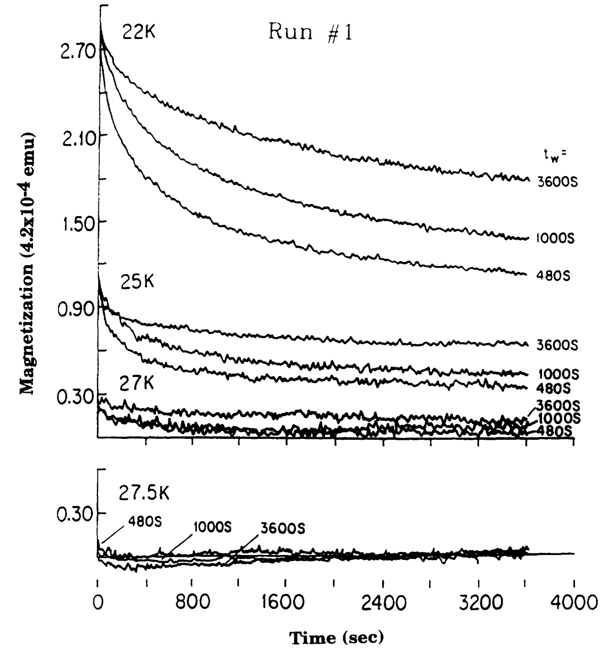

FIG. 2. HFTRM decays for three different waiting times, 480, 1000, and 3500 s, and four different temperatures during the first set of runs. The 22 K data are in the region of longitudinal freezing while the 25 and 27 K decays are in the region of transverse freezing. At 27.5 K we lose the ability to resolve either magnetization decays or waiting time effects.

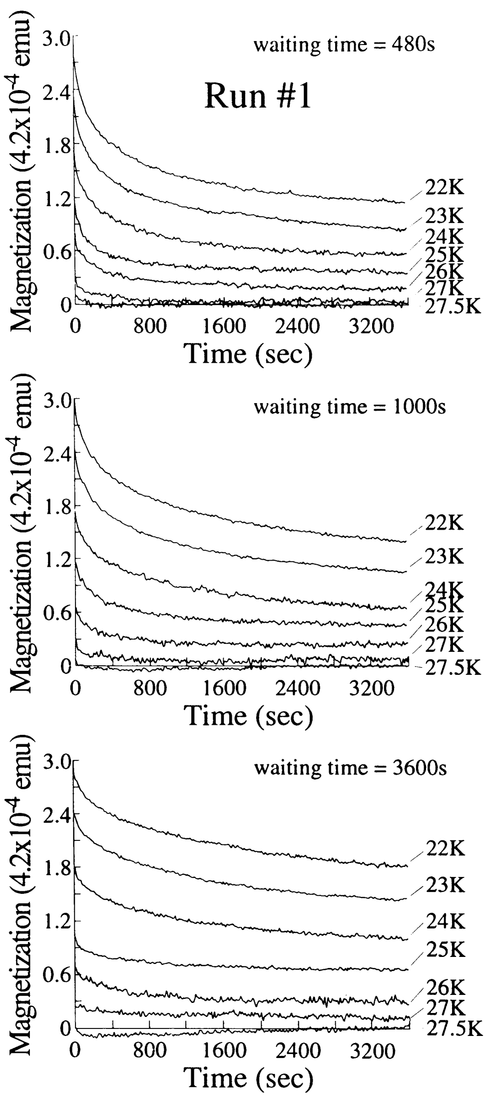

FIG. 3. HFTRM decays for three different waiting times, 480, 1000, and 3600 s, and seven different temperatures between 22 and 27.5 K during the first period of operation. Notice the change in shape of decays from 24 to 25 K.

amplifier coupled to the pickup coils. The same protocol was repeated with a -70 G field on the small solenoid. The two decays ($+70$ G, -70 G) are then subtracted to eliminate the systematic decay. The sample signal was approximately 70% (at 24 K) of the total decay measured during the first two periods of operation. After the introduction of a Hall probe used during the third period of operation, the sample signal to total decay ratio dropped to approximately 30%.

Figure 2 exhibits the HFTRM decays for the three different waiting times at four different temperatures (measured during the first period of operation), inclusive of both the longitudinal and transverse freezing regimes. Not only do long time magnetization decays persist within both longitudinal and transverse freezing regimes,

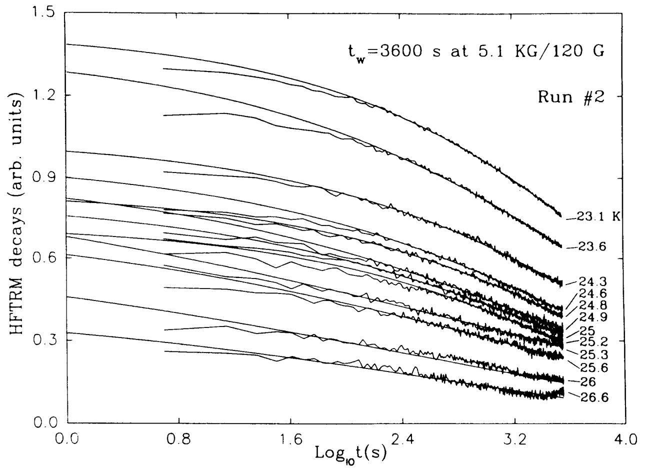

FIG. 4. HFTRM decays vs $\log_{10}(t)$ for the second set of runs with a waiting time of 3600 s. It shows more clearly the change in the form of decays which occurred between 25.2 and 25.3 K. The fits to the stretched exponential equation are superimposed on the data.

