Collective Phenomena in the LiHo$_x$Y$_{1-x}$F$_4$ Quantum Ising Magnet: Recent Progress and Open Questions

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Abstract. In LiHo$_x$Y$_{1-x}$F$_4$, the magnetic Holmium Ho$^{3+}$ ions behave as effective Ising spins that can point parallel or antiparallel to the crystalline c-axis. The predominant inter-Ho$^{3+}$ interaction is dipolar, while the Y$^{3+}$ ions are non-magnetic. The application of a magnetic field $B_x$ transverse to the c-axis Ising direction leads to quantum spin-flip fluctuations, making this material a rare physical realization of the celebrated transverse field Ising model. The problems of classical and transverse-field-induced quantum phase transitions in LiHo$_x$Y$_{1-x}$F$_4$ in the dipolar ferromagnetic ($x = 1$), diluted ferromagnetic ($0.25 \lesssim x < 1$) and highly diluted $x \lesssim 0.25$ dipolar spin glass regimes have attracted much experimental and theoretical interest over the past twenty-five years. Two questions have received particular attention: (i) is there an antiglass (quantum disordered) phase at low Ho$^{3+}$ concentration and (ii) what is the mechanism responsible for the fast $B_x$-induced destruction of the ferromagnetic ($0.25 \lesssim x < 1$) and spin glass ($x \lesssim 0.25$) phases? This paper reviews some of the recent theoretical and experimental progress in our understanding of the collective phenomena at play in LiHo$_x$Y$_{1-x}$F$_4$, in both zero and nonzero $B_x$.

1. The pure LiHoF$_4$ material

1.1. Dipolar ferromagnetism

The insulating rare-earth compound LiHoF$_4$ has the scheelite structure depicted in Fig. 1 [1]. In this material, only the Holmium ions, Ho$^{3+}$, are magnetic. LiHoF$_4$ forms a tetragonal structure (space group $C_{4h}^0$ – $I4_1/a$) with lattice constants $a = 5.175$ Å and $c = 10.75$ Å. There are 4 magnetic Ho$^{3+}$ ions per unit cell with fractional coordinates $(0,0,\frac{1}{2})$, $(0,\frac{1}{2},\frac{3}{4})$, $(\frac{1}{2},\frac{1}{2},0)$ and $(\frac{1}{2},0,\frac{1}{4})$. In Fig. 1, the lines connecting the central Ho$^{3+}$ ion with neighbouring F$^-$ ions and then to the nearest Ho$^{3+}$ ions illustrate the superexchange pathways between nearest-neighbour Ho$^{3+}$ cations [2]. As in most magnetic rare-earth compounds, the unfilled 4f electronic orbitals of the Ho$^{3+}$ ions are not very spatially extended. This makes the exchange interaction weak and causes magnetostatic dipolar couplings to be the strongest inter-ionic interactions. Indeed, LiHoF$_4$ is a dipolar Ising ferromagnet with a magnetic moment of $\sim 7\mu_B$ per Ho$^{3+}$ ion [2] and with a critical temperature $T_c \approx 1.53$ K [2]. In the mid-seventies, crystal-field calculations [3], susceptibility [4] and spectroscopic measurements [5] showed that LiHoF$_4$ is an Ising-like...
Figure 1. The left panel shows the crystalline tetragonal structure of LiHoF$_4$ with lattice spacing $a = 5.175$ Å and $c = 10.75$ Å. The thin solid lines illustrate the superexchange pathways between magnetic Ho$^{3+}$ ions (black circles) mediated via fluorine F$^-$ ions (green circles). In LiHo$_x$Y$_{1-x}$F$_4$, the magnetic Ho$^{3+}$ ions are substituted randomly by non-magnetic Y$^{3+}$ ions, with the lattice structure remaining the same for all $x$. The global Ising direction is along the $c$ axis and a magnetic field $B_x$ is applied perpendicular to that axis in transverse field experiments. The right panel shows a schematic temperature ($T$) – dilution ($x$) – transverse field ($B_x$) phase diagram of LiHo$_x$Y$_{1-x}$F$_4$. At high temperature $T$ or large $B_x$, the system is in a paramagnetic (PM) phase. For $x = 1$, LiHoF$_4$ is an Ising dipolar ferromagnet (FM) below $T_c = 1.53$ K. Upon the substitution of Ho$^{3+}$ by Y$^{3+}$, the FM phase persists down to $x \sim 0.25$. Upon cooling the diluted FM for $0.25 \lesssim x \lesssim 0.5$, magnetic susceptibility measurements find the development of a “ferroglass” regime (FM or SG?). As $x$ decreases, random frustration builds in and, for $x \lesssim 0.25$, a dipolar Ising spin glass (SG) phase develops (shaded blue region). Whether the dipolar SG exists down to $x = 0^+$ or an exotic quantum disordered “antiglass” (AG) phase occurs at $x > 0$ (red line segment) is not yet resolved and is the subject of much controversy. The application of a transverse field $B_x$ introduces quantum fluctuations causing a reduction of $T_c(x)$ (surface delineated by dotted lines) which, ultimately, drives a zero temperature quantum phase transition at zero temperature (dash-line curve in the $x - B_x$ plane).

