PHYSICAL REVIEW B VOLUME 33, NUMBER 5 1 MARCH 1986

Critical behavior of the transverse susceptibility in a *Cu*Mn spin glass

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(Received 19 November 1985)

Transverse ac susceptibility measurements on $CuMn_{4\,\mathrm{at.\%}}$ are used to examine the quantity δ, which is closely related to the transverse order parameter Δ^y introduced by Kotliar and Sompolinsky in a recent Letter. The δ data scale in the critical-phenomena sense, giving strong support for the existence of a phase transition in this weakly anisotropic spin glass. We show that this transition occurs along the Gabay-Toulouse line, as predicted by mean-field theory.

According to mean-field theory, Ising spin glasses undergo a phase transition along the de Almeida–Thouless (AT) line[1]

$$\tau_{\mathrm{AT}} = (H/H_1)^{2/3} \ , \tag{1}$$

where $\tau = 1 - T/T_g$ and $H_1 = 2k_B T_g/\sqrt{3}\mu_B$. Mean-field theory for a Heisenberg spin glass shows that the transition occurs in two stages.[2–4] The transverse degrees of freedom (i.e., spin components perpendicular to the applied field) first freeze along the Gabay-Toulouse (GT) line, given by

$$\tau_{\mathrm{GT}} = (H/H_3)^2 \ , \tag{2}$$

where $H_3 = 10k_B T_g/\sqrt{23}\mu_B$, followed at lower temperature by the freezing of the longitudinal spin components along the AT line. A dynamical study by Fischer[5] on the infinite-ranged vector spin-glass model confirms that a spin-glass phase transition occurs along the GT line but finds the AT line to represent a dynamical crossover rather than a phase transition. In real materials such as *Cu*Mn, prominent irreversibilities arise along an AT-like line[6–12] whose position depends on the measuring time,[10,11] suggesting that it is indeed dynamical in origin. The GT line, on the other hand, has proven to be very elusive.[13–17] An explanation was given by Sompolinsky, Kotliar, and Zippelius,[18] who showed that for an isotropic spin glass, the irreversibility associated with the GT line manifests itself only in the local susceptibility. The experimentally accessible uniform susceptibility remains reversible. Recently, Kotliar and Sompolinsky[19] (KS) extended their work to include random anisotropy. They show that the presence of anisotropy couples the local and uniform response to a magnetic field, allowing one to observe the local irreversibility through the uniform response.

In the present Rapid Communication we examine δ, which is closely related to the irreversible part of the local transverse susceptibility Δ^y, in $CuMn_{4\,\mathrm{at.\%}}$ as a function of the applied longitudinal field and temperature. We demonstrate that δ scales in the critical-phenomena sense for fields above 250 Oe. The scaled δ is a function of $x = \tau/H^2$ and vanishes at a nonzero value of x. The scaling form confirms the existence of a critical line given by Eq. (2), albeit with a characteristic field only 20% of the mean-field value. Our results differ from recent torque measurements in which a nearly field-independent irreversibility line was found.[20,21] Reasons for this discrepancy are discussed.

As noted above, Kotliar and Sompolinsky[19] extended the dynamical theory of the infinite-ranged Heisenberg spin-glass model to include a uniform magnetic field and the random anisotropic Dzyaloshinsky-Moriya (DM) interaction. The DM interaction[22] plays a crucial role, coupling the macroscopic response in a uniform magnetic field to the microscopic degrees of freedom. The macroscopic transverse susceptibility derived by KS for small fields is given by

