Quantum Spin Glass and the Dipolar Interaction

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Anisotropic dipolar systems are considered. Such systems in an external magnetic field are expected to be a good experimental realization of the transverse field Ising model. With random interactions, this model yields a spin glass to paramagnet phase transition as a function of the transverse field. We show that the off-diagonal dipolar interaction, although effectively reduced, induces a finite correlation length and thus destroys the spin-glass order at any finite transverse field. We thus explain the behavior of the nonlinear susceptibility in the experiments on $LiHo_xY_{1-x}F_4$, and argue that a crossover to the paramagnetic phase, and not quantum criticality, is observed.

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The study of quantum phase transitions (QPT) is of prime recent interest, as it is believed that the understanding of the physics at the vicinity of quantum critical points will shed light on some of the most interesting problems in condensed matter physics, such as the metal insulator transition, superconductor insulator transition, and high temperature superconductivity. Quantum magnets, and, specifically, their modeling by the transverse field Ising model (TFIM)

$$H = -\sum_{i,j} J_{ij}\tau_i^z\tau_j^z - \Delta\sum_i \tau_i^x, \qquad (1)$$

are a particularly good laboratory to study QPT. This model is rich enough to capture the interesting physics of QPT yet simple enough to allow theoretical treatment. Experimentally, much effort was invested to realize the TFIM. Maybe the best realization is in anisotropic dipolar systems, where the dipolar energy dominates the spin-spin interaction and the crystal field (CF) generates strong anisotropy. This results in a ground state (GS) Ising-like doublet for the single spins and an effective reduction of all but the longitudinal interaction terms.

Indeed, $LiHo_xY_{1-x}F_4$, with $x = 1$, was shown [1] to exhibit a ferromagnetic to paramagnetic (PM) transition as a function of transverse field H_t and temperature T. As x is reduced, the randomness in the position of the magnetic Ho atoms results in frustration, and for $x = 0.167$ a spin-glass (SG) phase was observed [2,3]. Furthermore, applying H_t induces quantum fluctuations, leading to a PM phase at large fields. Thus, this compound is considered to be the archetypal experimental realization of a quantum SG [4,5]. In this Letter, we show that, for anisotropic dipolar glasses in general, and for the $LiHo_xY_{1-x}F_4$ compound in particular, the off-diagonal dipolar (ODD) interaction terms, albeit effectively reduced, qualitatively change the physics of the problem. In particular, in the presence of H_t, the ODD terms reduce the symmetry of the system in comparison to the TFIM and render the latter inadequate in studying the system. A proper treatment of the ODD terms results in the absence of long-range SG order at any finite H_t and a reduction of the cusp in the nonlinear susceptibility (NLS) at the crossover (C-O) to the PM phase. Thus, we argue that the experimental line drawn at the peak values of the NLS [3] is not a phase transition line. Except for the point at $H_t = 0$, this line corresponds to a C-O between a paramagnet to a phase we denote a "quasi-spin glass." In this phase, the system separates into domains within which the random ordering of the spins is maintained. These domains have a typical size $\xi(H_t)$ which dictates the correlation length in the system, and its dependence on H_t is given by the critical exponent ν calculated below. The domain structure is maintained until the C-O field, where fluctuations between the relevant Ising-like states dominate and the system becomes PM (see Fig. 1). This C-O is expressed as a cusp in the NLS. Importantly, the reduction of ξ with increasing H_t results in the corresponding reduction of the cusp in the NLS, explaining the peculiar experimental result [3] where the cusp is reduced with decreasing T. Interestingly, we show below that at $T = 0$ the C-O takes place at a value of H_t which corresponds to $\xi \approx 1$ and, therefore, to a complete

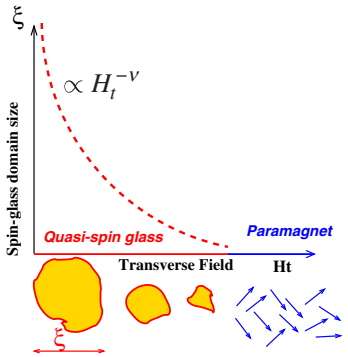

FIG. 1 (color online). Schematic picture for the $T = 0$ behavior of a dipolar Ising SG in a transverse field H_t. The typical size ξ of a SG ordered domain (depicted below the x axis) decreases with H_t, with a critical exponent ν (see text). At large enough H_t, the system becomes PM, via a C-O and not a quantum phase transition.

absence of a cusp in the NLS, as can be inferred from the experiment [3].

