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Critical effects in the reversible, nonlinear susceptibility of *Pd*Mn spin glasses

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The ac susceptibility of two *Pd*Mn spin glasses containing 5.0 and 5.5 at.% Mn has been measured as a function of temperature in a large number of static biasing fields between 0 and 500 Oe in the vicinity of the spin-glass (SG) temperature T_{SG}. Our analysis of the field dependence of these data both above and below T_{SG} provides the first experimental observation of a nearly symmetric divergence in the coefficients of the leading terms in H^2 and H^4 in the nonlinear reversible susceptibility as T approaches T_{SG} from above and below. The exponents which characterize the temperature dependence of the divergences are in good agreement with estimates obtained above T_{SG} by other authors.

After nearly two decades of both experimental and theoretical investigation, the question of whether the cusp in the zero-field ac susceptibility of spin glasses represents a true thermodynamic phase transition remains unresolved. Recently, the focus of these investigations has shifted towards examining the nonlinear components in the magnetization.[1-4] This change in emphasis was stimulated by theoretical predictions[5] that, when the magnetization is expanded in powers of the applied field H,

$$M(H,T)=\chi_0(T)H+a'(T)H^3+b'(T)H^5+\cdots , \quad (1)$$

the coefficients $a'(T)$ and $b'(T)$ of the leading nonlinear terms diverge as the spin-glass (SG) temperature T_{SG} is approached from above [$\chi_0(T)$ is the zero-field susceptibility]. Several recent experiments[1-4,6,7] have confirmed the general features of these predictions and have extracted the critical indices characterizing the temperature dependence of the divergence in these leading coefficients.

In the present paper, we report measurements of the ac susceptibility both above and below T_{SG} for two *Pd*Mn spin glasses containing 5 and 5.5 at.% Mn with spin-glass temperatures of $T_{SG}=2.840$ and 3.153 K, respectively. These data show that the leading coefficients $a'(T)$ and $b'(T)$ exhibit a nearly symmetric divergence about T_{SG}, a feature which, up to now, has not been observed.

We have chosen to investigate the *Pd*Mn system for a number of reasons. Our numerical calculations[8] based on an effective-field model indicate that the magnitude of these coefficients depends on the characteristics of the exchange distribution as well as on temperature; in particular, these coefficients are largest in systems where the ratio $\eta=\bar{J}_0/\bar{J}$ of the center to the width of the exchange distribution is close to but just less than 1. For *Pd*Mn, this ratio has recently been estimated[9,10] to be $\eta=0.97$ and 0.92 for the 5 and 5.5 at.% Mn samples, respectively. These concentrations also place T_{SG} in a convenient temperature range. Moreover, previous measurements on dilute ferromagnets[11] have shown that, under certain conditions, the reversible (ac) susceptibility in the vicinity of the ordering temperature is dominated by critical fluctuations, and indeed the present results appear to indicate that this is also the case for these *Pd*Mn spin glasses where spin-orbit coupling in the host leads to negligible irreversible effects[12] immediately below T_{SG}.

Figure 1 shows the ac susceptibility of the *Pd*–5.0 at.% Mn sample measured at 2 kHz in a driving field of 0.12 Oe rms as a function of temperature in the vicinity of the spin-glass temperature for several static biasing fields H. This figure clearly denonstrates the substantial field dependence of the susceptibility expected in a sample with η close to 1. Equation (1) implies a susceptibility of the form $\chi(H,T)=\chi_0(T)+a(T)H^2+b(T)H^4+\cdots$ and Fig. 2 illustrates the manner in which the H^2 coefficient $a(T)$ is obtained from the experimental data. Figure 2 also shows that as T approaches T_{SG} and the coefficient gets larger, the field range over which this H^2 term dominates decreases, in agreement with our numerical model calculations.[8] The data below T_{SG} exhibit a similar behavior. Figure 3 summarizes the temperature dependence of $a(T)$ for both the *Pd*–5 and 5.5 at.% Mn samples over the entire temperature range investigated. This figure clearly demonstrates the nearly symmetric divergence[13] in this coefficient about T_{SG}, obtained from the reversible susceptibility; this symmetric behavior has not been reported previously.