The right panel shows a schematic temperature ($T$) – dilution ($x$) – transverse field ($B_x$) phase diagram of LiHo$_x$Y$_{1-x}$F$_4$. At high temperature $T$ or large $B_x$, the system is in a paramagnetic (PM) phase. For $x = 1$, LiHoF$_4$ is an Ising dipolar ferromagnet (FM) below $T_c = 1.53$ K. Upon the substitution of Ho$^{3+}$ by Y$^{3+}$, the FM phase persists down to $x \sim 0.25$. Upon cooling the diluted FM for $0.25 \lesssim x \lesssim 0.5$, magnetic susceptibility measurements find the development of a “ferroglass” regime (FM or SG?). As $x$ decreases, random frustration builds in and, for $x \lesssim 0.25$, a dipolar Ising spin glass (SG) phase develops (shaded blue region). Whether the dipolar SG exists down to $x = 0^+$ or an exotic quantum disordered “antiglass” (AG) phase occurs at $x > 0$ (red line segment) is not yet resolved and is the subject of much controversy. The application of a transverse field $B_x$ introduces quantum fluctuations causing a reduction of $T_c(x)$ (surface delineated by dotted lines) which, ultimately, drives a zero temperature quantum phase transition at zero temperature (dash-line curve in the $x - B_x$ plane).

dipolar ferromagnet with a highly anisotropic $g$-tensor ($g_\perp \approx 0$ and $g_\parallel \approx 14$). The dipolar interaction has a long and interesting history and this physical realization rekindled interest in dipolar ferromagnetism. Luttinger and Tisza had found that the ground state of a dipolar ferromagnet depends on both the lattice structure and sample shape [6]. However, taking the demagnetization field into account, Griffiths showed that, in the absence of an applied magnetic field, the free energy is independent of the sample shape [7]. Using renormalization group arguments, Larkin and Khmelnitskii [8] and Aharony [9] showed that $d^* = 3$ is the upper critical dimension for mean-field critical behaviour in a dipolar ferromagnet. This observation led to significant experimental efforts to find the expected logarithmic corrections to mean-field theory in LiHoF$_4$ [10, 11, 12, 13, 14]. The demagnetization field also renders the ferromagnetic transition quite peculiar in the presence of dipolar interactions [4] as these oppose a uniform magnetization and leads to the formation of domains below the transition [5, 15]. The size and shape of the domains depend on an energy balance between surface and bulk contributions [16]. For LiHoF$_4$, the ground state domain configuration is expected to consist of parallel antiferromagnetically aligned sheets [17, 18, 19]. The size of the domains increases with decreasing demagnetization factor and, in the limit of infinitely long thin samples, a state of uniform magnetization is possible. Also, for thick samples, a branched pattern is predicted in the domain structure for
a dipolar Ising ferromagnet [17, 18]. LiHoF$_4$ is an excellent test case for theories of domain formation due to the availability of an accurate microscopic model of the material (see below). It remains an open experimental challenge to verify the nature of the predicted rich domain structure.