Theoretical considerations.—Our analysis is valid both specifically to the $LiHo_xY_{1-x}F_4$ system, as we further comment on below, as well as to any anisotropic dipolar system. The only requirement is that the single spins have a GS Ising-like doublet, with a large energy separation to the excited states. To emphasize the generality of our approach, we consider the following spin-s Hamiltonian

$$\mathcal{H} = -D\sum_i[(S_i^z)^2 - s^2] - \frac{1}{2}\sum_{\substack{i\neq j\\ \alpha,\beta}} V_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta - \mu_B H_t \sum_i S_i^x. \tag{2}$$

Here i and j denote the positions of the spins, randomly diluted on some lattice, $V_{ij}^{\alpha\beta}$ denotes the dipolar interaction ($\alpha, \beta = x, y, z$), and $D > 0$ is the CF anisotropy constant. For $H_t = 0$, the GS of a single spin is doubly degenerate, denoted $|\uparrow_s\rangle$ and $|\downarrow_s\rangle$, with $s_z = \pm s$. The first excited states have $s_z = \pm(s-1)$ and energy $\Omega_0 = (2s-1)D$. Throughout the Letter, we assume that $\Omega_0 \gg \mu_B H_t, V_{\max}$, where $V_{\max}$ is the largest dipolar energy between two spins in the system. We define $\mathcal{H} = \mathcal{H}_\parallel + \mathcal{H}_\perp$ such that

$$\mathcal{H}_\parallel = -D\sum_i[(S_i^z)^2 - s^2] - \frac{1}{2}\sum_{i\neq j} V_{ij}^{zz} S_i^z S_j^z; \tag{3}$$

$$\mathcal{H}_\perp = -\frac{1}{2}\sum_{i\neq j}\sum_{(\alpha\beta)\neq(zz)} V_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta - \mu_B H_t \sum_i S_i^x. \tag{4}$$

We assume that the dilution is such that $\mathcal{H}_\parallel$ is equivalent to the classical random bond Ising model with a low T SG phase. As this classical dipolar Ising SG is equivalent to the short-range Edwards-Anderson model [6,7] [Eq. (1) with random nearest neighbor J_{ij} and $\Delta = 0$ [8]], our analysis is done within the scaling ("droplet") picture [9], which accounts for its behavior at large sizes. The GS of $\mathcal{H}_\parallel$ is then twofold degenerate [9] with states $|\psi_o\rangle, |\bar{\psi}_o\rangle$, which are related by $S_z \to -S_z$ symmetry, and in which each spin is in either state $|\uparrow_s\rangle$ or $|\downarrow_s\rangle$. Importantly, adding a transverse field term preserves the above symmetry, and therefore the TFIM is the archetypal model for the quantum SG phase. However, when adding $\mathcal{H}_\perp$ which includes ODD terms, this symmetry is not preserved. The GS degeneracy breaks, and the system gains energy by choosing locally a state similar to $|\psi_o\rangle$ or $|\bar{\psi}_o\rangle$ according to which optimizes the energy gain due to $\mathcal{H}_\perp$.

Following the scaling picture of Fisher and Huse [9] and using an Imry-Ma-like [10] argument, we calculate this energy gain, i.e., the energy to flip a droplet of size L having $N \sim L^d$ spins, due to the addition of $\mathcal{H}_\perp$. This energy gain [see Eq. (9)] is then compared with the energy cost due to the domain wall formation, and the correlation length ξ is obtained [Eq. (11)]. Consider first

$$\mathcal{H}'_\perp = -\sum_{i\neq j} V_{ij}^{zx} S_i^z S_j^x - \mu_B H_t \sum_i S_i^x. \tag{5}$$

The addition of $\mathcal{H}'_\perp$ to $\mathcal{H}_\parallel$ changes $|\psi_o\rangle \to |\psi\rangle$ and $|\bar{\psi}_o\rangle \to |\psi'\rangle$ with energies E_ψ and $E_{\psi'}$, respectively. The energy the system gains by choosing locally the lowest energy state is $\delta E \equiv |E_\psi - E_{\psi'}|$, which we now calculate. In second order perturbation, $E_\psi = E_{\psi_o} + E_\psi^{(2)}$, where

$$E_\psi^{(2)} = -\frac{\langle\psi_o|(\sum_{i\neq j} V_{ij}^{zx} S_i^z S_j^x + \mu_B H_t \sum_i S_i^x)^2|\psi_o\rangle}{\Omega_0}. \tag{6}$$

