

Quantum and Classical Glass Transitions in $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$

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When performed in the proper low field, low frequency limits, measurements of the dynamics and the nonlinear susceptibility in the model Ising magnet in transverse field, $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$, prove the existence of a spin glass transition for $x = 0.167$ and 0.198 . The classical behavior tracks for the two concentrations, but the behavior in the quantum regime at large transverse fields differs because of the competing effects of quantum entanglement and random fields.

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Research on spin glasses has led not only to deep insights into disordered materials and the glassy state, but has generated novel approaches to problems ranging from computer architecture through protein folding to economics. The rugged free energy landscape characteristic of such systems defies usual equilibrium analyses, with pronounced non-linear responses and history dependence. From the discovery of spin glasses 35 years ago [1], it has been appreciated that measurements in the low frequency and small magnetic field limits are essential to derive meaningful results.

At low temperatures, and in cases where barriers to relaxation are tall and narrow, quantum mechanics can enhance the ability to traverse the free energy surface [2]. The $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ family of materials represents the simplest quantum spin model, the Ising magnet in transverse field, and it has been an especially useful system to probe the interplay of disorder, glassiness, random magnetic fields and quantum entanglement [3, 4, 5, 6, 7, 8, 9]. The parent compound, LiHoF_4 , is a dipole-coupled Ising ferromagnet with Curie temperature $T_C = 1.53\text{K}$. Applying a magnetic field H_t transverse to the Ising axis introduces quantum mixing of classical spin-up and spin-down eigenstates, or equivalently tunes the tunneling probabilities for walls between patches of ordered spins [10]. Hence, quantum fluctuations controllable by an external field can drive the classical order-disorder transition to zero temperature, resulting in a much studied ferromagnetic quantum critical point [11, 12, 13].

The nature of the ground state can be tuned by partially substituting non-magnetic Y for the magnetic Ho [14]. Dilution enhances the effects of the frustration inherent in the dipolar interaction, with the ferromagnet giving way to a spin glass at $x \sim 0.2$ (Fig. 1). The transverse component of the dipolar coupling introduces other phenomena. At large x , the ground state becomes a disordered ferromagnet, with the low temperature dynamics dominated by domain wall tunneling [10]. The high T and low H_t behavior reveal both the effects of Griffiths singularities [15] and the internal random fields due to the application of a uniform H_t to a disordered Ising

magnet [6, 7, 16]. For $x \lesssim 0.1$, the internal transverse fields induce quantum entanglement that prevents the system from freezing and stabilizes a spin liquid “antiglass” phase down to very low temperatures [5, 17]. In the intermediate range, where the spin glass phase is stable, the tendencies towards ferromagnetism and random field effects found at high x compete with the massive quantum entanglement of the antiglass. This competition might be expected to lead to very different statics and dynamics for small changes in x . Fig. 1a illustrates the essential physics. In the classical, $H_t = 0$ case, a small random field of strength h at site 1 and 0 at site 2 will produce a splitting of order h between the degenerate $|\uparrow\downarrow\rangle, |\downarrow\uparrow\rangle$ classical ground states. On the other hand, $H_t \neq 0$ will yield a single non-degenerate ground state $|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle$, on which the only effect of a small h will be an energy change $\sim h^2$.

The first work on the spin glass state in $\text{LiHo}_{0.167}\text{Y}_{0.833}\text{F}_4$ revealed a well-defined transition from the paramagnet to the glass, evidenced by both a sharp divergence in the nonlinear susceptibility, χ_3 , and a dynamical signature in the dissipative component of the linear susceptibility, χ_1'' [3, 4]. These measurements relied on H_t to speed up the system dynamics to the point where the $f \rightarrow 0$ limit could be probed directly, and led to considerable theoretical work [9, 15, 18], some with good qualitative agreement with our experiments [19]. Within the last three years, the concept that randomly placed classical dipoles [20] should undergo a spin glass transition has itself been questioned on account of numerical work on small cubic lattices [21]. More recent experiments [22] employing a μSQUID magnetometer did not reveal a divergence in χ_3 , leading the authors to a similar conclusion, namely that $\text{LiHo}_{0.167}\text{Y}_{0.833}\text{F}_4$ is not a spin glass. Unfortunately, the authors of [22] used large longitudinal fields and fast sweep rates, probing the system very far from equilibrium, and thus obscured the meaning of their data. In this Letter, we show explicitly that when data are acquired in the proper small longitudinal field, low frequency limit, clear evidence is seen for a classical spin glass transition. Moreover, we report the discovery that a minor change

