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Quantum spin glass in anisotropic dipolar systems

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Abstract

The spin-glass phase in the $LiHo_xY_{1-x}F_4$ compound is considered. At zero transverse field this system is well described by the classical Ising model. At finite transverse field deviations from the transverse field quantum Ising model are significant, and one must take properly into account the hyperfine interactions, the off-diagonal terms in the dipolar interactions, and details of the full $J = 8$ spin Hamiltonian to obtain the correct physical picture. In particular, the system is not a spin glass at finite transverse fields and does not show quantum criticality.

1. Introduction

The study of spin glasses, and in particular the classical transition between the spin-glass (SG) phase and the paramagnetic (PM) phase, have been thoroughly studied since the 1970s [1]. As interest in quantum phase transitions has grown, the understanding of quantum criticality at the SG to PM phase transition has drawn much theoretical interest [2–4]. Experimentally, the quantum phase transition is not accessible in most spin glasses, where the strong exchange interaction blocks quantum fluctuations at available magnetic fields. However, anisotropic magnetic dipolar systems, notably the $LiHo_xY_{1-x}F_4$ compound, have very weak exchange interactions, and seem ideal for the observation of such a transition [5]. In this system the dipolar interaction and single-ion anisotropy terms have magnitude $\sim$1 and $\sim$10 K respectively; with spin $J = 8$, appreciable quantum fluctuations are expected already at transverse fields $H_\perp \sim 1$ T. The strong easy-axis anisotropy means that, for $T \ll 10$ K, the Ho spins truncate to an Ising-like doublet; this anisotropy also strongly suppresses all but the longitudinal terms of the dipolar interaction. The $LiHoF_4$ compound can be diluted by exchanging Ho with the nonmagnetic Y atom, resulting in the $LiHo_xY_{1-x}F_4$ compound with any desired x [5]. The angular dependence of the dipolar interaction, in the presence of quenched randomness, results in frustration. Thus, the system which is ferromagnetic at $x = 1$ [6], turns into a spin glass for $x = 0.167$, with $T_c = 0.13$ K [7, 8]. For the latter dilution, the SG to PM transition was studied

as a function of T and $H_\perp$ [7, 8]. Both the linear and nonlinear susceptibility were measured. Despite the importance of this experiment, some of its very interesting features long remained without a proper explanation. Questions included

(i) the reduction of the cusp in the nonlinear susceptibility with *decreasing* T;
(ii) the observation that it is much easier to disorder the spin glass thermally than quantum mechanically;
(iii) the sharpness of the low-T crossover between a PM response and slow relaxation; and
(iv) the smallness of the low-T critical exponent, as observed in the nonlinear susceptibility near the transition.

Theoretically, the $LiHo_xY_{1-x}F_4$ system was considered to be a good realization of the transverse field Ising model (TFIM) in the electronic degrees of freedom. Recently [9–11] it was shown that the real system differs from the above model in two significant ways, which affect the physics considerably; (a) the hyperfine (hf) interaction between the Ho electronic and nuclear spins is strong, and for $x \ll 1$ in general, and in the SG experiments [8] in particular, dominate the physics [9, 11], (b) the off-diagonal dipolar terms, although effectively reduced, become essential at any finite $H_\perp$ as they reduce the symmetry of the model [10]. In this paper we show how these features answer the first two of the four questions noted above; we also show that a proper understanding of the off-diagonal dipolar terms requires going outside the simple TFIM.

2. Hyperfine interactions

The Hamiltonian describing the $LiHo_xY_{1-x}F_4$ system is given by a sum of crystal field [12, 13], Zeeman, hf, and inter-Ho interaction terms:

$$H = H_{\rm cf} + H_{\rm Z} + H_{\rm hf} + H_{\rm int}. \tag{1}$$

The Ho ion has a $J = 8$ angular momentum. $H_{\rm cf}$ splits the 17-fold degeneracy, leaving three relevant low-energy levels: an Ising-like doublet, denoted $|\uparrow\rangle, |\downarrow\rangle$, and a first excited state approximately 10 K higher in energy, denoted $|\Gamma_2^l\rangle$ [9, 12]. $H_{\rm Z} = -\sum_i g_J \mu_{\rm B} \vec{H} \cdot \vec{J}_i$ is the Zeeman energy, and $H_{\rm int} = -\sum_{ij} U_{ij}^{\alpha\beta} J_i^\alpha J_j^\beta$ is dominated by the dipolar interaction [13]. We denote the easy axes by z, and consider $H_\perp \parallel x$. It is common to neglect all but the longitudinal terms in the dipolar interaction, and drop the hf interaction, thus obtaining a low-energy TFIM effective Hamiltonian:

$$H = -\sum_{i,j} V_{ij}^{zz} \tau_i^z \tau_j^z - \Delta_0(H_\perp) \sum_i \tau_i^x, \tag{2}$$

where $\vec{\tau}_j$ is a Pauli vector describing the two-level effective electronic spin at spatial position $\mathbf{r} = \mathbf{r}_j$, and $\Delta_0 \propto {H_\perp}^2/\Omega_0$ for small $H_\perp$. However, both the hf and off-diagonal dipolar interactions are of crucial importance for the $LiHo_xY_{1-x}F_4$ system.