1.2. Microscopic model and transverse field Ising model for LiHoF$_4$

The magnetic properties of LiHoF$_4$ are determined by the 4$f^{10}$ electrons of the Ho$^{3+}$ ions. Applying Hund’s rules results in a 17-fold degenerate $^5I_9$ electronic ground state. The crystal field from the surrounding ions, described by a crystal field potential $V_{\text{CF}}$, lifts this degeneracy and gives rise to a two-fold non-Kramers degenerate ground state, $|\psi_0^\pm\rangle$, and an excited singlet, $|\psi_e\rangle$, at an energy of approximately 10 K above $|\psi_0^\pm\rangle$ [3, 5]. The minimal microscopic Hamiltonian in the presence of a transverse magnetic field $B = B_x \hat{x}$ can be written as [20]:

$$H = \sum_i V_{\text{CF}}(\mathbf{J}_i) - \frac{\mu_0}{4\pi} g_L \mu_B \sum_i B_x J_i^x + \frac{1}{2} \sum_{i \neq j} \frac{\mu_0}{4\pi} (g_L \mu_B)^2 \sum_{\mu, \nu} \mathbf{L}_{ij}^{\mu \nu} J_i^\mu J_j^\nu + \mathcal{J}_{\text{ex}} \sum_{(i,j)} \mathbf{J}_i \cdot \mathbf{J}_j + A \sum_i \mathbf{J}_i \cdot \mathbf{J}_i,$$  

where $\mu, \nu = x, y, z$. $\mathbf{L}_{ij}^{\mu \nu}$ is the magnetic dipole interaction, $\mathbf{L}_{ij}^{\mu \nu} = [\delta^{\mu \nu} |\mathbf{r}_{ij}|^2 - 3(\mathbf{r}_{ij})^\mu (\mathbf{r}_{ij})^\nu] / |\mathbf{r}_{ij}|^5$, $\mathcal{J}_{\text{ex}}$ is the nearest-neighbour exchange interaction. $A$ is the strength of the hyperfine interaction and $\mathbf{J}_i (i = 7/2)$ is the total angular angular momentum vector of the Ho-nucleus at the $i$-th site. Disregarding the hyperfine interactions, an effective spin $1/2$ model for LiHoF$_4$ from the above microscopic model of Eq. (1) was derived in Ref. [20] and revisited in Ref. [21]. Without hyperfine interactions, the single site Hamiltonian (first two terms in Eq. (1)) can be diagonalized numerically for arbitrary value of the transverse field $B_x$. The ground state doublet is split by $B_x$, with an energy $\Delta(B_x)$ between the two states. Meanwhile, the excited $|\psi_e\rangle$ singlet state remains more than 10 K above the split doublet [20, 21]. Consequently, the $\mathbf{J}_i$ operators can be projected onto the two-dimensional subspace of the two lowest energy eigenstates via the relationship $J_i^\mu = C_{\mu 0} + \sum_{\nu=x,y,z} C_{\mu \nu}(B_x) \sigma_i^\nu$, hence providing a description in terms of effective pseudospin-$1/2$ operators [20, 21]. The strongest effective interaction is $\propto (C_{zz})^2 \mathbf{L}_{ij}^{zz} \sigma_i^z \sigma_j^z$ and, to an accuracy of a few percent [20, 21], the effective model can be written (up to a field dependent constant) as

$$H_{\text{Ising}} = -\frac{1}{2} \Delta \sum_i \sigma_i^x + \frac{1}{2} \sum_{i \neq j} \frac{\mu_0}{4\pi} (g_L \mu_B C_{zz})^2 \sum_{\mu, \nu} \mathbf{L}_{ij}^{\mu \nu} \sigma_i^\mu \sigma_j^\nu + \mathcal{J}_{\text{ex}} (C_{zz})^2 \sum_{(i,j)} \sigma_i^z \sigma_j^z. \quad (2)$$

Note that the $C_{\mu \nu}$ parameters and the effective transverse field $\Gamma$, with $\Gamma \equiv \Delta/2$, depend on the applied physical transverse field $B_x$. In the limit of small $B_x$, $\Delta(B_x) \propto B_x^2$ [20, 21]. In Eq. (2), $\Delta/2$ plays the role of an effective transverse field $\Gamma$ acting on the $x$-component of the pseudospin $\sigma_i^x$, hence the realization of a transverse field Ising model (TFIM) in LiHoF$_4$ [22].

