Quenching of the Nonlinear Susceptibility at a $T = 0$ Spin Glass Transition

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$LiHo_{0.167}Y_{0.833}F_4$ is a dilute dipolar-coupled Ising magnet with a spin glass transition which can be crossed with temperature T ($T_g = 0.13$ K) or with an effective transverse field Γ ($\Gamma_g = 1$ K at $T = 0$). The nonlinear susceptibility contains a diverging component which dominates at $T = 98$ mK, but disappears by 25 mK. At the same time, the onset of spin glass behavior in the dissipative linear susceptibility becomes sharper. We conclude that, contrary to theoretical expectations, quantum transitions can be qualitatively different from thermally driven transitions in real spin glasses.

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The zero-temperature metal-insulator transition in disordered systems is the best known and most studied quantum phase transition [1]. Although quantum fluctuations undeniably drive the transition, the field has been handicapped by the difficulty of readily identifying a suitable order parameter. As an alternative, one can investigate the $T = 0$ physics of spin systems with quenched randomness [2–4]. Magnetic phase transitions in the classical limit have been investigated extensively, and, as thermal fluctuations give way to quantum fluctuations, one expects that the generalized magnetic susceptibility will continue to reveal the critical behavior.

The Ising spin glass in a transverse field is a simple and, as recently demonstrated [5], experimentally accessible example of a classical disordered magnetic system which can be converted into its quantum analog. The model Hamiltonian is

$$H = -\sum_{i,j}^{N} J_{ij}\sigma_i^z\sigma_j^z - \Gamma\sum_{i}^{N}\sigma_i^x , \qquad (1)$$

where the σ's are Pauli spin matrices and the J_{ij}'s are random longitudinal couplings. The commutator $[H,\sigma_j^z]$, which vanishes for zero transverse field Γ, becomes nonzero for $\Gamma \neq 0$, making Heisenberg's equation, $(\hbar/i) \times \partial\sigma^z/\partial t = [H,\sigma_j^z]$ germane. Thus, a finite Γ introduces channels for quantum relaxation which bypass the activation barriers determining the classical spin glass dynamics, and so depress the freezing temperature [5,6]. As these quantum fluctuations are tuned by the transverse field, a zero-temperature phase boundary arises between the spin glass and paramagnetic ground states.

Following our experience with thermally driven magnetic transitions, it is natural to ask about the critical behavior at the quantum spin glass transition, and to compare results in the zero-temperature and classical limits. In the present paper, we describe the first experiment to address this question. The key result is that the $T = 0$ transverse field-induced transition in the dipolar-coupled spin glass [7] $LiHo_{0.167}Y_{0.833}F_4$, as characterized by the linear and nonlinear susceptibilities, appears *first order.* At finite temperatures, the same transition is associated with a finite critical regime and a diverging nonlinear susceptibility, the hallmark of classical spin glass transitions [8].

$LiHo_{0.167}Y_{0.833}F_4$ is a site-diluted (nonmagnetic Y for magnetic Ho) and isostructural derivative of the dipolar-coupled Ising ferromagnet $LiHoF_4$ ($T_c = 1.53$ K). A magnetic field H_t applied perpendicular to the easy magnetic axis, which is parallel to c in this tetragonal material, yields a splitting of the Ho^{3+} ground state doublet [9]. It is this splitting Γ, proportional in lowest order to H_t^2, which plays the role of the transverse field in Eq. (1). We plot in Fig. 1 the spin glass paramagnet phase boundary in the Γ-T plane, determined from the dynamical behavior of the linear susceptibility (filled circles) [5]. Note that thermal fluctuations with $T_g = 0.13$ K are far more effective in destroying spin glass order than quantum fluctuations, for which the critical strength is $\Gamma_g(T = 0) = 0.98$ K.

