# From Classical to Quantum Glass

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We study the effects of a transverse magnetic field on the dynamics of the randomly diluted, dipolar coupled, Ising magnet $LiHo_{0.167}Y_{0.833}F_4$. The transverse field mixes the eigenfunctions of the ground-state Ising doublet with the otherwise inaccessible excited-state levels. We observe a rapid decrease in the characteristic relaxation times, large changes in the spectral form of the relaxation, and a depression of the spin-glass transition temperature with the introduction of quantum fluctuations.

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Ensembles of randomly distributed magnetic spins with competing interactions exhibit many of the hallmarks of glassy behavior. As the temperature is lowered, long-time relaxation processes become important, leading to a slowing down and a broadening in the dynamic response. At sufficiently low temperature, the spins freeze with zero net magnetization. Intense theoretical and experimental efforts over many decades have revealed essential aspects of the freezing process [1], where the spins are considered as classical degrees of freedom. Only at the lowest temperatures, well below the glass transition, does quantum mechanics become important as different parts of the free-energy surface can be linked through tunneling.

If present in sufficient strength quantum fluctuations could have an enormous impact on the nature of the collective interactions as the spins start to freeze, and on the actual transition itself. New routes to relaxation become available, possibly even threatening the stability of the spin-glass state. The Ising model for $N$ interacting spins in transverse field $\Gamma$, with Hamiltonian

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

is a theoretical construct which introduces quantum mechanics to a classical problem in a natural way. Here, spins $i$ and $j$ are connected by a random exchange $J_{ij}$ and the $\sigma$'s are Pauli spin matrices. The transverse field serves the role of an operator which mixes formerly pure spin eigenstates. Various authors [2–4] have considered spin glasses in transverse fields, with predictions ranging from the destruction of the spin-glass state [3] to an enhancement in the transition temperature [4] with the introduction of quantum fluctuations. We report here static and dynamic measurements on a physical realization of the Ising spin glass in a transverse magnetic field. While proton tunneling in the glass state of mixed hydrogen-bonded ferroelectrics [5] can be represented by a transverse field $\Gamma$, our magnetic system permits $\Gamma$ to be tuned all the way from the classical to the quantum limit.

$LiHo_xY_{1-x}F_4$ is an isostructural dilution series where magnetic $Ho^{3+}$ and nonmagnetic $Y^{3+}$ ions randomly occupy the rare-earth ($R$) sites in the body-centered tetragonal $LiRF_4$ lattice [6]. The single-ion anisotropy is Ising with the moments ($\mu_{\rm eff}=7\mu_B$) derived from the ground-state doublet of $Ho^{3+}$ lying parallel to the $c$ axis. The dominant interaction between moments is dipolar, as directly demonstrated by neutron diffraction [7,8]. The pure compound ($x=1$) is a ferromagnet, with an essentially perfect (due to the long range of the dipolar interaction) mean-field transition [9] at $T_c=1.53$ K. Sufficient dilution destroys any long-range order, and the disorder and frustration (arising from the mix of ferromagnetic and antiferromagnetic interactions in a dipolar-coupled system) combine to make an $x=0.167$ sample a spin glass with a transition temperature $T_g$ of order 0.1 K [8].

The first excited crystal-field level in $LiHo_xY_{1-x}F_4$ is 9.4 K above the ground-state doublet. At the low temperatures of our experiments, $0.025 \leq T \leq 0.25$ K, only the Ising doublet is appreciably populated. As the states in the doublet are eigenstates of $J_{zz}$ (z$\parallel$c), the dipoles have no first-order response to a magnetic field $H_t$ applied perpendicular to the $c$ (Ising) axis. $H_t$, however, can mix the eigenfunctions of the ground-state doublet, the first excited-state singlet, and, to a lesser extent, the higher-level states. We plot in the inset of Fig. 1 the evolution with $H_t$ of the lowest eigenvalues of the Hamiltonian for the single ion in the $LiRF_4$ crystal field. The principal effect of $H_t$ is to introduce a splitting $\Gamma \propto H_t^2$ (at low $H_t$) of the doublet ground state. It is $\Gamma$ and not $H_t$ which appears in the model Hamiltonian of Eq. (1). In this context, the eigenstates of $\sigma_i^x$ correspond to the two lowest-lying states of the single-ion Hamiltonian.

