Quantum spin glass and the dipolar interaction

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Systems in which the dipolar energy dominates the magnetic interaction, and the crystal field generates strong anisotropy favoring the longitudinal interaction terms, are considered. Such systems in 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 function of the transverse field. Here we show that the off-diagonal dipolar interaction, although effectively reduced, destroys the spin glass order at any finite transverse field. Moreover, the resulting correlation length is shown to be small near the crossover to the paramagnetic phase, in agreement with the behavior of the nonlinear susceptibility in the experiments on LiHo$_x$Y$_{1-x}$F$_4$. Thus, we argue that the in these experiments a cross-over to the paramagnetic phase, and not quantum criticality, was observed.

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, superconducting insulator transition and high temperature superconductivity. Quantum magnets, and specifically their modeling by the transverse field Ising model (TFIM)

$$H = - \sum_{i,j} J_{ij} \hat{\tau}_i^z \hat{\tau}_j^z - \Delta \sum_i \hat{\tau}_i^x. \quad (1)$$

are a particularly good laboratory to study QPT, as 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, and maybe the best realization is in anisotropic dipolar systems. In this systems the dipolar energy dominates the spin-spin interaction, and the crystal field generates strong anisotropy resulting in a ground state Ising like doublet for the single spins and an effective reduction of all but the longitudinal interaction terms.

Indeed, LiHo$_x$Y$_{1-x}$F$_4$ with $x=1$ was shown [1] to exhibit a ferromagnetic to paramagnetic (PM) transition as 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 a transverse magnetic field 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$_x$Y$_{1-x}$F$_4$ compound in particular, the off-diagonal terms of the dipolar interaction, albeit effectively reduced, qualitatively change the physics of the problem. In particular, in the presence of a transverse field the off-diagonal dipolar 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 off-diagonal dipolar terms results in the absence of long-range SG order at any finite $H_t$, and a reduction of the cusp in the non-linear susceptibility at the crossover to the PM phase. Thus, we argue that the experimental line drawn at the peak values of the non-linear susceptibility [6] is not a phase transition line. Except for the point at $H_t=0$, this line corresponds to a cross-over between a paramagnet to a phase we denote a “quasi spin-glass” (QSG). 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 $\nu$ calculated below. The domain structure is maintained until the crossover field, where fluctuations between the relevant Ising like states dominate and the system becomes PM, via a crossover and not a quantum critical point.

FIG. 1: Schematic picture for the $T=0$ behavior of a dipolar Ising SG in a transverse field $H_t$. The typical size $\xi$ of a SG ordered domain (depicted below the x-axis) decreases with $H_t$, with a critical exponent $\nu$ (see text). At large enough $H_t$ the system becomes PM, via a crossover and not a quantum critical point.

Theoretical considerations.— Our analysis below is valid
both specifically to the LiH04-Y1−xF4 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 ground state 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

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

(2)

Here \(i, j\) 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 anisotropy constant due to the crystal field. For \(H_i = 0\) the GS of a single spin is doubly degenerate with \(s_z = \pm s\) and zero energy. The corresponding states are denoted \(|\uparrow_s\rangle\) and \(|\downarrow_s\rangle\). The first excited states have \(s_z = \pm (s-1)\) and energy \(\Omega_x = (2s-1)D\). Throughout the paper we assume that \(\Omega_x \gg \mu_B H_i, V_{\text{max}}\) where \(V_{\text{max}}\) is the largest dipolar energy between two spins in the system. We now define \(\mathcal{H} = \mathcal{H}_\parallel + \mathcal{H}_\perp\) such that

\[ \mathcal{H}_\parallel = -D \sum_i [(S_i^z)^2 - s^2] - \frac{1}{2} \sum_{i \neq j} V_{ij}^{\alpha \beta} S_i^\alpha S_j^\beta, \]  

(3)

and

\[ \mathcal{H}_\perp = -\frac{1}{2} \sum_{i \neq j} \sum_{(\alpha \beta) \neq (zz)} V_{ij}^{\alpha \beta} S_i^\alpha S_j^\beta - \mu_B H_i \sum_i S_i^z. \]  