In an attempt to perform a quantitative analysis of the temperature dependence of the coefficient $a(T)$ in Fig. 3,

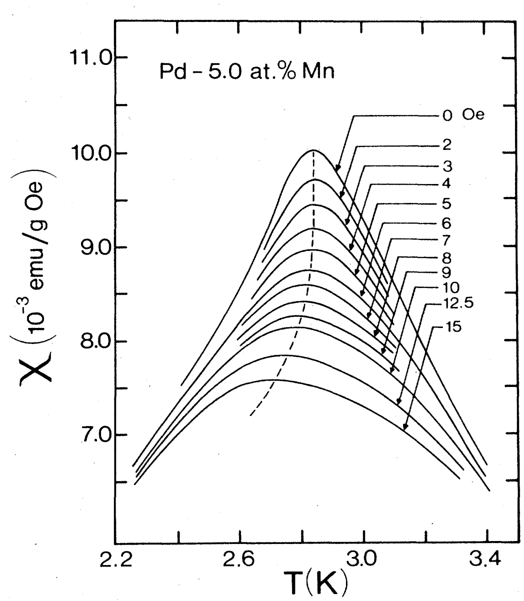

FIG. 1. The field-dependent ac susceptibility χ of *Pd*–5.0 at.% Mn plotted against temperature T in the vicinity of $T_{SG}=2.84$ K. The numbers marked against each curve are the static biasing fields.

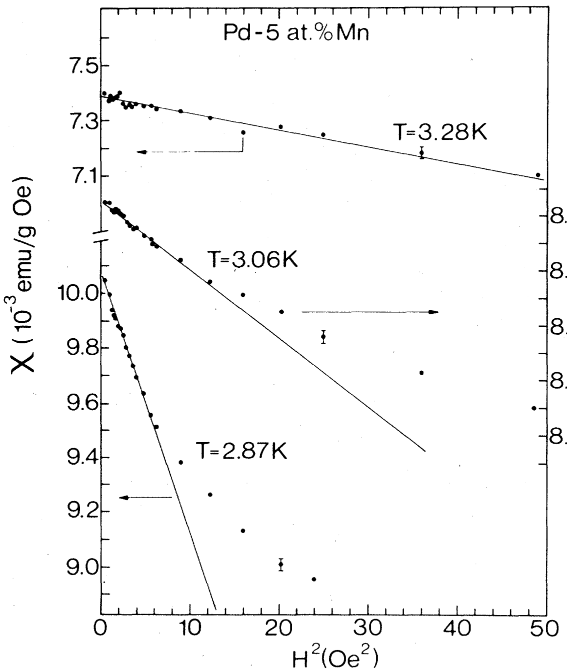

FIG. 2. The ac susceptibility of *Pd*–5 at.% Mn plotted as a function of H^2 for three different temperatures above T_{SG}.

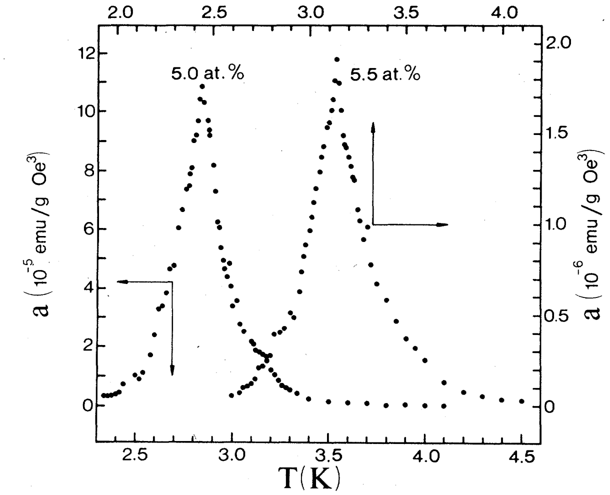

FIG. 3. The temperature dependence of the H^2 coefficient $a(T)$ for the *Pd*–5 and 5.5 at.% Mn samples.