The Ho atom is a pure isotope $I = 7/2$ nuclear spin with contact hf interaction $H_{\rm hf} = A_J \sum_i \vec{I}_i \cdot \vec{J}_i$. Due to the strong anisotropy we consider first the longitudinal part of the hf interaction $H_{\rm hf}^{\parallel} = A_J I^z J^z$. This term splits each of the states $|\uparrow\rangle, |\downarrow\rangle$ into an eightfold multiplet of nearly equidistant levels, with separation ∼205 mK [12] between adjacent levels (figure 1). This splitting, larger than the typical dipolar energy and the relevant experimental temperatures [8], influences significantly the physics of the system. The Ising doublet states have now a definite nuclear spin, i.e. $I_z = -7/2$ for the electronic state $|\uparrow\rangle$ and $I_z = 7/2$ for the electronic state $|\downarrow\rangle$. A transverse field $H_\perp$ then couples $a \equiv |\uparrow, -7/2\rangle$ with $b \equiv |\downarrow, -7/2\rangle$ and $\bar{a} \equiv |\downarrow, 7/2\rangle$ with $\bar{b} \equiv |\uparrow, 7/2\rangle$ (see figure 1). Thus, the transverse field by itself does not

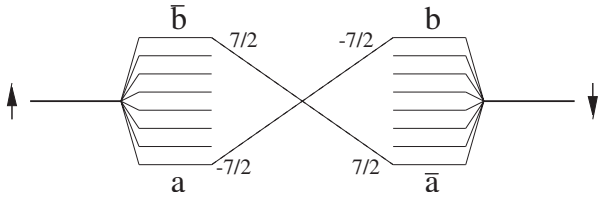

Figure 1. Splitting of the electronic low-energy doublet (↑ and ↓) due to the longitudinal hf interaction. The doublet ground states, a and $\bar{a}$ have a definite and opposite nuclear spin, $\pm 7/2$. Transverse magnetic field couples states with the same nuclear spin, as is shown by the dashed lines.

induce quantum fluctuations between the relevant Ising doublet ground states (GSs), but only re-normalizes their effective spin. The *transverse* hf interactions $H_{\rm hf}^{\perp} = A_J(I^+J^- + I^-J^+)/2$ allow simultaneous changes in I_z and J_z, and thence quantum fluctuations between the Ising doublet states. However, as is detailed in [9, 11], this mechanism is negligible for $H_\perp \ll \Omega_0/\mu_B$. Thus, the inclusion of the hf interactions results in three energy scales which dictate the behaviour of the system, and in particular the position of the crossover between the SG and PM phases as a function of T and $H_\perp$ [9, 11]. At zero field, $T_c(0)$ is dictated by the dipolar interaction V_0, since the nature of the Ising doublet is not important for the classical transition. The behaviour of $T_c(H_\perp)$ with $H_\perp$ is governed by the strength of the hf interaction and the crossover at $T = 0$ is governed by the anisotropy energy Ω_0, as quantum fluctuations between the relevant Ising states become significant only when the state $|\Gamma_2^l\rangle$ becomes appreciably hybridized with $|\uparrow\rangle, |\downarrow\rangle$. All three energy scales become apparent in the position of the line separating the SG and PM phases, and in particular the relation $V_0 \ll \Omega_0$ explains the fact that it is much easier to disorder the spin glass thermally rather than quantum mechanically [9, 11].