$$\chi_\perp = M/H - \Delta^y(1 + HM_r/K)^{-1} \ , \tag{3}$$

where Δ^y is the irreversible part of the local transverse susceptibility $(\chi_{\perp(\mathrm{dc})}^{\mathrm{local}} - \chi_{\perp(\mathrm{ac})}^{\mathrm{local}})$, M_r is the longitudinal remanent magnetization, and K is a macroscopic anisotropy constant. In the limit of vanishing K, the longitudinal susceptibility is given by the ratio of the longitudinal equilibrium magnetization M and the applied field—the susceptibility remains isotropic even in the presence of a nonzero Δ^y. According to KS the transverse order parameter Δ^y exhibits at least three scaling regimes, depending on the magnitude of H relative to the strength of the DM interaction. In the low-field limit local and macroscopic modes are tightly coupled and the system behaves in an Ising-like manner; Δ^y becomes nonzero along the AT line in this strong anisotropy region. At the opposite extreme of large fields relative to the DM interaction, called the weak-anisotropy region, the phase transition occurs along the GT line, with a dynamical crossover along the AT line. At intermediate values of anisotropy and field the phase transition line is relatively insensitive to the field. The results of the KS theory are expected to apply to spin glasses such as *Cu*Mn, where the DM interaction is known to play an important role.[22,23]

We cannot measure Δ^y directly in an experiment. However, by rearranging Eq. (3) we may define the measurable quantity δ,

$$\delta \equiv M/H - \chi_\perp(T) = \Delta^y(1 + HM_r/K)^{-1} \ , \tag{4}$$

which is closely related to Δ^y. The equilibrium susceptibility M/H may be measured directly in a field-cooled magnetization experiment, or approximated with the projection hypothesis.[24] The latter states that the magnetization below the spin-glass transition is independent of temperature. Thus Eq. (5) becomes

$$\delta \simeq \chi_\perp(T_g) - \chi_\perp(T) \ , \tag{5}$$

where T_g is the spin-glass transition temperature. While the projection hypothesis is not strictly obeyed in real systems, we will demonstrate that it is an accurate approximation for *Cu*Mn in the proper field and temperature regime. We also show that δ, calculated with or without the aid of the projection hypothesis, has a very simple scaling form over a wide range of H and T and, consequently, that it mirrors the critical behavior of the spin-glass transition. The quantity δ was first shown to be of importance in an earlier work on the amorphous spin-glass $Fe_{10}Ni_{70}P_{20}$.[25]

A sample of $Cu\text{Mn}_{4\,\text{at.\%}}$ was prepared for this study from high purity Cu and Mn. The alloy was fabricated in the standard fashion: The melt was gradually cooled to 900 K, annealed there for 24 h, and then slowly cooled to room temperature. The sample was machined into a cylinder and etched in 50% nitric acid to remove surface impurities and surface strains. The measurements were performed with a unique ac susceptometer whose measuring coils are thermally isolated from the sample, preventing errors due to temperature variations in the coil resistance. The inductive mismatch between the coils was balanced using a trimming coil, driven with active electronics, surrounding the empty secondary. The measurements were performed at 76 Hz, with an oscillating transverse probe field of 0.5 Oe. The longitudinal dc field was provided by an electromagnet. In zero field a sharp peak in the ac susceptibility occurred at the freezing temperature $T_g = 19.7$ K.

The field-cooled transverse susceptibility of this $Cu\text{Mn}_{4\,\text{at.\%}}$ sample was measured from 60 to 8 K for 23 fields between 40 and 3400 Oe. Representative susceptibility data are displayed in Fig. 1. Field-cooled dc susceptibility data, taken on a commercial superconducting-quantum-interference-device- (SQUID-) based magnetometer,[26] are also presented in Fig. 1. Data were taken at cooling rates of 0.075 K/s and 2.5×10^{-4} K/s to investigate nonequilibrium effects;[27] none were detected. Observe that the dc susceptibility is nearly temperature independent below T_g for large fields, suggesting that Eq. (5) represents a good approximation. Consequences of this approximation will be discussed in greater detail shortly. Using Eq. (5), the pertinent quantity δ is calculated and presented in Fig. 2. We can see that the various isotherms of δ behave similarly, decreasing gradually for small values of H, then rapidly at intermediate values, and falling off gradually again at higher fields. Extrapolation to zero, as suggested by the dashed lines in Fig. 2, gives an estimate for the critical field. Because the functional form of δ is not known the extrapolation cannot be done accurately. However, a scaling analysis, suggested by the similarity of the δ vs H curves, serves as a powerful and accurate tool in examining the critical region. It will allow us to use data over a wide range of H and T to determine the functional form of the transition line.