Here we used the fact that the only relevant excited states are those in which one spin changes its state from $s_z = \pm s$ to $s_z = \pm(s-1)$, and their energy is Ω_0 in leading order. Thus, the sum over the excited states can be taken out as the identity operator. A similar equation holds for $E_{\psi'}$. One can show that the terms with even powers of H_t are equal for E_ψ and $E_{\psi'}$, while the term linear in H_t is equal in magnitude but has opposite signs for E_ψ and $E_{\psi'}$ [7]. Using the fact that, since $k \neq l$, the operators commute, we obtain

$$\delta E = \frac{4}{\Omega_0}\langle\psi_o|\mu_B H_t \sum_i S_i^x \sum_{k\neq l} V_{kl}^{zx} S_k^z S_l^x|\psi_o\rangle \tag{7}$$

and therefore

$$\delta E = 4\frac{s\mu_B H_t}{2\Omega_0}\sum_{k\neq i} V_{ki}^{zx}\langle\psi_o|S_k^z|\psi_o\rangle = \frac{2s\mu_B H_t}{\Omega_0}\sum_i h_i^x, \tag{8}$$

where we define $h_i^x \equiv \sum_k V_{ki}^{zx}\langle S_k^z\rangle$ as an effective transverse magnetic field at site i. For each i, all of the V_{ki}'s are small except the few for which the sites i and k are spatially close. Because of the randomness of the sign, retaining for each i the term with the largest absolute value, denoted $\tilde{V}_i$, gives a good approximation for δE up to a numerical factor c of order unity. Since $\tilde{V}_i$ is random in sign, the average energy gained by flipping a droplet of N spins is given by

$$\langle\delta E\rangle = c\frac{s^2\mu_B H_t V\sqrt{N}}{\Omega_0}, \tag{9}$$

where V is the average magnitude of $|\tilde{V}_i|$, and we choose $|\psi_o\rangle$ and $|\bar{\psi}_o\rangle$ such that $\delta E > 0$.

The above result (9) is central to our analysis, and, in order to check our approximation of randomness leading to it, we calculated the gap between the GS and the first excited state numerically using Lanczos exact diagonalization (ED) [11]. We consider system sizes in the regime where they are much smaller than ξ. This is important for our calculation, since then we are dealing with single domains, and, therefore, the two lowest states correspond to $|\psi\rangle$ and $|\psi'\rangle$ and the gap to δE. To reproduce the experimental situation, we focus on three-dimensional finite size clusters, randomly distributing N spins at the rare earth sites of the $LiHo_xY_{1-x}F_4$ lattice. Since we are interested in small fields, it is sufficient to use $s = 1$ particles with on-site anisotropy Ω_0. We therefore study the spin-1 version of $\mathcal{H} = \mathcal{H}_\parallel + \mathcal{H}'_\perp$:

$$\mathcal{H}_1 = -\sum_{i\neq j}\left[\frac{1}{2}V_{ij}^{zz}S_i^zS_j^z + V_{ij}^{zx}S_i^zS_j^x\right] - \mu_B H_t\sum_i S_i^x - \Omega_0\sum_i([S_i^z]^2 - s^2). \quad (10)$$

Here and below, all energies are in units of the typical nearest neighbor dipolar energy V_0. We fixed the dilution to $x = 18.75\%$ by using $2\times 2\times N/3$ unit cells, N being the total number of spins (there are 4 rare earth sites per unit cell). For this dilution, we find that $V = 0.8$. Lanczos ED has been performed in the *full* $d = [2s+1]^N$-dimensional Hilbert space for $N = 6$, 9, and 12 $s = 1$ spins [12]. For each size, the gap has been computed over 10^4 random samples. In Fig. 2, we present the numerical results obtained in the perturbative regime [$\Omega_0 \gg \mu_B H_t$ and $(\mu_B H_t)^2/\Omega_0 \ll V_{\max}$]. The $\sqrt{N}$ scaling of δE as stated in Eq. (9) is clearly demonstrated, as we found a very good data collapse for the distribution of $\ln(\delta E/\sqrt{N})$. The inset in Fig. 2 also shows that the disorder average gap $\langle\delta E\rangle = \alpha\sqrt{N}$. Confronting the numerical estimate obtained for the prefactor α with Eq. (9), we get $c \simeq 1$.