For $B_x = 0 (\Delta = 0)$, the effective dipolar model (2) has been analyzed using mean-field theory [2, 4] and classical Monte Carlo simulations. Numerical Monte Carlo simulations are complicated by the difficult nature of the conditionally-convergent dipolar lattice sums. An early Monte Carlo simulation using free boundary conditions for rather small system sizes found a critical temperature of 1.89 K [23]. To obtain better accuracy with long range dipolar interactions, there are two standard approaches that can be implemented: Ewald summation [19, 21, 24, 25, 26, 27] and cavity methods [20, 28]. An early Monte Carlo simulation using the Ewald summation
method to a dipolar Ising ferromagnet on a body-centered lattice was carried out by Xu et al. [24, 25]. Monte Carlo simulations using the cavity method [20] and the Ewald method [19, 21] have determined the critical temperature for the dipolar model in Eq. (2) with $\Delta = 0$ on the LiHoF$_4$ lattice and estimated the nearest-neighbour (antiferromagnetic) exchange interaction $J_{\text{ex}}$ needed to lower the Monte Carlo $T_c$ value in order to match it to the experimental value of 1.53 K [2]. A more recent study, using the Ewald summation method and system sizes up to 32 000 dipoles, found that the magnetization, specific heat and susceptibility do match experimental results at a quantitative level [19]. The pure LiHoF$_4$ material can therefore be viewed as one of the best realizations of a dipolar Ising model. A review of materials that behave like Ising systems can be found in Ref. [29].

1.3. Hyperfine interaction
As in other Ho-based materials [30, 31], there is a strong hyperfine coupling ($A = 0.039$ K) between the electronic ($J = 8$) and nuclear ($I = 7/2$) moments of Ho$^{3+}$ in LiHoF$_4$. The hyperfine contribution dominates the specific heat below 0.5 K [2], and leads to a substantial increase in the critical transverse field at low temperatures [22]. As we shall discuss in Section 2, the hyperfine interaction also strongly influences the phase diagram of the dilute compound LiHo$_x$Y$_{1-x}$F$_4$ in a transverse field [32, 33]. The hyperfine interaction is therefore important and may not be omitted in a proper quantitative description of the low-temperature properties of LiHo$_x$Y$_{1-x}$F$_4$.

1.4. Transverse field effects
At a continuous phase transition, the order parameter fluctuates coherently over increasing length scales as the critical point is approached. At finite temperatures, the fluctuations are classical in nature sufficiently close to the critical temperature. In some systems, at temperatures near absolute zero, a phase transition can be induced by tuning an external parameter such as a magnetic field, pressure, doping or disorder. In such a case, the fluctuations are quantum mechanical in nature and the transition is called a quantum phase transition (QPT) [34, 35]. The archetypical model for a quantum phase transition is the transverse field Ising model (TFIM) [36, 37]. There are, however, not many real Ising magnets with experimentally accessible QPTs [38] since the required transverse field usually far exceeds laboratory fields (currently less than 50 T for DC fields). This would be the case for dipolar Ising spin ice materials [39], but not for the Dy(OH)$_3$ and Ho(OH)$_3$ dipolar Ising ferromagnets [38]. Due (i) to the weak dipolar interaction and (ii) the low-lying singlet $|\psi_0\rangle$ above the ground Ising doublet $|\psi^+\rangle$, LiHoF$_4$ is one of very few magnetic systems where one can observe the vanishing of the magnetization as a transverse field is applied [22]. Interestingly, recent neutron scattering measurements of LiHoF$_4$ have revealed that the relatively strong hyperfine interactions in Ho mask the true QPT of the hybridized electronuclear critical mode, and the energy gap in the solely electronic degrees of freedom remains finite as the system passes through the QPT [40].