3. Off-diagonal terms of the dipolar interaction

The longitudinal term of the dipolar interactions, $\propto J_i^z J_j^z$, has direct matrix elements within the low-energy Ising states $|\uparrow, -7/2\rangle, |\downarrow, 7/2\rangle$. All other interaction terms involve the state $|\Gamma_2^l\rangle$ in second or higher perturbation expansion. Since $\Omega_0 \gg V_0$, one is tempted to neglect all but the longitudinal interaction. However, the off-diagonal terms, and in particular the terms $\propto J_i^z J_j^x$, become important at $H_\perp \neq 0$, as they change the symmetry of the system. The $J_z \to -J_z$ symmetry, while maintained by $H_\perp$ without the off-diagonal terms, is destroyed. This reduction of symmetry results in a generation of an effective random field at each site [10]. As a result, within the droplet picture of Fisher and Huse [14], and using an Imry–Ma-like argument [15], one obtains [10] a magnetic field-dependent finite correlation length. The system with *transverse* magnetic field and off-diagonal dipolar interactions becomes equivalent to the random field Ising model, i.e., a spin glass in the presence of random *longitudinal* magnetic field. Thus, the scaling theory of Fisher and Huse [14] predicts an instability of the SG phase to finite $H_\perp$ in our case, equivalent to its prediction of the absence of a de-Almeida–Thouless line in the RFIM. Interestingly, to obtain the correct physical picture of the system at finite field one has to consider the large Ho spin, going beyond the simplified Ising picture. The reason is that it is the fluctuations between each of the single Ho GSs and its first excited states, and not the much smaller quantum fluctuations between the two Ising states $a, \bar{a}$ [10], that govern the magnitude of the effective random field and the reduction of the correlation length.

Within the scaling picture of Fisher and Huse [14] the spin glass at zero transverse field has two time-reversed GSs, denoted ψ and $\bar{\psi}$. Each Ho ion in the GS ψ is in either state a or $\bar{a}$,

and in the opposite state in the GS $\bar{\psi}$. Consider a single Ho ion at $H_\perp \neq 0$. For small magnetic fields the fluctuations between the Ising states a and $\bar{a}$ are negligible [9]. Yet, the energy of each of the Ising states is reduced by an energy proportional to ${H_\perp}^2/\Omega_0$, due to fluctuations to the relevant excited states ($|\Gamma_2^l, -7/2\rangle$ for a and $|\Gamma_2^l, 7/2\rangle$ for $\bar{a}$). If we choose an arbitrary region in state ψ, the energy reduction due to the field is just the sum over all spins of the single spin energy gain. With $J_z \to -J_z$ symmetry, the same energy reduction occurs for the state $\bar{\psi}$. Now consider the effect of the off-diagonal dipolar terms (in particular, the term $V_{ij}^{zx} J_i^z J_j^x$) on the different domains. In second-order perturbation, the domain energy shift is given by [10]

$$E_\psi^{(2)} = -\frac{\langle\psi_0|(\sum_{i\neq j} V_{ij}^{zx} J_i^z J_j^x + \mu_B H_\perp \sum_i J_i^x)^2|\psi_0\rangle}{\Omega_0}. \tag{3}$$

The dipolar terms have randomly the same or the opposite sign to that of the magnetic field, and typically, by flipping a domain of N spins one gains an energy of [10]

$$\langle\delta E\rangle = c\frac{j^2\mu_B H_\perp V_0\sqrt{N}}{\Omega_0}, \tag{4}$$

where $j = \max J_z$. Comparing this energy gain to the energy cost of flipping the domain [10], $\approx j^2 V_0 L^{\theta_d}$, where L is the domain linear size, one finds a finite correlation length at any $H_\perp$ given by [10]

$$\xi \approx \left(\frac{\Omega_0}{\mu_B H_\perp}\right)^{\frac{1}{(3/2)-\theta_d}}. \tag{5}$$

Importantly, this correlation length depends only on $H_\perp$ and Ω_0. In the experiment [8], as T is decreased the crossover to the PM phase occurs at higher $H_\perp$, dictating a smaller correlation length and a reduced cusp in the nonlinear susceptibility [10]. Note that the cusp is further reduced due to the renormalization of the effective spin [9]. Interestingly, the finite correlation length results in an enhanced transverse field in the x direction [10], which was anticipated by the comparison of the experimental and theoretical positions of the crossover line between the SG and PM phases [9]. Importantly, our results here are easily generalized to any anisotropic spin glass, as long as a dipolar interaction exists [10].

In principle, one could also get a finite correlation length in a spin-half Ising model, by introducing a longitudinal interaction $\propto V_0$ and a reduced off-diagonal interaction $\propto \alpha V_0$ [16]. Similar considerations to the ones above lead to a correlation length $\xi \approx [V_0/(\alpha\mu_B H_\perp)]^{1/(3/2-\theta_d)}$ [16], coming from quantum fluctuations between the Ising doublet states, with a ξ depending on V_0. A notable difference between this result and the correct one is the size ξ at the crossover to the PM phase at $T = 0$. This crossover actually occurs at $H_\perp \approx \Omega_0$ for the case of large spin [9, 10], leading to $\xi \approx 1$. However, in the effective spin-half model the crossover occurs at $H_\perp \approx V_0$ and $\xi \approx 1/\alpha$ at the crossover, emphasizing the inadequacy of the spin-$\frac{1}{2}$ model. Recently, Tabei *et al* considered the anisotropic dipolar spin glass in transverse field [17]. They used a spin-half model with effective random fields in both longitudinal and transverse directions, and infinite range interaction. They find a diminishing of the cusp of the nonlinear susceptibility with reduction of temperature and increase of applied transverse field. However, neither the experimental temperature dependence of the cusp's peak nor its smearing as the temperature is reduced, are reproduced in their results.