but also waiting time effects are present for all temperatures below 27 K. At 27.5 K no decay in the HFTRM or waiting time effects are observed. The temperature range between 27 and 27.5 K at 5.8 kG is plotted in Fig. 1 as a thick line. It is coincident with the previous "static" determination of the Gabay-Toulouse line [8].

Plots of HFTRM decays for several representative temperatures between 20 and 27.5 K and three different waiting times are shown in Fig. 3. The most interesting aspect of these decays is that there is clearly a change in the magnetization decay *shape* between 24 and 25 K at all waiting times. This change is clearly evident in Fig. 4, a plot of HFTRM decays vs $\log_{10}(t)$ from the second period of operation (5.1 kG on the superconducting magnet, 120 G on the small solenoid). Figure 4 also includes fits of the data to the stretched exponential form

$$M(t)=M_0\exp\left[-\left(\frac{t}{\tau_p}\right)^{1-n}\right]. \tag{1}$$

All of the data for $130\ \mathrm{s} \le t \le t_w$ can be reasonably fitted with this shape. For all temperatures there is deviation from the stretched exponential decay during the time regime 5 to 40 s. This may be due to power law behavior previously observed for short times in spin glasses in Ref. [9], but the smallness of the time window precludes a precise analysis.

The stretched exponential exponent n and characteristic time τ_p defined by Eq. (1) are plotted as a function of temperature in Fig. 5 for the three different periods of operation. A clear change in the value of the exponent n is observed in the temperature regime where there is a change in the shape of decays. Within this same temperature regime, τ_p appears to begin to decrease more rapid-

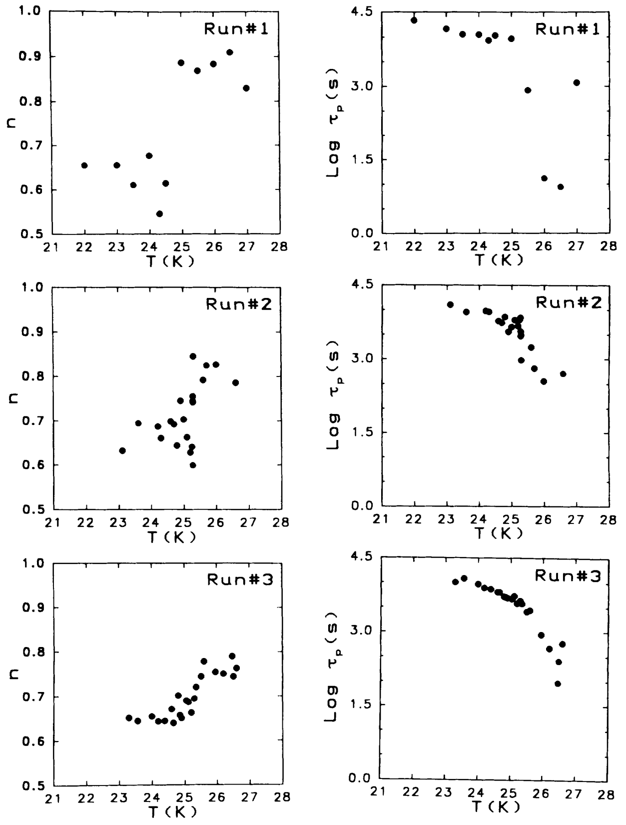

FIG. 5. Parameters n and τ_p vs temperature of the stretched exponential fits for $t_w = 3600$ s during the three periods of runs.

ly as a function of temperature. The larger scatter in the values of the parameters above this temperature regime for the change in n is caused by the decrease of HFTRM signal-to-noise ratio when we approach the Gabay-Toulouse line. This regime where n changes is at a slightly higher temperature than where we have previously observed the change from weak to strong irreversibility in the longitudinal magnetization [10].

In the low field Ising regime, when the spin glass phase transition temperature is approached from below, the stretched exponent n increases towards unity [11]. The absence of such systematic n vs T behavior in our HFTRM decays when n changes is consistent with a crossover transition from longitudinal to transverse freezing as predicted by Gabay and Toulouse [2].

In summary, we have made a series of measurements of the longitudinal component of the HFTRM decays in magnetic fields between 5 and 6 kG. We find very long time magnetization decays and waiting time effects at all temperatures below the temperature we have previously determined to be the Gabay-Toulouse transition temperature. We find that the form of the magnetization decay changes at a temperature only slightly larger than our previous determination of the high field longitudinal crossover temperature. These measurements are consistent with the mean field theories which predict the existence of two different phases in the Heisenberg spin glass regime, transverse and longitudinal freezing, separated by a crossover line.

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