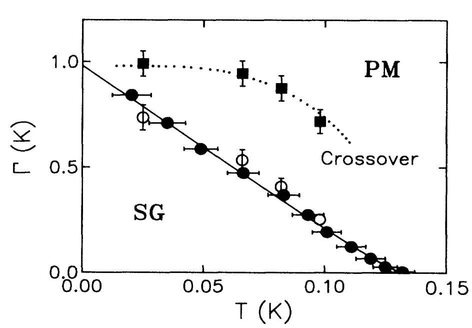

FIG. 1. Phase diagram of the diluted dipolar-coupled Ising spin glass $LiHo_{0.167}Y_{0.833}F_4$ in the transverse field (Γ)—temperature (T) plane. SG: spin glass; PM: paramagnet. Filled circles follow from dynamical measurements, open circles from the nonlinear susceptibility, and squares indicate where $\chi_1''(f = 1.5\ \text{Hz})$ begins to rise (see Fig. 3). The dotted line is the mean-field phase boundary associated with an ordered magnet whose critical temperature and field are the same as $T_g(\Gamma = 0)$ and $\Gamma_g(T = 0)$, respectively.

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We suspended single crystals of $LiHo_{0.167}Y_{0.833}F_4$ of typical dimensions $1.6\times1.6\times5$ mm^3 from the mixing chamber of a helium dilution refrigerator inside the bore of a superconducting magnet with field direction perpendicular to the Ising (long) axis of the crystals. A trim coil, oriented parallel to the Ising axis, provided a controllable longitudinal field h varying less than 0.1% over the sample region, and compensated for sample misalignment (less than 0.3° for each of three runs). We measured the total susceptibility, $\chi_{tot}(T,H_t,f)$, using a digital lock-in technique [10] and a standard low-temperature gradiometer configuration.

We show in Fig. 2(a) the real part of χ_{tot} as a function of h at $T=98$ mK and $f=1.5$ Hz for a series of transverse fields. Clearly, parabolas with small quartic corrections give an excellent account of the data, implying that, in practice,

$$\chi_{tot}(T,H_t,f)=\chi_1-3\chi_3h^2+5\chi_5h^4-\cdots . \quad (2)$$

In Eq. (2), χ is the linear susceptibility, while χ_3 is the lowest order nonlinear susceptibility. For our sample, fits of Eq. (2) to data such as those plotted in Fig. 2(a) show that χ_1, χ_3, and χ_5 are of order 100 emu/mol Ho, 10^{-4} emu/mol Oe^2, and 10^{-9} emu/mol Oe^4, respectively, requiring longitudinal fields of up to 100 Oe to discern χ_3 accurately. As expected given the large g-factor anisotropy of this dipolar-coupled Ising magnet, the data also show that terms containing odd powers of h and H_t, which one might consider adding to Eq. (2), are negligible. Indeed, the lowest order term of this type, proportional to H_th, would manifest itself as an offset in the χ_{tot} vs h curves in Fig. 2(a). Throughout our experiment, the only offsets observed were consistent with the less than 0.3° misalignments of the crystal and transverse field magnet axes. Finally, the imaginary part of χ_{tot} varies negligibly with h, implying that $|\chi_3''|<5\times10^{-6}$ emu/mol Oe^2 for the fields, temperatures, and frequencies probed in this experiment. As one approaches the spin glass transition from above ($H_t^c=5.74$ kOe), the parabolic curvature increases rapidly, indicating a sharp augmentation of the nonlinear contribution. Moreover, at $H_t=5.75$ kOe a clear signature of a higher order contribution to the susceptibility appears in the nonparabolic nature of the data at highest h. Although χ_5 is generally much smaller than χ_3, it is expected to diverge more strongly at the transition [8,11]. Varying χ_5 in our fit to χ_{tot} does not change the χ_1 and χ_3 values from those found when χ_5 was fixed at zero.

The lower frame of Fig. 2 illustrates the frequency dependence of χ_3 at this T. There is a strong rolloff for $f>10$ Hz which reflects the characteristic correlation time of the system [11]. Below $f=2$ Hz, however, χ_3 is essentially constant for all $H_t\geq H_t^c$. At lower T and correspondingly larger critical H_t, the roll-off frequency in-