Since δ is related to a spin-glass order parameter, it should obey thermodynamic scaling relations if a true phase transition is present. However, the analysis is complicated by the presence of the term $\phi \equiv HM_r/K$ in the denominator of Eq. (4). If ϕ is small compared to unity we have $\delta \simeq \Delta^y$. If, on the other hand, ϕ is large then $\delta \simeq K\Delta^y/HM_r$. Since both K and Δ^y vanish at the critical line δ may still be treated as an order parameter. At intermediate values of ϕ the critical behavior of Δ^y will still be reflected in δ. We do not, at this point, know enough about ϕ to predict which of the above situations is applicable in the present experiment. However, regardless of the magnitude of ϕ, δ may be used to obtain the critical line.

According to the KS theory, δ should vanish along a criti-

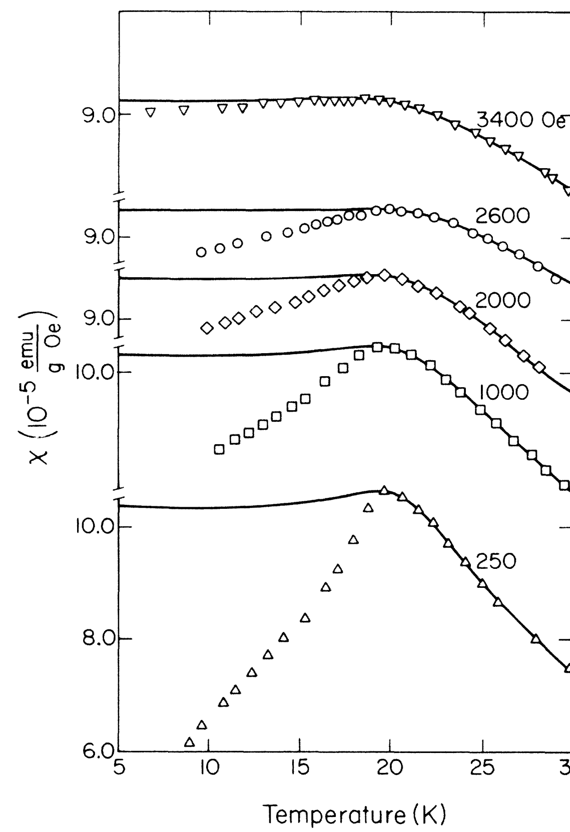

FIG. 1. The transverse ac susceptibility (open symbols) and the dc susceptibility (solid line) are plotted as a function of temperature for several fields. The scale for each curve is the same, but the zeros have been offset for clarity. The ac measurements were performed at 76 Hz with an oscillating field of 0.5 Oe.

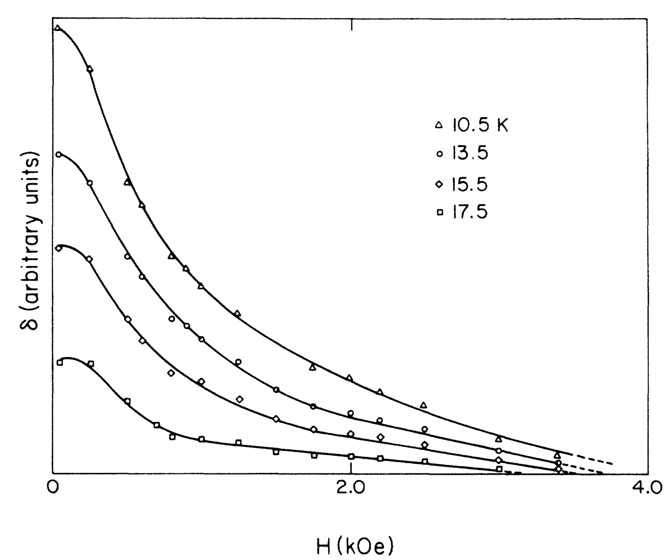

FIG. 2. Some representative isotherms of $\delta \equiv \chi_\perp(T_g) - \chi_\perp(T < T_g)$ are shown as a function of the applied longitudinal field. The isotherms display similar behavior, suggestive of a scaling relation. The fields are in kOe. The lines drawn are guides for the eye.