In order to obtain ξ, i.e., the typical domain size, we compare the domain's energy gain (9) to the energy cost due to the formation of a domain wall. For the short-range Ising SG, this energy is $\propto L^\theta$, with $\theta \approx 0.2$ in 3 dimensions [13,14]. Furthermore, under quite general conditions, Fisher and Huse argued [9] that $\theta \leq (d-1)/2$. For the dipolar Ising SG, we expect the same scaling behavior with a similar exponent $\theta_d \simeq \theta$ to hold [6,7,9], and the energy of flipping a domain is therefore $\approx Vs^2L^{\theta_d}$. As a result, for L such that $(s^2\mu_B H_t V\sqrt{N})/\Omega_0 > Vs^2L^{\theta_d}$, it will be preferable for domains to choose their state between $|\psi\rangle$ and $|\psi'\rangle$ as the one that locally minimizes $E_\psi^{(2)}$. This results in a finite ξ;

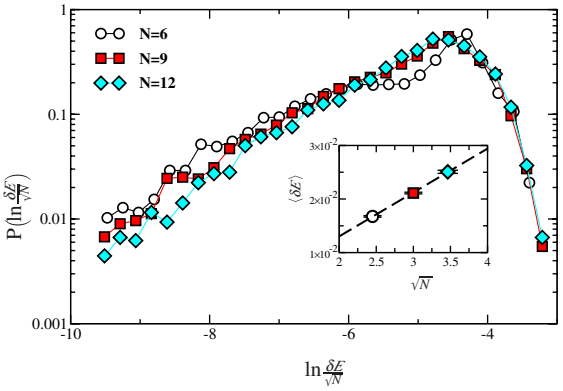

FIG. 2 (color online). Distribution $P[\ln(\delta E/\sqrt{N})]$ plotted in a semilog scale. Lanczos ED data obtained for the spin-1 Hamiltonian (10) with $\Omega_0 = 50$ and $\mu_B H_t = 0.5$ have been collected over 10^4 random samples with the $LiHo_xY_{1-x}F_4$ structure and $x = 18.75\%$, for $N = 6$, 9, and 12. Inset: The disorder average gap $\langle\delta E\rangle$ vs $\sqrt{N}$. The dashed line is a linear fit $\langle\delta E\rangle = \alpha\sqrt{N}$, with $\alpha \simeq 0.008$.

$$\xi \approx \left(\frac{\Omega_0}{\mu_B H_t}\right)^{1/(3/2-\theta_d)}. \quad (11)$$

For the Ising SG in longitudinal field, it was argued [9,15] and then verified experimentally [16] and numerically [17] that there is no de Almeida-Thouless line [18] and no SG phase at any finite field. At $H_t \ll \Omega_0/\mu_B$, our system is equivalent to the above model in small longitudinal fields, and we thus argue that there is no SG phase at any finite *transverse* field when the interaction is dipolar, and as $H_t \to 0$, ξ diverges with the same form [9] of the critical exponent $\nu = 1/[(3/2) - \theta_d]$.

In our treatment, the only dipolar terms we considered are the longitudinal and the zx terms. However, one can show that all the neglected terms [see the terms present in $\mathcal{H}_\perp$—Eq. (4)—but not in $\mathcal{H}'_\perp$—Eq. (5)] do not contribute to δE in the second order perturbation expansion [7].

Interestingly, the two effects of H_t, i.e., inducing the C-O to the PM phase and the reduction in ξ calculated above, behave very differently as functions of H_t. The former is dictated by fluctuations between the two single spin Ising GSs, which depend on H_t to a high power, of order s, and are practically negligible as long as $H_t \ll \Omega_0/\mu_B$. However, the fluctuations that dictate the reduction of ξ at low transverse fields are between each single spin GS and its first excited state at energy Ω_0. The latter depend on H_t to second order and result in a reduction of ξ which depends on $1/H_t$ to a power ν close to unity. Therefore, the disordering of the SG order by H_t occurs in two stages. At low field, domains of size ξ are formed, within which the GS is very similar to either of the two zero field SG GSs. At $H_t \approx \Omega_0/\mu_B$, a C-O occurs where the order within each domain is destroyed, and each spin fluctuates independently. Importantly, when reaching the C-O region at very low T, one is already in the regime where $\xi \approx 1$ in units of interspin spacing, resulting in small features in the relevant susceptibilities, in agreement with experiment [2,3]. We emphasize that the understanding of the scenario above *requires* a model in which the large spins are considered and the anisotropy is explicitly taken into account. Indeed, the anisotropy energy Ω_0 enters explicitly into Eqs. (9) and (11). A presumably simpler spin-half model with effectively reduced ODD terms will not be sufficient [7], since in such a model the reduction in ξ and the C-O to the PM phase are both induced by fluctuations between the Ising GSs and have the same scale in H_t.