(4)

We assume that the dilution is such that \(\mathcal{H}_\parallel\) is equivalent to the classical Ising model with random interactions and exhibits a SG phase at low temperature. As this classical dipolar Ising SG is equivalent to the short range Edwards-Anderson model \(\mathcal{H}_J\) (Eq. (1) with random nearest neighbor \(J_{ij}\) and \(\Delta = 0\)), our analysis is done within the scaling (“droplet”) picture \(\mathcal{H}_J\) which accounts for its behavior at large sizes. The GS of \(\mathcal{H}_J\) is then a two-fold degenerate states \(|\psi_0\rangle, |\tilde{\psi}_0\rangle\) with states \(|\psi_0\rangle\), \(|\tilde{\psi}_0\rangle\), which are related by \(S_z \rightarrow -S_z\) 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 the off-diagonal dipolar terms, this symmetry is not preserved. The GS degeneracy breaks, and the system gains energy by choosing locally a state similar to \(|\psi_0\rangle\) or \(|\tilde{\psi}_0\rangle\) according to which optimizes the energy gain due to \(\mathcal{H}_\perp\).

Following the scaling picture of Fisher and Huse \(\mathcal{H}_J\) and using an Imry-Ma \(\mathcal{H}_J\) like 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. (5) below) is then compared with the energy cost due to the domain wall formation, and the correlation length is obtained \(\xi\). Although, as is shown below, one can define an effective longitudinal random field at each site and use a direct analogy to the Ising SG in a random field, we would proceed by calculating directly the energy gain.

Consider first

\[ \mathcal{H}_\perp' = - \sum_{i \neq j} V_{ij}^{zz} S_i^z S_j^z - \mu_B H_i \sum_i S_i^z. \]  

(5)

The addition of \(\mathcal{H}_\perp'\) to \(\mathcal{H}_\parallel\) changes \(|\psi_0\rangle\) to \(|\psi\rangle\) and \(|\tilde{\psi}_0\rangle\) to \(|\psi'\rangle\) with energies \(E_\psi\) 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 theory \(E_\psi = E_\psi + E^{(2)}_{\psi}\) where

\[ E^{(2)}_{\psi} = -\frac{(\langle \psi | \sum_{i \neq j} V_{ij}^{zz} S_i^z S_j^z + \mu_B H_i \sum_i S_i^z | \psi \rangle)^2}{\Omega_o}. \]  

(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 \(s_z = \pm (s-1)\). Therefore the energy of all relevant excited states is \(\Omega_o\) in leading order, and 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 \(E_\psi\) and \(E_{\psi'}\), while the term linear in \(H_t\) is in equal magnitude but has opposite signs for \(E_\psi\) and \(E_{\psi'}\). Using the fact that since \(k \neq l\) the operators commute we obtain

\[ \delta E = \frac{4}{\Omega_o} \langle \psi_0 | \mu_B H_i \sum_i S_i^z \sum_{k \neq i} V_{ki}^{zz} S_k^z S_i^z | \psi_0 \rangle \]  

(7)

and therefore

\[ \delta E = 4 \frac{\mu_B H_i}{2\Omega_o} \sum_{k \neq i} V_{ki}^{zz} \langle \psi_0 | S_k^z | \psi_0 \rangle = \frac{2s^2 \mu_B H_i}{\Omega_o} \sum_i h_i^z, \]  

(8)

where we define \(h_i^z \equiv \sum_k V_{ki}^{zz} \langle S_k^z \rangle\) as an effective transverse magnetic field at site \(i\). For each \(i\) all the \(V_{ki}\)’s are small except the few for which the sites \(i\) and \(k\) are spatially close. Due to 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 \(\delta E\) 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

\[ \langle \delta E \rangle = 2s^2 \mu_B H_t V \sqrt{N} / \Omega_o, \]  

(9)

where \(V\) is the average magnitude of \(|\tilde{V}_i|\), and we choose \(|\psi_0\rangle\) and \(|\tilde{\psi}_0\rangle\) such that \(\delta E > 0\).