4. Numerical results

The scaling relation (4) has been checked against Lanczos exact diagonalization (ED) computations on finite size clusters [10]. In order to get closer to the experiment, we randomly

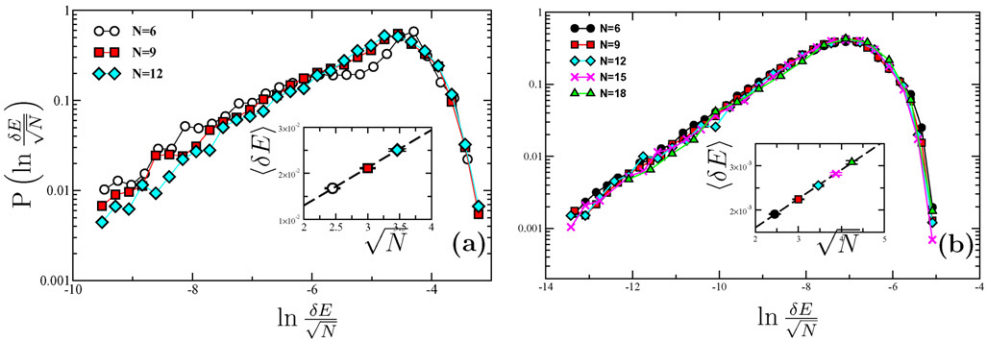

Figure 2. Distributions of the finite size gaps δE rescaled by $\sqrt{N}$ and plotted in a semi-log scale. Lanczos ED data collected over 10 000 random diluted $LiHo_xY_{1-x}F_4$ samples for each size N. Insets: Linear dependence of the disorder average gap $\langle \delta E \rangle$ versus $\sqrt{N}$. (a) Results obtained for the spin-1 Hamiltonian (6) with $\Omega_0/\mu_B H_t = 100$ for three different sizes, with $x = 18.75\%$. (b) Results obtained for the spin-$\frac{1}{2}$ model (7) with $\alpha = 0.1$, $\mu_B H_t = 0.0025$ and five different sizes with a dilution $x = 1/12$.

(This figure is in colour only in the electronic version)

distribute N moments at the rare earth sites of three-dimensional $LiHo_xY_{1-x}F_4$ diluted lattices, and focus on $s = 1$ particles with an on-site anisotropy $\Omega_0 \simeq 10$ K which accounts for the crystal field. Therefore, the following transverse field dipolar spin-1 Hamiltonian,

$$\mathcal{H}_1 = -\sum_{i\neq j}\left[\tfrac{1}{2}V_{ij}^{zz}S_i^zS_j^z + V_{ij}^{zx}S_i^zS_j^x\right] - \mu_B H_t\sum_i S_i^x - \Omega_0\sum_i\left([S_i^z]^2 - s^2\right), \tag{6}$$

has been diagonalized on $LiHo_xY_{1-x}F_4$ lattices with $x = 18.75\%$ for various sizes, and over 10 000 independent random samples for each size. In the perturbative regime, we have computed the finite size gap δE for each sample and the $\sqrt{N}$ scaling stated in equation (4) is clearly demonstrated, as shown in figure 2(a).

A similar calculation was done for the spin-$\frac{1}{2}$ Ising model [16]:

$$\mathcal{H}_{\frac{1}{2}} = -\sum_{i\neq j}\left[\tfrac{1}{2}V_{ij}^{zz}S_i^zS_j^z + \alpha V_{ij}^{xz}S_i^xS_j^z\right] - \mu_B H_t\sum_i S_i^x. \tag{7}$$

Numerically speaking, the $s = 1/2$ problem is easier since it leads to a smaller Hilbert space dimension and allows us to check the $\sqrt{N}$ scaling relation over a broader range of sample sizes, as shown in figure 2(b). However, here too the essential difference between the spin-$\frac{1}{2}$ and spin-1 models is apparent. In the spin-$\frac{1}{2}$ model, since the significant fluctuations are between the Ising doublet states, the strength of the dipolar interaction replaces Ω_0 in the denominator of equation (3), which results in better convergence of the numerics at the smallest sizes.

5. Conclusion

Of the four experimental puzzles mentioned in the introduction, the first two are explained by our analysis. Since we believe that the Hamiltonian (1) includes all the physics relevant to the experiment [8], we expect that the solution to the latter two puzzles lies within the same framework of considerations above.

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