Given the accuracy of the effective Ising model for LiHoF$_4$ in the absence of a transverse field, the material would seem to constitute an ideal testing ground for the inclusion of non-commuting quantum-mechanical terms in realistic spin models of magnetic materials. As outlined above, there is a well defined procedure to describe the effect of the transverse field $B_x$ in LiHoF$_4$ within a TFIM [20, 21]. However, a quantum Monte Carlo calculation [20] of the critical temperature, $T_c$, vs $B_x$ phase diagram reveals only a qualitative agreement with the experiment of Ref. [22]. In the low temperature region (below 0.5 K), the effect of the hyperfine interactions on the $B_x - T$ phase diagram becomes important, and this is only approximately taken into account in the simulation of Ref. [20]. Recent theoretical work proposes an alternative way to incorporate the effect of the hyperfine interactions in an effective model of LiHoF$_4$ in nonzero $B_x$ [33]. Notwithstanding the importance of hyperfine effects at low temperature, one notes that even
close to the classical critical temperature of $T_c = 1.53$ K, the experimental [22] phase boundary rises much more steeply than in the model of Eq. (2) [20, 21]. To compound the puzzle, a very recent experiment using dilatometry [41] confirms the previous $T_c(B_x)$ experimental phase diagram obtained on the basis of susceptibility measurements [22]. The reason for this stability of the ordered state with respect to weak $B_x$ is an open question. Possible explanations include magnetostriction effects, small terms that are neglected in the effective model of Eq. (2) (see Refs. [20, 21] for a discussion of those terms) or anisotropic exchange terms, such as $J_{uv}^{ex}(r_{ij}).J_i^uJ_j^v$, altogether ignored in the microscopic model of Eq. (1). While the latter interactions may largely be inconsequential at the classical transition, when projected within the manifold of $|v_0^\mp\rangle$ Ising doublets when $B_x = 0$ [42], these could perhaps end up affecting the $T_c(B_x \neq 0)$ phase diagram when $B_x \neq 0$.

2. LiHo$_x$Y$_{1-x}$F$_4$ – transverse field and random field physics

2.1. Disordered magnetic systems

All real materials contain a certain amount of frozen-in random disorder such as impurities, interstitials and crystalline defects. One may then ask how the thermodynamic properties and the stability of the ground state of an otherwise idealized pure system are affected by various types of random disorder [43, 44]. Since the mid 1970s, magnetic systems have proven to be almost ideal paradigms to study the role of random disorder in condensed matter systems. Examples of cornerstone questions in the field of random disordered systems are: Does a transition that is first order in a pure system remains first order in the presence of random disorder [45]? For a second order transition, does an arbitrary small amount of random disorder change the critical exponents and hence modify the universality class [46]? Does the long range order exhibited by a pure system survive arbitrary small random fields conjugate to the order parameter [47]? When the disorder is large enough and the interactions are in strong competition and lead to random frustration, is there a thermodynamic transition at nonzero temperature to a spin glass state [48, 49]? Under what conditions does a random magnet exhibit nonanalytical behavior above its critical temperature, in the so-called Griffiths phase, which is a remnant of the long range order phase displayed by the parent disorder-free system [50]? The above questions were originally by and large investigated for classical phase transitions at nonzero temperature. In the late 1980s, those and similar questions started to attract considerable interest in the context of quantum phase transitions [34, 35]. Here again, the LiHoF$_4$ material, or more precisely, its Y$^{3+}$ diamagnetically diluted LiHo$_x$Y$_{1-x}$F$_4$ variant, has proven to be a most interesting material to investigate quantum phase transitions in a disordered system that is tuned by an external parameter. As in the pure LiHoF$_4$ material, the parameter tuning the level of quantum (spin flip) fluctuations in LiHo$_x$Y$_{1-x}$F$_4$ is a magnetic field $B_x$ applied perpendicularly to the Ho$^{3+}$ Ising spin direction.