We use the dynamic magnetic response of the system as a whole to probe the evolution of the superposition of eigenstates with $H_t$, performing, in essence, a *collective* electron-spin-resonance experiment. We measured the complex ac susceptibility, $\chi(f)=\chi'(f)+i\chi''(f)$, of a single crystal of $LiHo_{0.167}Y_{0.833}F_4$ over the frequency range $10^{-1}$–$10^5$ Hz using a computer-based digital lock-in

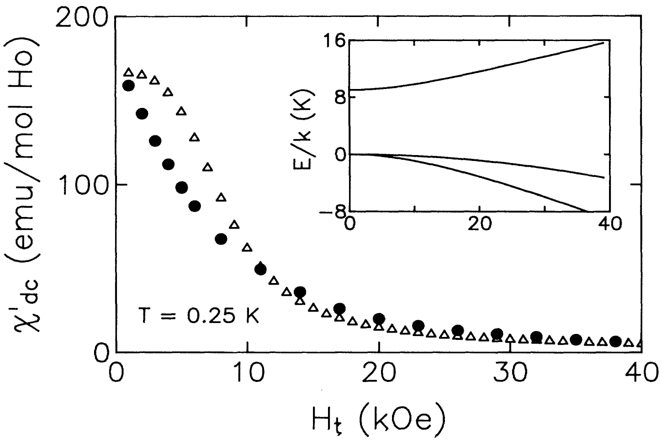


FIG. 1. The measured dc magnetic susceptibility $\chi'_{dc}$ (solid circles) as a function of transverse magnetic field $H_t$ for the Ising spin glass $LiHo_{0.167}Y_{0.833}F_4$ at a temperature approximately twice $T_g$. Open triangles are a mean-field calculation (see text). Inset: Evolution with $H_t$ of the single-ion energy levels. When the splitting between the $H_t=0$ Ising doublet becomes larger than any other energies involved ($H_t \gtrsim 10$ kOe), quantum mixing of the states dominates and the mean-field approximation holds.

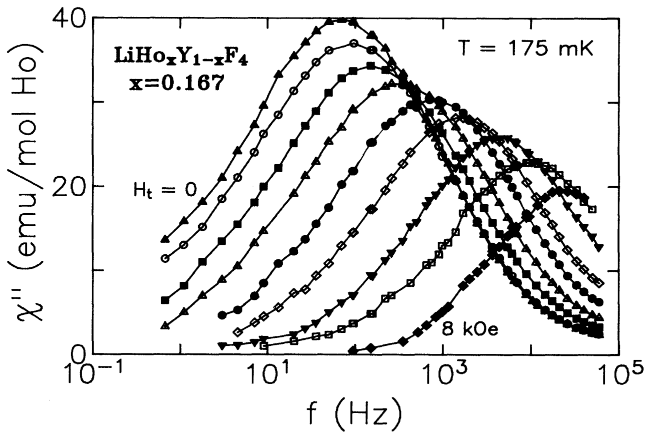


FIG. 2. Imaginary part of the susceptibility $\chi''$ over many decades in frequency $f$ at $T > T_g$ for a series of transverse magnetic fields $H_t$ in 1-kOe intervals. The glassy response speeds up dramatically as the transverse field introduces new quantum routes to relaxation.

technique [10]. The sample of dimensions $1.6\times1.6\times5.1$ $mm^3$ was suspended from the mixing chamber of a helium dilution refrigerator inside the bore of an 80-kOe superconducting magnet, with its Ising (long) axis oriented perpendicular to the field direction. Heat sinking was achieved via sapphire rods, spring loaded inside a standard gradiometer configured to measure the $\hat{z}$ component of $\chi$. A compensation coil, oriented parallel to the sample's Ising axis, was used to null out any longitudinal field component by maximizing the $\chi'$ response at each transverse field. From the values of the compensating field, we found geometric misalignments of 0.5° and 0.3°, respectively, in two separate runs.

We show in the main part of Fig. 1 the measured transverse field dependence (solid circles) of $\chi'_{dc}$, the $f\to 0$ limit of $\chi'(f)$, in the paramagnetic state at $T=0.25$ K. We also plot the mean-field result [11] (open triangles), $\chi_{mf}=\chi_0/[1-(\theta/C)\chi_0]$, where $\theta=0.16$ K is the ($H_t=0$) Curie-Weiss temperature previously determined [8] for $LiHo_{0.167}Y_{0.833}F_4$, $C$ is the Curie constant given by the free-spin moments, and $\chi_0$ is the single-ion susceptibility [12]:

$$\chi_0=\frac{2}{Z}\sum_{m\neq n}\frac{|\langle m(H_t)|g\mu_B J_z|n(H_t)\rangle|^2}{\varepsilon_n(H_t)-\varepsilon_m(H_t)}\times\exp[-\varepsilon_n(H_t)/kT]. \quad (2)$$

Here, $\varepsilon_n(H_t)$ is the energy of the $n$th eigenstate in the presence of a transverse magnetic field $H_t$ and $Z=\sum_n\exp[-\varepsilon_n(H_t)/kT]$. In our calculations, we have used the known [13] crystal-field level scheme and wave functions of $Ho^{3+}$ ions in $LiRF_4$ and we have restricted the sum in Eq. (2) to the two states that evolve with transverse field from the $H_t=0$ doublet, shown in the inset of Fig. 1. The in-field eigenstates $|n(H_t)\rangle$ were computed via explicit diagonalization of the transverse-field Hamiltonian $g\mu_B H_t J_x$ in the full $17\times17$ $Ho^{3+}$ eigenfunction space.