we first point out that mean-field theory[5] predicts the H^2 coefficient to diverge as $t^{-\gamma_a}$ where $t=|(T-T_{SG})T_{SG}^{-1}|$, with $\gamma_a=1$ for $T>T_{SG}$. While mean-field theory is frequently correct in predicting the occurrence of a divergence, the precise values predicted for the exponents are seldom observed in real systems and, in fact, experimentally determined values[1,2,4] for γ_a above T_{SG} range from 3.25 to 3.65. From the double logarithmic plots in Fig. 4 we see that the value obtained for γ_a for *Pd*–5 at.% Mn from data *above* T_{SG} is $\gamma_a=3.19\pm0.34$, in good agreement with the other estimates. (The corresponding value for the *Pd*–5.5 at.% Mn sample is $\gamma_a=3.17\pm0.43$.) While the data in Fig. 4 extend down to a reduced temperature of $t\cong3\times10^{-3}$, which is closer to the conventional critical region than any previous investigation, the γ_a values are obtained over a restricted reduced-temperature range $t\geq9\times10^{-2}$. The question which naturally arises is whether the rounding of the data below this temperature represents a crossover to a regime with a smaller value for the exponent γ_a. We believe that this is not the case and that the rounding is caused either by attempting to define an H^2 coefficient over a reduced-field range which extends outside the region where this leading term dominates the critical behavior, or by dynamical effects. In ferromagnets, where critical effects appear directly in the susceptibility, the range of reduced fields h and temperatures t over which critical behavior can be observed is known to be limited.[14] In this regard the problem of investigating critical behavior in spin glasses is inherently more difficult since the relevant effects appear in the *nonlinear* susceptibility χ_{NL}. This requires the application of fields which are both large enough to establish deviations from linearity yet small enough to ensure that the critical behavior is dominated by, for example, the H^2 term. With this point in mind we have extended our measurements to lower static applied fields (down to 0.65 Oe) than other investigators. In spite of these precautions, a comparison between the experimental data of Fig. 2 and our numerical calculations[8] indicates that for reduced temperatures $t\lesssim2\times10^{-2}$, the region of validity of H^2 dominance in χ_{NL} is restricted to applied fields less than 1 Oe in this system. This is far smaller than that used experimentally to determine the H^2 coefficient in this temperature range, leading to an underestimate of the coefficient which becomes progressively more marked as $t\rightarrow0$.

Figure 4 also shows a similar plot for data acquired below T_{SG}. This is the new feature associated with the present investigation. It shows that the nonlinear ac susceptibility

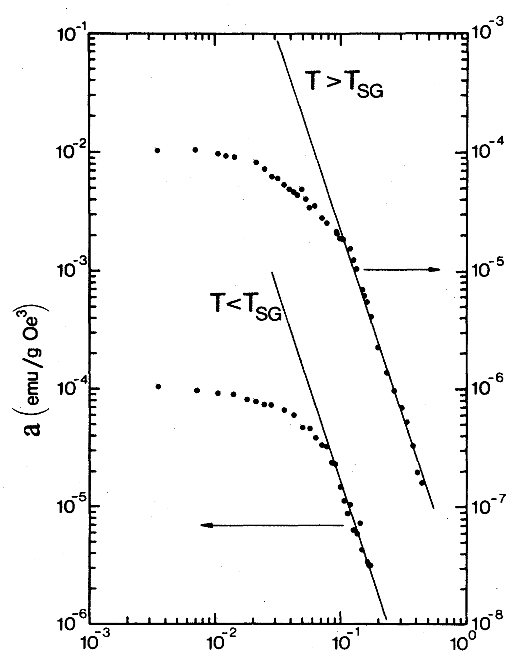

FIG. 4. Double logarithmic plot of a as a function of reduced temperature t above and below T_{SG} for *Pd*–5.0 at.% Mn. The estimates for γ_a and γ_a' were obtained from the slopes of the straight lines.

below T_{SG} diverges with a similar temperature dependence to that observed above T_{SG}; in fact, we obtain $\gamma_a' = 3.23 \pm 0.62$ for $t \geq 9 \times 10^{-2}$. (The corresponding value in the *Pd*–5.5 at.% Mn sample is $\gamma_a' = 3.20 \pm 0.43$.)