2.2. Transverse field and random field effects in LiHo$_x$Y$_{1-x}$F$_4$

The LiHo$_x$Y$_{1-x}$F$_4$ compound forms a solid solution from the pure dipolar ferromagnetic LiHoF$_4$ to the non-magnetic LiYF$_4$ without a change of the crystalline structure and, thanks to the close ionic radius of Ho$^{3+}$ and Y$^{3+}$, LiHo$_x$Y$_{1-x}$F$_4$ displays a minimal dependence of the $a$ and $c$ lattice parameters on $x$. The critical ferromagnetic transition temperature, $T_c(x)$, decreases as the magnetic Ho$^{3+}$ ions are substituted by non-magnetic Y$^{3+}$. For $0.25 \lesssim x < 1$, LiHo$_x$Y$_{1-x}$F$_4$ can be described as a diluted ferromagnet (see Fig. 1). As discussed in Section 1.1, the main interactions between the Ho$^{3+}$ ions are magnetostatic dipole-dipole interactions. The dipolar interaction is inherently frustrated since its sign, that of $L_{ij}^{uv}$ in Eq. (1), depends on the orientation of the magnetic moments with respect to $\vec{r}_{ij} \equiv \vec{r}_j - \vec{r}_i$, the position vector of ion $j$ relative to ion $i$. Hence, as a result of the random substitution Ho$^{3+} \rightarrow$ Y$^{3+}$, random frustration builds up as $x$ decreases until, for $x \approx 0.25$, the diluted ferromagnetic state gives
way to a dipolar spin glass phase (see Fig. 1). We note that there has been much debate as to whether such a spin glass phase occurs for $x \lesssim 0.25$ in LiHo$_x$Y$_{1-x}$F$_4$ [51, 52, 53]. We proceed in this section with the assumption that a thermodynamic spin glass phase does exist, at least for $0.15 \lesssim x \lesssim 0.25$. We return in Section 3 to the question of the existence of a spin glass phase in LiHo$_x$Y$_{1-x}$F$_4$ for $x \lesssim 0.25$, below the concentration where the diluted ferromagnetic phase has disappeared (see Fig. 1).

The essence of the research on transverse field induced quantum fluctuations and quantum phase transitions in LiHo$_x$Y$_{1-x}$F$_4$ originally lay in the expectation that this material may be a physical realization of a transverse field Ising model (TFIM) with Hamiltonian

$$H_{\text{TFIM}} = -\sum_{i>j} \Omega_{ij}^{zz} \sigma_i^z \sigma_j^z - \Gamma \sum_i \epsilon_i \sigma_i^z,$$

with effective transverse field $\Gamma$. In Eq. (3), the randomly frustrated $\Omega_{ij}^{zz}$ couplings model, or rather mimic, the site-diluted $\mathcal{L}_{ij}^{zz} \rightarrow \epsilon_i \epsilon_j \mathcal{L}_{ij}^{zz}$ in Eq. (2), where $\epsilon_i = 1$ when the site $i$ is occupied by a magnetic Ho$^{3+}$ ion and $\epsilon_i = 0$ when it is occupied by non-magnetic Y$^{3+}$. In theoretical studies, important simplifications have been made by taking a Gaussian distribution for $\Omega_{ij}^{zz}$ of mean $\Omega_0$ and standard deviation $\Omega$ and setting $\epsilon_i = 1 \forall i$ [54, 55, 56, 57]. Of particular interest in these studies was the prediction of Griffiths phase physics near the QPT both in two and three dimensional spin glass versions ($\Omega_0 = 0$) of Eq. (3) [56].

Two puzzles were identified when the behaviour of LiHo$_x$Y$_{1-x}$F$_4$ subject to $B_x \neq 0$ was first compared with expectations from theoretical and numerical studies. Firstly, while $dT_c/\partial x \propto x$ in the diluted ferromagnetic (FM) regime ($0.25 \lesssim x < 1.0$, see Fig. 1), as in mean-field theory [58], the rate at which $T_c(B_x)$ is reduced by $B_x$ gets progressively faster than mean-field theory predicts as the Ho$^{3+}$ concentration $x$ decreases [59]. This means that, in comparison with the energy scale for FM order that determines the classical transition at $T_c(B_x = 0, x)$, $B_x$ becomes more efficient at destroying FM order the lower the concentration $x$ is [59]. Secondly, for $B_x = 0$, LiHo$_{0.167}$Y$_{0.833}$F$_4$ has generally been believed to exhibit a conventional spin glass (SG) transition [51] (see however Ref. [52, 53]), with the transition signaled by a nonlinear magnetic susceptibility, $\chi_3$, diverging at $T_g$ as $\chi_3(T) \propto (T - T_g)^{-\gamma}$ [48]. However, $\chi_3(T)$ in LiHo$_{0.167}$Y$_{0.833}$F$_4$ becomes less singular as $B_x$ is increased from $B_x = 0$ [60, 61]. This suggests that, in contrast with numerical work [55, 56], no $B_x$-driven quantum phase transition between a polarized quantum paramagnetic state and a SG state occurs as $T \rightarrow 0$.