This simple mean-field theory, without any adjustable parameters, accounts well for the data in the high-field limit. When $H_t>10$ kOe, the splitting of the $H_t=0$ Ising doublet is much greater than $\theta$ and the quantum mixing of the states dominates the physics. At smaller transverse fields the exact nature of the spin-spin interaction is important, and the mean-field approximation is seen to be inadequate. Of particular interest is the apparent approach of $\chi'_{dc}$ to its $H_t=0$ value with finite slope, while $d\chi'_{mf}/dH_t|_{H_t=0}$ clearly vanishes. This unusual behavior may be a manifestation of Griffiths singularities [14], which are expected because, even though $T=0.25$ K is well above $T_g$, it is well below the Curie point (1.53 K) of the ferromagnetic parent $LiHoF_4$.

Quantum-mechanical effects are most apparent in the frequency-dependent response [15]. In Fig. 2 we show $\chi''(f)$ for various $H_t$ at $T=0.175$ K, a temperature still above the glass transition. The broad spectral width of up to 3 decades in frequency FWHM (compared to the single time Debye fixed width of 1.14 decades) reflects the multiple routes to relaxation characteristic of glassy systems. The application of a transverse field radically affects the time scale of the Ising system's response. The frequency of the peak $f_p$ of $\chi''$ increases by 2 orders of magnitude in only 6 kOe. Moreover, the low-frequency tails of $\chi''(f)$ are greatly suppressed, indicating that the new quantum routes to relaxation most profoundly affect the long-time modes. We have checked that a longitudinal field primarily depresses the amplitude of the

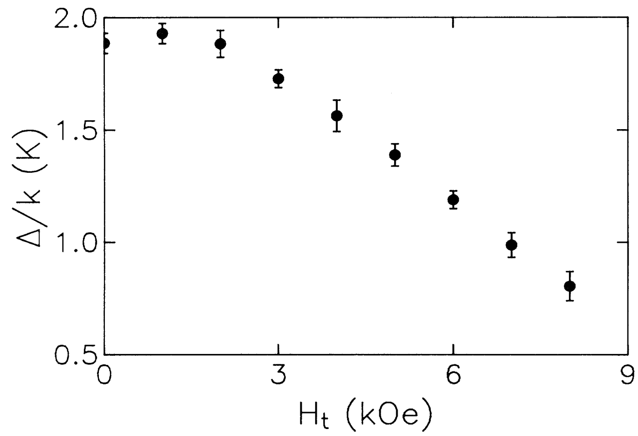


FIG. 3. Energy barrier to relaxation, Δ, from the Arrhenius law for $T > T_g$ as a function of transverse field.

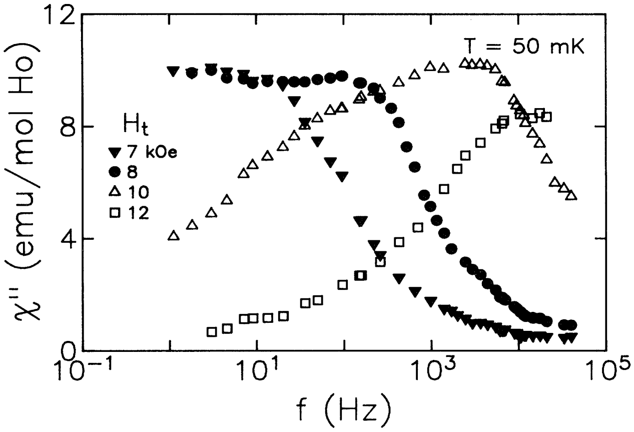


FIG. 4. The counterpart to Fig. 2 for $T < T_g(H_t = 0)$. The flat response at low $f$ for small transverse fields is characteristic of spin glasses. With increasing $H_t$ the spectral response becomes paramagnetic.

response and cannot account for the large observed shifts in $f$.

We can parametrize the dependence of $f_p$ on $H_t$ through the Arrhenius law familar from studies of glasses: $f_p = f_0 \exp(-\Delta/kT)$, where $f_0$ is a microscopic attempt frequency, $k$ is Boltzmann's constant, and $\Delta$ is an energy barrier to relaxation. As expected, the Arrhenius form describes our data for $T$ well above $T_g$, and we plot in Fig. 3 the progression with transverse field of the energy barrier $\Delta$. Halving $\Delta$ profoundly affects $f_p$ because of the exponential dependence in the Arrhenius law. Qualitatively, we can understand the result of Fig. 3 as arising from the basal-plane $g$ factor induced by $H_t$, presumably lessening the energy barrier for a spin to pass through the plane as it changes its orientation along the Ising axis.