The above result 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) \(\mathcal{H}_J\). We consider system sizes in the regime where they are much smaller than \(\xi\). This is important for our calculation, since then we are dealing with single domains, and therefore the two lowest states correspond to \(|\psi\rangle, |\psi'\rangle\), and the gap to \(\delta E\). 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$_x$Y$_{1-x}$F$_4$ lattice. Since we are interested in small fields, it is sufficient to use $s = 1$ particles with on-site anisotropy $\Omega_o$. We therefore study the spin-1 version of $\mathcal{H} = \mathcal{H}_\parallel + \mathcal{H}_\perp$:

$$\mathcal{H}_1 = -\sum_{i \neq j} \left[ \frac{1}{2} V_{ij}^z S_i^z S_j^z + V_{ij}^{yz} S_i^y S_j^z + V_{ij}^{yz} S_i^z S_j^y \right] - \mu B H_i \sum_i S_i^z - \Omega_o \sum_i \left( (S_i^z)^2 - s^2 \right).$$

Here and below all energy scales are expressed in units of the typical n.n. dipolar energy $V_0$. We fix the dilution to a constant $x = 3/16 = 18.75\%$ 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 $V = 0.8$. Thus the Lanczos ED have been performed in the full $d = [2s + 1]N$ dimensional Hilbert space for $N = 6, 9$ and 12 $s = 1$ spins [12]. Then, the gaps have been computed for 10,000 independent random samples for each size.

For the Ising SG in longitudinal field it was argued [8, 16] and then verified experimentally [17, 18, 19] and numerically [20, 21] that there is no de Almeida Thouless line [22], and no SG phase at any finite field. At $H_i \ll \Omega_o/\mu_B$ 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 $H_i \to 0$ the transverse length diverges with the same form [3] of the critical exponent $\nu = \frac{1}{2(3/2 - \delta d)}$.

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

Interestingly, the two effects of the transverse magnetic field, i.e. inducing the crossover to the paramagnetic phase, and the reduction in $\xi$ calculated above, behave very differently as function of $H_i$. The former is dictated by fluctuations between the two single spin Ising ground states, which depend on $H_i$ to a high power, of order $s$, and are practically negligible as long as $H_i \ll \Omega_o/\mu_B$. However, the fluctuations that dictate the reduction of $\xi$ at low transverse fields are between each single spin ground state and its first excited state at energy $\Omega_o$. The latter depend on $H_i$ to second order and result in a reduction of $\xi$ which depends on $1/H_i$ to a power $\nu$ close to unity. Therefore, the disordering of the SG order by $H_i$ occurs in two stages. At low field domains of size $\xi$ are formed, within which the GS is very similar to either of the two zero field SG ground states. At $H_i \approx \Omega_o/\mu_B$ a crossover occurs where the order within each domain is destroyed, and each spin in the system fluctuates independently. Importantly, when reaching the crossover region at very low $T$ one is already in the regime where $\xi \approx 1$ in units of inter-spin spacing, resulting in small features in the relevant susceptibilities, in agreement with experiment [2, 3]. We would like to emphasize that the understanding of the scenario above requires a model in which the large spins are considered and the anisotropy in explicitly taken into account. Indeed, the anisotropy energy $\Omega_o$ enters explicitly into Eqs. (2, 11). A presumably simpler model in which one treats spin-half particles and models the effective reduction of the off-diagonal terms in the dipolar interaction by a multiplicative reduction factor will not be sufficient, since in such a model both the re-
duction in the correlation length and the crossover to the PM phase are induced by fluctuations between the Ising ground states, and therefore have the same scale in magnetic field.

In addition to changing the symmetry of the system at $H_t 
eq 0$, resulting in the destruction of the SG phase and the QPT to the PM phase, the off-diagonal terms of the dipolar interaction also enhance the effective transverse field $\tilde{H}_t$. Although, in principle, $h_{t}^{z}$ [see Eq. (6)] is a random quantity, domains of size $\xi$ choose to be in a state equivalent to $|\psi\rangle$ or $|\psi\rangle$ by the maximization of $\sum_{k} h_{t}^{z}$. As a result a net magnetic field in the $x$ direction, $\langle h_{t}^{x}\rangle = (\delta E)/N \propto \xi^{-3/2}$, is added to the external transverse field. As the crossover region is approached $\xi$ is small and the effective transverse field due to the off-diagonal dipolar interaction is significant. We thus give a precise physical origin to the conjecture made in Ref. [23].