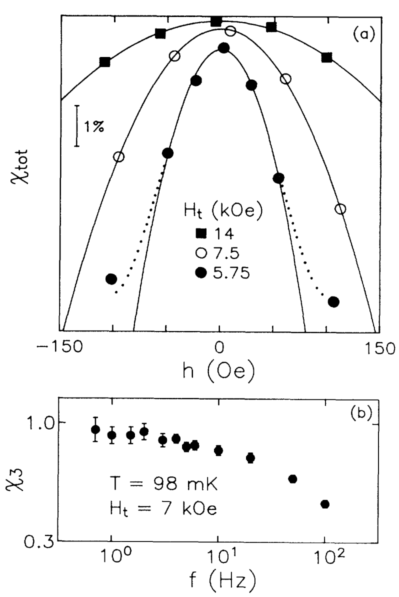

FIG. 2. (a) The total magnetic susceptibility [Eq. (2)] vs longitudinal field h for a series of transverse magnetic fields H_t at $T=98$ mK. The sharply increased parabolic curvature approaching the spin glass transition at $H_t=5.74$ kOe corresponds to the divergence of χ_3. At $H_t=5.75$ kOe, the dotted line includes a χ_5 component in the fit. (b) The frequency dependence of the nonlinear component. All fixed frequency data are taken at $f=1.5$ Hz, where χ_3 has saturated.

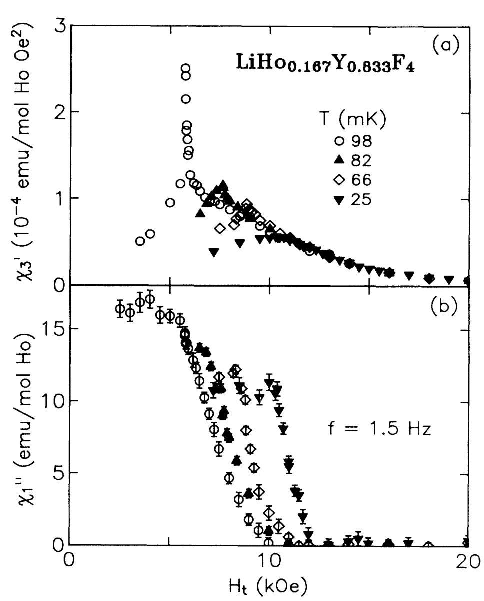

FIG. 3. (a) The nonlinear susceptibility as a function of transverse magnetic field at four temperatures. The clear divergence of χ_3 at the spin glass transition in the high T, small H_t (small Γ) classical limit becomes merely a small flat maximum in the low T, large H_t (large Γ) quantum limit. (b) Corresponding behavior of the imaginary part of the linear susceptibility. Here, there is a clear signature of the spin glass transition in both the classical and quantum regimes.

creases, exceeding 10^3 Hz at $T=25$ mK and $H_t=11$ kOe. Hence, data taken at 1.5 Hz maximize the inductive response while still being in the frequency-independent regime for all temperatures and transverse fields probed.

The primary result of our study is captured in Fig. 3. The divergence of the nonlinear susceptibility is seen as expected in the classical limit, but becomes suppressed and effectively disappears as $T \to 0$. By $T=25$ mK, χ_3 only shows a flat maximum. Furthermore, at this T and unlike what we find at 98 mK, we do not observe any deviation from a parabolic dependence on h up to 260 Oe for any H_t, precluding a diverging higher order susceptibility. The (unscaled) overlap of the $\chi_3(H_t)$ data above 12 kOe defines the regime where the splitting Γ of the Ising doublet dominates any thermal or spin-spin interaction energy.

The quenching of the divergence of the nonlinear susceptibility in the quantum limit raises the question of whether a well-defined spin glass transition still occurs. We show in Fig. 3(b) simultaneous measurements of the imaginary part of the linear susceptibility in the zero frequency limit. There is a clear dynamical signature of the transition at all temperatures, even sharpening at low T and high H_t. With increasing H_t, χ_1'' decreases sharply from its low H_t plateau at a critical field indistinguishable from that determined by the χ_3' measurement. Thus, χ_3' at $H_t=5.74$ kOe is indeed most singular at the glass transition defined by the spin dynamics. As described elsewhere [5], the sudden increase in the dissipation is due to the development of a flat, frequency-independent tail in $\chi_1''(f \to 0)$, which, by the fluctuation-dissipation theorem, corresponds to characteristic $1/f$ noise in the spin glass magnetization [12].