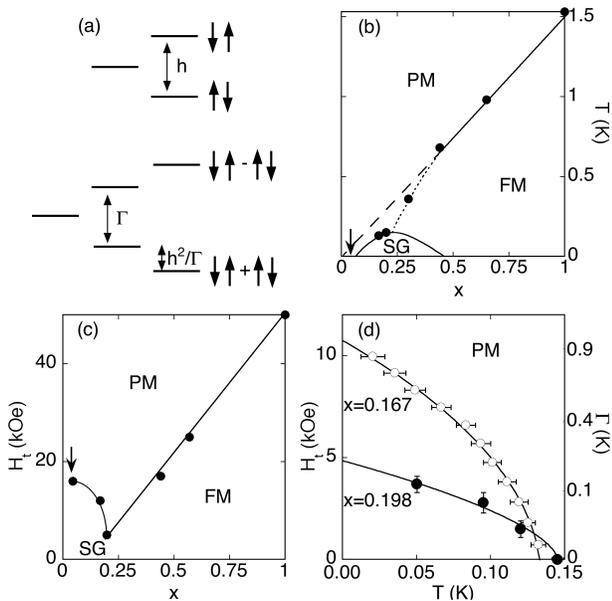


FIG. 1: Energy levels and phase diagram for $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ (a) Schematic of energy levels for antiferromagnetically coupled spins in a (top) random field and (bottom) uniform transverse field Γ , and then a random field h . (b) Magnetic behavior vs. x and temperature T . Arrow denotes spin liquid “anti-glass” phase. (c) Critical transverse field, above which $\chi''(H_t)$ decreases, vs. x . (d) Spin glass/paramagnet phase boundaries for $x = 0.167$ and 0.198 .

in x from 0.167 to 0.198 - approaching the multicritical point where spin glass and ferromagnetic phases coexist (Fig. 1b) - results in dramatic changes in the quantum (H_t -dependent) behavior.

We performed ac susceptibility measurements from 1 to 10^5 Hz on single crystal needles of $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ mounted on the cold finger of a dilution refrigerator. The magnitude of χ_1 was consistent with the Curie-Weiss law for Ho^{3+} ions at high T . The susceptibility was corrected for demagnetization effects using the demagnetization factor of rods of the same aspect ratio. Ho concentrations x were determined to ± 0.001 by a differential weighing technique. Static transverse magnetic fields up to 8 T and longitudinal fields, h_ℓ , up to 0.03 T were supplied by a superconducting solenoid and Helmholtz coils, respectively. The ac excitation amplitude was restricted to less than $A = 0.02$ Oe to ensure linear response and to control heating. For measurements of the nonlinear susceptibility, the dc longitudinal field was swept at 0.04 Oe/s so that this rate was smaller than the effective sweep rate at $f = 1.5$ Hz of $2\pi Af = 0.2$ Oe/s.

We plot in Fig. 1d the $T - H_t$ phase diagrams for both the $x = 0.167$ and 0.198 spin glasses. The transition is defined by the emergence of a flat spectral response at low f in χ''_1 , corresponding to $1/f$ noise in the magnetization. This dynamically-determined phase boundary coincides with that derived from the maxima of $\chi_3(f \rightarrow 0)$. The