As mentioned in our discussion of the data in Fig. 2, deviations from the H^2 dominance of the nonlinear susceptibility become apparent as $T \rightarrow T_{SG}$ both from above and below. These deviations were analyzed for an H^4 contribution,[15] and the temperature dependence of the corresponding coefficient $b(T)$ is summarized in Fig. 5 for the two samples studied. We again wish to emphasize the nearly symmetric divergence in this coefficient about T_{SG}. Quantitative analysis based on double logarithmic plots of $b(T)$ as a function of reduced temperature t, both above and below T_{SG}, while subject to considerable error, lead to values for the temperature exponent of $\gamma_b(5\text{ at.\%}) = 7.5 \pm 1.4$ and $\gamma_b(5.5\text{ at.\%}) = 6.9 \pm 1.1$ above T_{SG}, in agreement with Omari, Prejean, and Souletie,[4] and $\gamma_b'(5\text{ at.\%}) = 7.5 \pm 2.7$ and $\gamma_b'(5.5\text{ at.\%}) = 6.8 \pm 1.6$ below T_{SG}, once again demonstrating the symmetric nature of the transition . As before, these γ values were obtained from a restricted temperature interval at the upper end of the experimental temperature range and above the region of severe flattening; specifically, the γ_b's are valid for $t \geq 10^{-1}$ and 1.2×10^{-1}, respectively, while the γ_b''s are valid for $t \geq 9 \times 10^{-2}$.

In summary, measurements of the reversible susceptibility of two *Pd*Mn spin glasses yield coefficients for the leading terms in H^2 and H^4 in the nonlinear susceptibility, which diverge in a nearly symmetric way as T approaches T_{SG} both from above and below; the critical exponents which characterize this temperature divergence agree very well with other estimates obtained above T_{SG} by other authors. We would like to point out that such a symmetric divergence is predicted by mean-field Ising models[8] provided that the usual coupled equation for m and q remain valid for temperatures just below T_{SG}. Such a situation does appear plausible in light of the phase diagram recently predicted by Kotliar and Sompolinsky[16] for spin glasses with random anisotropy, aspects of which have been verified experimentally.[17]

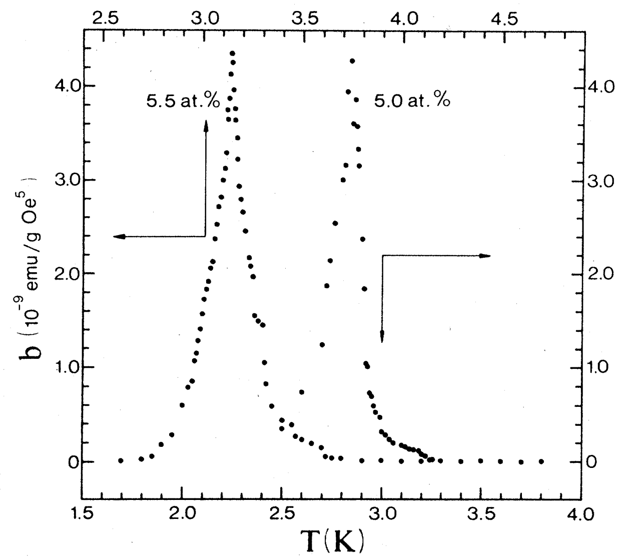

FIG. 5. The temperature dependence of the H^4 coefficient $b(T)$ for the *Pd*–5 and 5.5 at.% Mn samples.

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