We can fit the divergence of the nonlinear susceptibility at $T=98$ mK to the critical form, $\chi_3 \sim [(\Gamma-\Gamma_g)/\Gamma_g]^{-\gamma_{\mathrm{eff}}}$. As seen in Fig. 4, this form fits well over the range 10^{-3} to 1 in the reduced variable, coinciding with the interval over which χ_1'' changes from zero to its plateau value [Fig. 3(b)]. We find best fit values $\Gamma_g=0.253(8)$ K and $\gamma_{\mathrm{eff}}=0.20(2)$. By comparison, Monte Carlo simulations [13] of the classical short-range Ising spin glass find critical exponents $\gamma=2.9(1)$, while experiments on both metallic Ag:Mn and insulating $Eu_{0.4}Sr_{0.6}S$ spin glasses [11] yield diverging nonlinear susceptibilities with $\gamma=2.3(2)$.

The values for $\Gamma_g(T)$ determined from χ_3 are plotted as open circles in the phase diagram of Fig. 1, along with the squares which mark the onset of dissipation. The open circles fall on the phase boundary defined by the appearance of frequency-independent behavior at small f in $\chi_1''(f)$. While this phase boundary is simply a straight line, the crossover line delineated by the squares displays considerable curvature. It can be described using the mean-field formula for the phase boundary of an ordered magnet in a transverse field [14], $\coth(\Gamma/\alpha k T_c)=J/\Gamma$,

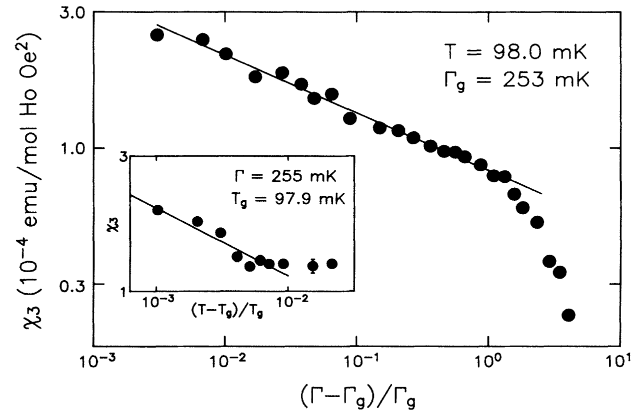

FIG. 4. Critical behavior of the nonlinear susceptibility in both reduced transverse field and reduced temperature (inset), described by the same effective critical exponent (slope). Axes scale by the ratio of the relative strengths of the quantum and thermal fluctuations (see text).

where J is a measure of the interaction between spins and α is a dimensionless constant associated with the effective ratio of ground and excited state degeneracies. This ratio rises above the single ion value of two with the complexity of the fundamental units, e.g., clusters of strongly coupled spins [7], undergoing the mean-field transition. The parameter values yielding the best account (dotted line in Fig. 1) of the data are $\alpha=7.9(4)$ and $J=1.00(6)$ K. The calculated $T_c(\Gamma=0)=J/\alpha=0.13\ \mathrm{K}=T_g(\Gamma=0)$, thus making the measured ($T\leq 0.098$ K) crossover points for $LiHo_{0.167}Y_{0.833}F_4$ lie on the mean-field transition line associated with an ordered magnet whose critical temperature and field are the same as $T_g(\Gamma=0)$ and $\Gamma_g(T=0)$, respectively. That $J=\Gamma_g(T=0)$ is much closer to $T_c(x=1)$ than it is to T_g or even the mean-field ferromagnetic $T_c(x=0.167)=xT_c(x=1)=0.26$ K, underlines the great importance of rare spin configurations involving strong J_{ij}'s in the quantum regime.