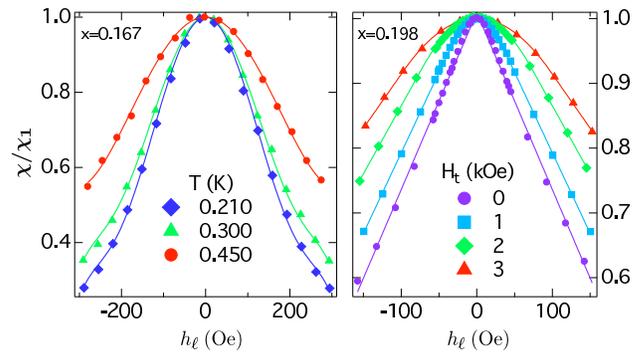


FIG. 2: (color online) Scaled susceptibility for $x = 0.167$, $H_t = 2.1$ kOe (left) and $x = 0.198$, $T = 0.25$ K (right) as a function of longitudinal field. Both exhibit pronounced non-linear responses, but with different forms most likely due to random field effects [16] appearing for the more concentrated sample.

classical spin glass transition $T_g(H_t = 0)$ increases with increasing x , but lies below the mean-field ferromagnetic $T_C(x) = xT_C(x = 1)$. However, once the transverse magnetic field is turned on and the relative importance of quantum entanglement and random field effects becomes germane, the samples respond very differently and the phase boundaries actually cross.

Fig. 2 illustrates the pronounced sensitivity to fields applied parallel to the Ising axis and the evolution of the nonlinear response with x . $\chi(h_\ell)$ for $x = 0.167$ can be described by a conventional power series expansion: $\chi = \chi_1 - 3\chi_3 h_\ell^2 + 5\chi_5 h_\ell^4 + \dots$, with all orders of the susceptibility growing as the glass transition is approached from above. The longitudinal field dependence of the susceptibility for $x = 0.198$ also exhibits strong nonlinearities, but has qualitatively different behavior. The parabolic χ_3 at small h_ℓ rolls over to a linear dependence at large field, consistent with a tendency towards the singular linear behavior, attributed to random fields, seen for the disordered ferromagnet with $x = 0.44$ in the classical low H_t , high T regime [16]. Fig. 2 not only provides insight into the different underlying physics as one moves toward the ferromagnet with increasing x , but also underscores the well-known requirement that only the field range where χ is actually quadratic in field should be used to extract χ_3 .

We show in Fig. 3 the temperature dependence of the linear and nonlinear terms in the susceptibility for $x = 0.167$ at $H_t = 2.1$ kOe. As expected, increasing orders of the susceptibility diverge increasingly more strongly [23], reflecting the approach to a phase transition. This is in accord with the results reported by Wu et al. [4] and in disagreement with the recent results of Jönsson et al. [22]. The discrepancy can be understood by looking at the different limits in which the system was examined. In Ref [4] and in the present work, great care was taken to accumulate data in the $h_\ell \rightarrow 0$ limit.

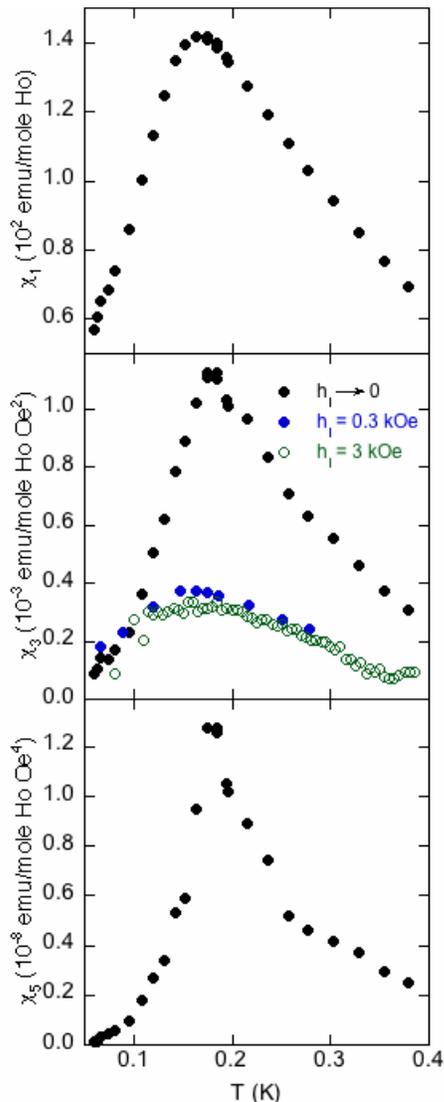


FIG. 3: (color online) Linear and nonlinear susceptibilities for $\text{LiHo}_{0.167}\text{Y}_{0.833}\text{F}_4$ derived from data akin to Fig. 2a and fit to $\chi = \chi_1 - 3\chi_3 h_\ell^2 + 5\chi_5 h_\ell^4 + \dots$. In (b) we demonstrate the danger of not explicitly accounting for high order, non-linear effects. Fitting our data to a simple parabola out to 0.3 kOe or applying large fields as in Ref. [22] (open circles) suppress the divergence of χ_3 and mask the true physics.