cal line of the form

$$\tau/H^{\eta}=\text{const} \quad , \tag{6}$$

where the value of η depends on the anisotropy region being considered. Equation (6) suggests a scaling relation of the form

$$\delta=H^{\sigma}F(\tau/H^{\eta}) \quad , \tag{7}$$

or, equivalently,

$$\delta=\tau^{\sigma/\eta}G(H/\tau^{1/\eta}) \quad . \tag{8}$$

If the functions $F(x)$ or $G(y)$ vanish at a nonzero value of their argument then the critical line given by Eq. (6) exists; otherwise, δ vanishes only in the limit $H\rightarrow\infty$.

We have plotted the quantity δ/H^{σ} as a function of τ/H^{η} using both Eqs. (4) and (5) to calculate δ. In both instances the data collapse to a single curve for $\sigma=1.0\pm0.1$ and $\eta=2.0\pm0.15$. The scaled data are displayed in the inset of Fig. 3 for 250 Oe $< H \leq 2600$ Oe and $0.09<\tau<0.5$. Data for $\tau<0.009$ are not used because experimental uncertainties lead to large errors in this region of small δ. We see that the scaling of the data calculated with the use of the projection hypothesis is as good as that incorporating the true dc susceptibility. However, both approaches do not yield the same curve. Using the true dc susceptibility gives a lower value of δ than that obtained using the projection hypothesis. This difference is greatest at large values of τ and small values of H, but vanishes for small τ and large H, as Fig. 3 suggests. Therefore, in the latter region, corresponding to the critical region, the projection hypothesis may be used as a valid approximation. While it is not necesssary to use this approximation, it does have some practical advantages. In particular, the experimental errors introduced by subtracting the true dc susceptibility from the transverse susceptibility are eliminated, allowing for a more accurate examination of the critical region.

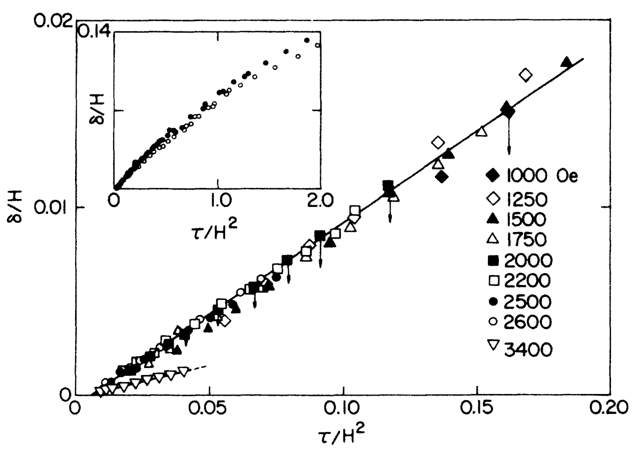

FIG. 3. The inset shows the scaling of δ in the range 250 Oe $< H \leq 2600$ Oe and $0.09<\tau<0.5$. Equation (4) was used to calculate the open circles, while the solid circles were calculated with Eq. (5). Note the convergence of the two curves near the origin. Approximately 140 data points are involved in this scaling. The larger figure is an enlarged view of the critical region. Equation (5) was used to calculate δ. Arrows indicate the position of a few points if Eq. (4) were used. Again, note the convergence near the origin. The solid line is a linear least-squares fit showing the existence of a critical point. A change in scaling is seen at 3400 Oe, but the critical point remains the same. The fields are in kOe.