Our analysis above could equally be done by defining $\delta E$ in Eq. (8) as $\sum_{i} h_{k}^{z}(S_{i}^{z})$, where $h_{k}^{z} = (2s_{\mu B}H_{t}/\Omega_{o}) \sum_{i} V_{ki}^{z}$. 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 $\delta E$ performed above.

Experimental consequences.— The crystal field Hamiltonian in LiHo$_x$Y$_{1-x}$F$_4$ is different from the one given in Eq. [2]. Furthermore, the strong hyperfine interactions strongly re-normalize the parameters of the TFIM, invalidating the simple model in the electronic degrees of freedom [23]. Still, for our purpose here an equivalent physical picture emerges: the two relevant (electro-nuclear) Ising states of each Ho ion couple very weakly at small transverse field, and the relevant excited states are at $\approx 10K$ above the ground states. 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$_x$Y$_{1-x}$F$_4$ system [2, 3], with $\Omega_{o} \approx 10K$ and suggest that LiHo$_x$Y$_{1-x}$F$_4$ is not a SG at any $H_t \neq 0$. Furthermore, the peculiar experimental result [6] where the cusp in the nonlinear susceptibility is reduced with decreasing $T$ is naturally explained: as $T$ is reduced the crossover to the PM phase occurs at larger transverse fields. This results in smaller correlation length $\xi$, and therefore a diminishing of the cusp in the nonlinear susceptibility [3]. In addition, the re-normalization of the effective spin $\tilde{S}$ specific to the LiHo$_x$Y$_{1-x}$F$_4$ compound further reduces the nonlinear susceptibility near the crossover.

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. [6]. As it is clear by our analysis here that it is a crossover and not a phase transition that was 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 crystal field terms which induce quantum fluctuations between the Ising like doublet (such as $(S_{2}^{z} + S_{2}^{z})$ terms added to the Hamiltonian [2] for integer spin systems).

Recently there is an increasing experimental [17, 18, 19] and numerical [20, 21] support for the validity of the droplet picture in describing short range Ising SG in general, and to its prediction [3, 14] of the non-existence of a de Almeida-Thouless [22] in particular. For the anisotropic dipolar systems discussed here the crossover to the PM phase at $H_t \approx \Omega_{o}/\mu_B$ is a result of quantum fluctuations, and there is no analog to the de Almeida-Thouless line. However, at $H_t \ll \Omega_{o}/\mu_B$ the system is equivalent to a classical Ising SG in a small random longitudinal field. Thus, the above numerical and experimental results [17, 18, 19, 20, 21] support the validity of the droplet picture for dipolar Ising systems in small transverse field as well. Still, we believe that experiments that would directly observe whether dipolar Ising glasses in general and LiHo$_x$Y$_{1-x}$F$_4$ in particular have a SG phase at a finite transverse magnetic field are of much interest, both as a verification of our results, and as an additional support for the droplet picture in general.

Finally, our analysis is applicable also to any Ising SG where the dipolar interactions are present, even if the interaction that governs the ordering is different. In such case the correlation length will be given by [7] $\xi_{J} \approx \left(\frac{\Omega_{o}/J}{\mu_B H_{t}}\right)^{(3/2-d)^{-\theta}}$, where $J$ is the strength of the dominant interaction. The qualitative picture will remain similar, only now the size of the domains at the quantum crossover to the PM phase would be $\approx \left(\frac{\Omega_{o}}{J}\right)^{1/(3/2-d)^{-\theta}}$.

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Computationally speaking, the situation here is extreme in the sense that the spin Hamiltonian (10) on the randomly diluted LiHo$_x$Y$_{1-x}$F$_4$ lattice has lost all possible space and internal-spin symmetries.

Note a finite size effect due to a short range interaction which is visible for $N = 6$ but disappears at larger $N$.

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