By contrast, the μSQUID technique of Ref [22] requires measuring hysteresis loops using large longitudinal fields (± 3 kOe, 15 times the scale of Fig. 2) at sweep rates of up to 50 Oe/s $\gg 2\pi A f$ for our experiment, resulting in a non-equilibrium, large-field estimate for χ_3 . We demonstrate this explicitly in Fig. 3b by fitting our $\chi(h_\ell)$ data to a parabola over the entire field range (± 0.3 kOe). Fitting to large fields without accounting for the full non-linear response effectively mimics the data of Jönsson et al., improperly eliminating the cusp in χ_3 and hiding the true physics.

We plot in Fig. 4 the spectroscopic response of the

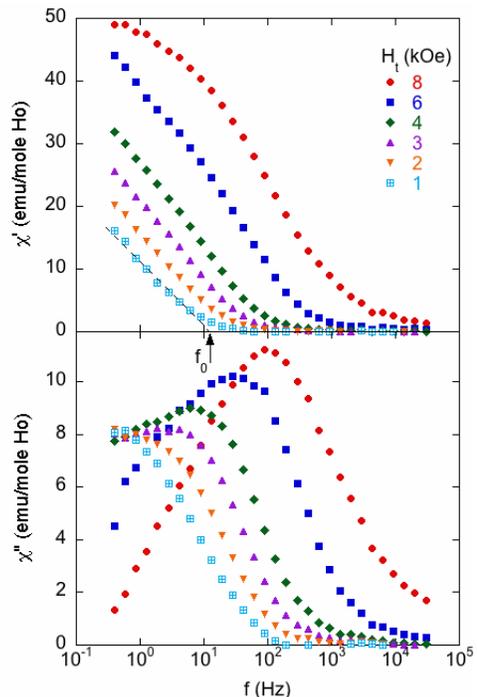


FIG. 4: (color online) Spectral response of the magnetic susceptibility of $\text{LiHo}_{0.198}\text{Y}_{0.802}\text{F}_4$ at $T = 0.05$ K for a series of transverse fields. The spin glass transition is marked by a flat low-frequency response in χ'_1 , corresponding to a logarithmic dependence of χ'_1 with onset f_0 and, via the fluctuation-dissipation theorem, $1/f$ noise in the magnetization.

$x = 0.198$ spin glass over 5 decades of f at a series of H_t . This sample is slower than its $x = 0.167$ counterpart [3], where quantum fluctuations promoted by the off-diagonal elements of the dipolar interaction more effectively speed the long-time relaxation. We characterize the approach to the spin glass from above by fitting the low frequency tail of χ'_1 to a power law form, f^α . $T_g(H_t)$ is defined dynamically when $\alpha \rightarrow 0$ and fluctuations occur on all (long) timescales. At this point χ'_1 grows logarithmically with f , where the onset frequency f_0 defines the fastest relaxation process available to the system and characterizes the quantum tunneling rate. The quantum dynamics accelerate with increasing H_t and quantitatively follow a WKB form [10].

We summarize our major findings in Fig. 5. Magnetic glass transitions for both $x = 0.167$ and 0.198 are defined classically (a–d) and quantum-mechanically (e–h) by $\alpha \rightarrow 0$. When it is possible to reach the $f \rightarrow 0$ limit at modest H_t , then a sharp, dynamical feature in χ''_1 and a divergence of χ_3 also serve to define $T_g(H_t)$. The onset frequency for relaxation, f_0 , follows an Arrhenius law, $e^{-\Delta/k_B T}$, in the classical limit where thermal fluctuations dominate and a WKB form in the quantum limit [10]. As a function of T with $H_t = 0$, f_0 for the two concentrations are indistinguishable. By contrast, at base temperature the H_t -dependent f_0 curves look differ-

ent for the two values of x , with faster relaxation at low H_t in the $x = 0.198$ sample with the suppressed quantum glass transition.