The linear spin glass phase boundary of Fig. 1 mandates the same effective exponent whether approaching the transition in Γ or T. We verify this accord in the inset to Fig. 4, where the decade spacing is the same as in the main part of the figure. χ_3 diverges with $(T-T_g)/T_g$ at $\Gamma=255$ mK, giving a best fit effective critical exponent $\gamma_{\mathrm{eff}}=0.24(6)$. The data sets, in fact, can be shifted horizontally on top of each other by simply multiplying the abscissa of the inset by 7.5, the ratio of $\Gamma_g(T=0)/T_g(\Gamma=0)=(0.98\ \mathrm{K})/(0.13\ \mathrm{K})$, the quantum and classical end points of the spin glass phase diagram (Fig. 1).

This correspondence allows us to rule out single ion effects as a determining factor in the divergent properties of χ_3. Although χ_3 can vary rapidly with Γ as $1/\Gamma^3$ at fixed T, it does not increase appreciably with decreasing T at fixed Γ because of the singlet nature of the transverse-field split ground state. Calculations in the full 17×17 eigenfunction space of the Ho^{3+} ion do indeed confirm that the single ion nonlinear susceptibility is effectively independent of T over the reduced temperature range of the inset to Fig. 4.

Mean-field treatments of both infinite-range quantum Ising [15] and quantum rotor [16] spin glasses predict unusual behavior for the nonlinear susceptibility at the $T=0$ transition, but retain its divergence. Specifically, they give $\gamma_{\rm eff}=\frac{1}{2}$ and a phase boundary $\Gamma_g(T)-\Gamma_g(T=0)\sim T^2$ at low T. Our experiment yields $\gamma_{\rm eff}<\frac{1}{2}$ at all T measured, with an essentially linear phase boundary. If we attempt to fit the weak divergence of χ_3 with transverse field by a critical form for the lower temperatures, then we are faced with the physically unreasonable situation of a progressively decreasing, temperature-dependent critical exponent, $\gamma_{\rm eff}$, indistinguishable from zero by $T=25$ mK. Hence, in the $T=0$ limit, the classical view that the nonlinear susceptibility diverges at a spin glass transition does not seem to apply. Yet, the dynamical evidence clearly points to an extant transition with an appreciable crossover region.

One response to our experiments is that the phenomena we find is somehow related to the suppression of χ_3 in classical Heisenberg spin glasses on application of modest external fields [11]. This is highly improbable given (1) the care we have taken to trim and align the transverse field perpendicular to the measuring field and the Ising axis of the crystal, (2) the fact that for a corresponding field-induced reduction in χ_3, T_g is reduced very substantially for $LiHo_{0.167}Y_{0.833}F_4$, but not for the classical systems, and (3) the underlying Hamiltonians and measurements are fundamentally different. In particular, an effective Ising (not Heisenberg) Hamiltonian, Eq. (1), describes $LiHo_{0.167}Y_{0.833}F_4$, and in contrast to the studies of Ref. [11], our experiment features a measuring field h oriented perpendicular to the applied field H_t.

Even though it is difficult to relate our results to previous experiments on spin glasses, a number of possible explanations exist. The nonlinear susceptibility may no longer couple to the order parameter in the quantum case [17], a radical suggestion given its observed divergence in the classical limit and the dramatic step in $\chi_1''(f)$ at Γ_g for low T. More likely, the $T=0$ quantum spin glass transition is first order. While the nonergodicity of the spin glass state precludes the standard test of hysteresis at a first order transition, this hypothesis is consistent with both the abrupt onset of linear dissipation and the absence of a pretransitional divergence of χ_3 (and the spin glass correlation length). Moreover, in quantum systems of finite size, first order transitions in the form of level crossings are the rule rather than the exception. It is plausible, especially given the large critical transverse field at $T=0$ in $LiHo_{0.167}Y_{0.833}F_4$, that level crossings associated with strong but relatively rare bonds dominate the physics, as occurs in random transverse-field Ising chains [4].

In summary, we tune a physical realization of the Ising spin glass in a transverse field [Eq. (1)] from the $\Gamma=0$ classical to the $T=0$ quantum limit. Both regimes are accessible for kOe magnetic fields and mK temperatures in our system of randomly distributed dipoles because of suitable g-factor anisotropies and crystal field splittings. We find a clear dynamical signature of the spin glass to paramagnet transition whether dominated by thermal or quantum fluctuations. However, in contrast to the obvious second order nature of the classical transition, the quantum counterpart appears first order.

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