Below $T_g(H_t = 0)$, the frequency dependence of the magnetic susceptibility is most unusual. In Fig. 4 we plot $\chi''(f)$ for various $H_t$ at $T = 0.05$ K. In sufficient transverse field, $\chi''(f)$ mimics the data for $T > T_g(H_t = 0)$ (see Fig. 2). As $H_t$ is decreased, the spectral response also moves to lower $f$. However, it becomes enormously broad and even essentially flat over decades in frequency as $f \rightarrow 0$.

The $f \rightarrow 0$ limit of $\chi''(f)$ can be described by the power-law form $f^\alpha$. We have used this form to fit the low-frequency portion of our data and we show in Fig. 5 the results for $\alpha$ vs $H_t$ at several $T$. In other spin glasses, most notably $Eu_xSr_{1-x}S$, $T_g$ is associated [16] with $\alpha$ approaching and then saturating at a small value, $\lesssim 0.1$. Furthermore, it is generally believed that the spin-glass state itself is characterized by $1/f$ noise in the magnetization [17], which implies, via the fluctuation-dissipation theorem, that $\alpha = 0$. We therefore identify the temperature at which $\alpha$ approaches zero with $T_g$. Defining $T_g$ in this manner is not possible at $H_t = 0$ because $\alpha \rightarrow 0$ at a frequency too low for us to probe. Given that the effect of the transverse field is to introduce quantum fluctuations which aid the relaxation process, shifting the spectral response to higher $f$ and increasing $\alpha$, we expect $T_g$ to decrease with increasing $H_t$. In addition, the application of $H_t$ moves the spin-glass signature, $\alpha = 0$, into an accessible frequency window. At $T = 0.15$ K, $\alpha$ never reaches zero, indicating that $T_g(H_t = 0) < 0.15$ K. As $T$ is lowered, we find increasingly robust regions of flat response, with $\alpha$ remaining zero to progressively higher $H_t$.

In the inset of Fig. 5 we show $T_g$ as a function of $H_t$, where the open circles are determined by plotting $\alpha$ vs $H_t$ at constant $T$ (as in the main part of the figure) and the solid circles are determined by plotting $\alpha$ vs $T$ at constant $H_t$. The solid line is a least-squares fit by the form $T_g(H_t) = T_g(0)[1 - (H_t/H_c)^\beta]$, with $T_g(0) = 0.133$

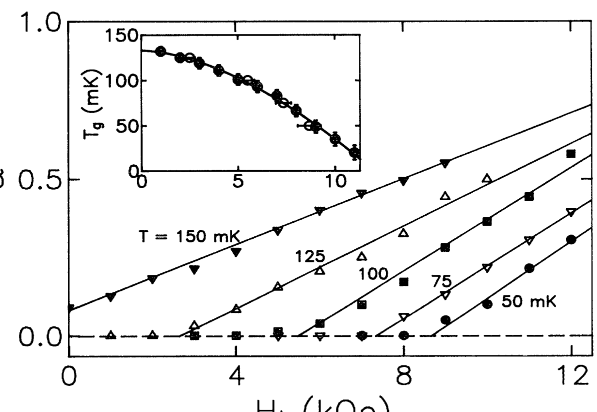


FIG. 5. The power in the form $f^\alpha$ fitted to the $f \rightarrow 0$ limit of $\chi''(f)$ vs transverse field at five different temperatures. We define spin-glass freezing when $\alpha \rightarrow 0$. Inset: Depression with $H_t$ of the spin-glass transition temperature $T_g$ so determined. Solid line is a least-squares fit with $T_g(0) - T_g(H_t) \sim H_t^{1.7 \pm 0.1}$ (see text).

$\pm 0.005$ K and $\beta = 1.7 \pm 0.1$. If this functional form continues below our lowest-temperature data at $T = 25$ mK, then the critical transverse field to completely suppress spin-glass freezing is $H_c = 12.0 \pm 0.4$ kOe. This corresponds to a splitting $\Gamma/k = 1.0$ K in the model Hamiltonian, Eq. (1). This is in contradiction to the theoretical expectation [2] that $\Gamma/k \sim T_g(H_t = 0) = 0.13$ K. Thus, thermal fluctuations appear to more easily destroy the spin-glass state in $LiHo_{0.167}Y_{0.833}F_4$ than do quantum fluctuations.

In summary, we have investigated an experimental realization of the Ising spin glass in transverse magnetic field. Modest $H_t$, of order 1–10 kOe, profoundly modify the classical (zero-field) behavior of randomly distributed dipoles, and allow us to introduce quantum fluctuations in a controlled fashion.

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