We have verified that in low transverse fields the dilute dipolar-coupled magnet, $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$, can display the static and dynamical signatures of a conventional spin glass. These results are in agreement both with early theory [20] and experiments of fifteen years ago, and in disagreement with incorrect (as shown above) conclusions drawn from interesting recent experiments [22] where the same material was subjected to very strong and rapid perturbations away from equilibrium. While the static signature of the spin glass transition - a diverging non-linear susceptibility - seems to disappear for high H_t , the dynamical signature - the appearance of a flat $\chi''(f \rightarrow 0)$ - persists and indeed becomes sharper. This suggests that internal random fields [6, 7, 16] notwithstanding, there is a distinct quantum glass state that can be entered via a first order transition [3, 19] for which χ_3 would not diverge. The striking new discovery that we make here is that the (quantum) critical field H_c for this state is a non-monotonic function of x , with a lower value for $x = 0.198$ than for both the $x = 0.167$ spin glass and the $x = 0.44$ ferromagnet. We suspect that for $x = 0.198$ the random field effects seen near the Curie point for the $x = 0.44$ sample are important, and suppress the magnetic glass phase. On the other hand, the glass-like state for $x = 0.167$ is more robust because of the larger quantum entanglement derived from the relatively greater population of antiferromagnetically coupled spins. As x is lowered even more, we land in the antighlass spin liquid phase. The quantum spin glass then acquires new meaning as a resonating valence bond state where there are multiple ways of drawing the bonds to construct pairs, while the spin liquid for $x = 0.045$ is a non-degenerate liquid with a unique pattern of valence bonds.

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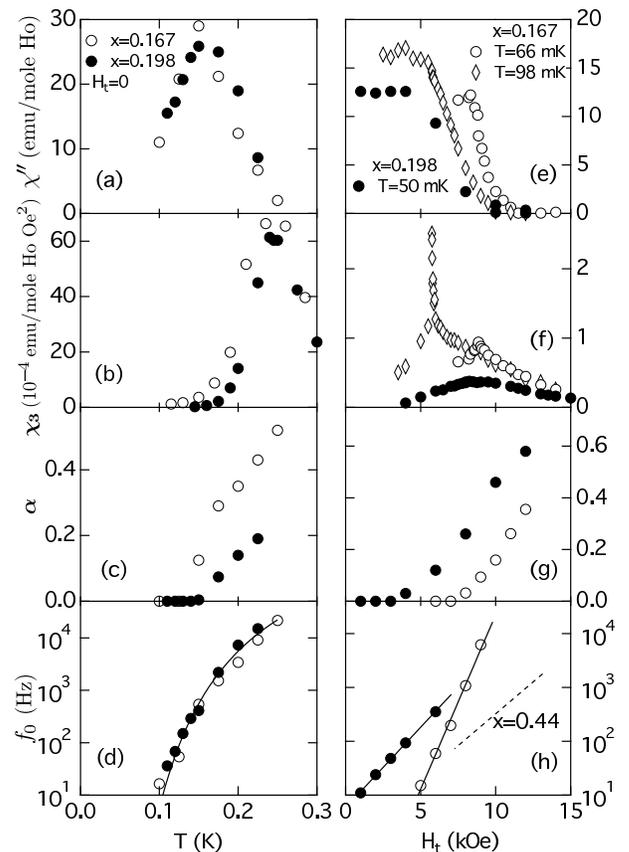


FIG. 5: Dynamical and non-linear signatures of the spin glass transition in the classical ($H_t = 0$, a-d) and quantum ($T \rightarrow 0$, e-h) regimes. The contrasting behavior of the two concentrations with transverse field reflect the competing effects of quantum entanglement at small x and random fields at large x (see text). Solid lines in d and h are Arrhenius and WKB fits, respectively. Dashed line is the slope for $x = 0.44$ [10].

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