Using the projection hypothesis we continue our study of the critical region. Figure 3 suggests that the scaling function $F(x)$ is linear for small x and vanishes at a nonzero value of x. To test the last assertion more fully, we have made linear least-squares fits to the scaled data for $x<0.4$ $(\text{kOe})^{-2}$. For one fit, the intercept was free to assume a nonzero value; for the other it was forced to vanish. Forcing $F(x)$ to pass through the origin dramatically increases the standard error to fit. We, therefore, conclude that

$$\delta\sim H[\tau/H^2-(13.0\pm1.7\ \text{kOe})^{-2}] \quad , \tag{9}$$

verifying Eq. (2) for the Gabay-Toulouse line, and showing that we are in the weak-anisotropy region. Again it should be emphasized that this result does not depend on the projection hypothesis being precisely obeyed. In Fig. 3 we have drawn arrows indicating the position of a few points if Eq. (4) were used instead of Eq. (5). The slope of the line would be slightly diminished, but the intercept, which is the important quantity, would be the same. Having presented strong evidence that δ is a viable spin-glass order parameter, we can make quantitative comparisons with mean-field theory. The most striking agreement with theory is, of course, the existence of a phase transition, as shown by the presence of a critical point in the scaling behavior of δ. The nature of the field-dependent transition agrees with the predictions of Gabay and Toulouse for the transverse freezing line. Given this compelling evidence for the GT line, along with the KS theory and the dynamical calculations of Fischer, one concludes that this is the true spin-glass transition and that the previously observed AT-like irreversibility lines are dynamical and not related to a phase transition. It is also clear that for fields above 250 Oe we are in the weak-anisotropy region of the KS theory. As can be seen in Fig. 3 a change in scaling occurs at high fields. The origin of this is not clear, but is most likely the result of a change in ϕ or a breakdown of Eq. (3) which might be expected in high fields. Preliminary results on Au-doped samples suggest that the latter is correct. The change in scaling does not effect the critical line since δ vanishes for the same value of τ/H^2.

The agreement with mean-field theory is not perfect, however. The measured characteristic field H_3 is smaller than mean-field predictions. Using the value of $4.4\mu_B$ per Mn atom[28] we calculate $H_3=67$ kOe, a factor of 5 larger than observed. The unavoidable presence of ferromagnetic coupling in *Cu*Mn will undoubtedly magnify the applied field. The molecular fields will then be larger than the external field by a factor of $(1-J_0/J)^{-1}$, where J and J_0 are, respectively, the width and mean of the distribution of exchange interactions.[29] From the high-temperature Curie-Weiss behavior we estimate this factor to be between 2 and 3 for $Cu\text{Mn}_{4\ \text{at.\%}}$. While this does not entirely account for the difference, the failure of mean-field theories to predict such constants accurately is not uncommon. The exponent σ cannot at this time be compared to mean-field theory since the magnitude of ϕ is not known. Careful measurements of M_r and K will be necessary to solve this problem.

In summary, we have demonstrated that the transverse spin-glass order parameter can be obtained experimentally. The scaling behavior of this order parameter is strong evidence for a true spin-glass phase transition, and that this transition occurs along the Gabay-Toulouse transverse freezing line. It is thus clear, at least in the vicinity of the phase transition, that mean-field theory is of quantitative value. Our results differ from recent torque measurements showing the existence of a nearly field-independent irreversibility line in $Cu\mathrm{Mn}_{1\,\mathrm{at.\%}}\mathrm{Au}_x$ alloys.[20,21] It is conceivable that the torque measurements were done in the intermediate-anisotropy region, where the transition line is insensitive to the field. Another possible explanation lies in the interpretation of the torque measurements. It is assumed that the torque experiments are sensitive to the degrees of freedom perpendicular to the induced macroscopic anisotropy. For small fields this is probably true. However, at high fields rotation of the macroscopic anisotropy will occur, obscuring the meaning of the torque data. We are presently extending these measurements to $Cu\mathrm{Mn}_{4\,\mathrm{at.\%}}\mathrm{Au}_x$ alloys to examine these possibilities, and to further explore the validity of the KS theory. The present study has provided a rare occasion in which spin-glass theory and experiment can be compared quantitatively.

We would like to acknowledge informative conversations with G. Kotliar and H. Sompolinsky. This research was supported by the National Science Foundation, Materials Research Laboratories (MRL) Program under MRL Grant No. DMR-83-16981.

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