In addition to changing the symmetry of the system at $H_t \neq 0$, resulting in the destruction of the SG phase and the QPT to the PM phase, the ODD terms also enhance the effective transverse field [19]. Although h_i^x [see Eq. (8)] is a random quantity, we have shown that domains of size ξ choose to be in a state equivalent to $|\psi\rangle$ or $|\psi'\rangle$ by the maximization of $\sum_i h_i^x$. As a result, a net magnetic field in the x direction $\langle h_i^x\rangle = \langle\delta E\rangle/N \propto \xi^{-3/2}$ is added to the external one. As the C-O is approached, ξ is small and the effective transverse field due to the ODD interaction is

significant. We thus give a precise physical origin to the conjecture made in Ref. [19].

Our analysis above could equally be done by defining δE in Eq. (8) as $\sum_k h_k^z \langle S_k^z \rangle$, where $h_k^z \equiv (2s\mu_B H_t/\Omega_0)\sum_i V_{ki}^{zx}$. Using this definition, one can make the analogy between the current problem to the Ising SG in random longitudinal field, as an alternative to the direct calculation of δE performed above.

Experimental consequences.—The CF Hamiltonian in $LiHo_xY_{1-x}F_4$ is different from the one given in Eq. (2). Furthermore, the hyperfine interactions strongly renormalize the parameters of the TFIM, invalidating the simple model in the electronic degrees of freedom [19]. Still, for our purpose here, an equivalent physical picture emerges: The two relevant (electronuclear) Ising states of each Ho ion couple very weakly at small H_t, and their corresponding excited states are at ≈ 10 K above the GSs. Thus, the requirements for the validity of our theory given before Eq. (2) are fulfilled. Our analysis and results [and, in particular, Eq. (11)] are therefore directly applicable to the SG experiments in the $LiHo_xY_{1-x}F_4$ system [2,3], with $\Omega_0 \approx 10$ K, and suggest that $LiHo_xY_{1-x}F_4$ is not a SG at any $H_t \neq 0$. Furthermore, the peculiar experimental result [3] where the cusp in the NLS is reduced with decreasing T is naturally explained: As T is reduced, the C-O to the PM phase occurs at larger H_t. This results in smaller ξ and, therefore, a diminishing of the cusp in the NLS. In addition, the renormalization of the effective spin [19] specific to the $LiHo_xY_{1-x}F_4$ compound further reduces the NLS near the C-O.

From the experimental point of view, our analysis changes the status of the field. The only claim for the observation of the QPT between the SG and PM phases was made in Ref. [3]. As we show that long-range SG order is unstable to applied H_t, and, therefore, a phase transition was not observed at low temperatures in the above experiment, an experimental observation of this QPT is still awaiting. Our analysis also points to the direction one should take in seeking such a QPT: systems in which SG order and quantum fluctuations compete, and either or both are controllable by a parameter which does not change the symmetry responsible for the GS degeneracy of the ordered state. An example would be the change, with applied pressure, of CF terms which induce quantum fluctuations between the Ising-like doublet [such as $(S_+^2 + S_-^2)$ terms added to the Hamiltonian (2) for integer spin systems].

Recently, there is increasing experimental [16] and numerical [17] support for the validity of the droplet picture in describing short-range Ising SG, in general, and to its prediction [9,15] of the nonexistence of a de Almeida-Thouless line [18], in particular. For the anisotropic dipolar systems discussed here, the C-O to the PM phase at $H_t \approx \Omega_0/\mu_B$ is a result of quantum fluctuations, and there is no analog to the de Almeida-Thouless line. However, at $H_t \ll \Omega_0/\mu_B$, the system is equivalent to a classical Ising SG in a small random longitudinal field. Thus, the above numerical and experimental results [16,17] support the validity of the droplet picture for dipolar Ising systems in small H_t as well. Still, we believe that experiments that would directly observe whether dipolar Ising glasses, in general, and $LiHo_xY_{1-x}F_4$, in particular, have a SG phase at a finite H_t are of much interest, both as a verification of our results and as additional support for the droplet picture in general.

Finally, our analysis is also applicable to any Ising SG where dipolar interactions are present, even if the interaction that governs the ordering is different. The correlation length is then given by [7] $\xi_J \approx (\Omega_0 J/\mu_B H_t V)^{1/(3/2-\theta)}$, where J is the dominant interaction. The qualitative picture is similar, only the size of the domains at the quantum C-O to the PM phase is $\approx (\frac{J}{V})^{1/(3/2-\theta)}$.

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