The resolution of these two puzzles lies in the observation that, in the properly derived effective low-energy theory akin to Eq. (2) when the occupation factor $\epsilon_i \neq 1$ [62, 63], the transverse field $B_x$ induces not only quantum fluctuations, but also random fields [43, 44, 47], $h_i^z$, that couple linearly to the $\hat{z}$ component of $\bar{\sigma}_i$ [32, 33, 62, 63, 64, 65]. There are two key physical ingredients responsible for these random fields. Firstly, the magnetic field $B_x$, perpendicular to the $\hat{z}$-axis Ising direction, induces a finite expectation value of the Ho$^{3+}$ magnetic moments along $\hat{z}$, $\langle \mu_i^z \rangle = g \mu_B \langle \hat{J}_i^z \rangle$, at all temperatures. Quantum mechanically, this moment arises from the $B_x$-induced admixing of the (initially strictly Ising) $|\psi_0^z\rangle$ ground doublet with the excited crystal field states. Secondly, the bare microscopic Hamiltonian in Eq. (1), as opposed to the effective TFIM of Eq. (2) and its simplified version in Eq. (3), contains off-diagonal (anisotropic) $\mathcal{L}_{ij}^{\alpha\beta}$ ($\alpha \neq \beta$) terms of dipolar origin that couple the various components $J_i^\mu$ of $\bar{J}_i$. In pure LiHoF$_4$, perfect mirror symmetries make the lattice sum $h_i^z \equiv \sum_j \mathcal{L}_{ij}^{zz} \langle J_j^z \rangle$ vanish identically. For diluted LiHo$_x$Y$_{1-x}$F$_4$, $h_i^z$ no longer vanishes; the transverse magnetic moments $\langle \mu_i^z \rangle$ induce, via the random $\epsilon_i \mathcal{L}_{ij}^{zz} \langle J_j^z \rangle$ terms, internal random fields that act on the $z$ component of $\bar{J}_i$, $\bar{J}_i^z$. In other words, diluted LiHo$_x$Y$_{1-x}$F$_4$ subject to a transverse field $B_x$ maps onto a TFIM with the addition of correlated random fields [62, 63]. For $0.25 \lesssim x < 1$, this makes LiHo$_x$Y$_{1-x}$F$_4$ a unique example of a random field Ising fermion magnet model [43, 44, 47], albeit with the random
fields being correlated in LiHo$_x$Y$_{1-x}$F$_4$. Indeed, the experimental procedure for creating such a random field Ising system is convoluted and typically considers a diluted Ising antiferromagnet in a uniform longitudinal field $B_z$ [66, 67]. Analytical [33, 62, 63, 64, 65] and numerical [62, 63, 64] studies have shown, rather convincingly, that the presence of $B_x$-induced random fields $h_i^z$ can explain the two aforementioned puzzles observed in LiHo$_x$Y$_{1-x}$F$_4$ in nonzero $B_x$, in both the diluted FM regime and in the SG regime. In particular, the $B_x$-induced random fields $h_i^z$ eliminate the divergence of the nonlinear susceptibility $\chi_3$ in the spin glass regime $x \lesssim 0.25$ [32, 62, 64].

2.3. Open questions regarding the random field physics in LiHo$_x$Y$_{1-x}$F$_4$

Because of space constraint, we have focused above on the role of the induced random fields on the phase transitions that occur in the diluted ferromagnetic and spin glass regimes. As the critical (ferromagnetic or spin glass) temperature decreases, and drops below approximately 0.5 K, the role of the hyperfine interaction in inhibiting quantum fluctuations and strongly renormalizing the critical transverse field cannot be ignored [32, 33]. While progress has been achieved as to how to proceed [33], work remains to be done to investigate via quantitatively accurate calculations (e.g. quantum Monte Carlo simulations) the role of hyperfine interactions on the global $x - T - B_x$ phase diagram of LiHo$_x$Y$_{1-x}$F$_4$.

In the context of the physics of induced random fields, it is worthwhile to note that theoretical work predicts that in the simplest TFIM of Eq. (3), the addition of uncorrelated random fields should destroy the zero temperature quantum criticality and drive the renormalization group flow towards the fluctuationless (classical) zero temperature random field fixed point [68]. It would be interesting to investigate the implication of that result for the correlated $h_i^z$ random fields discussed above and that exist in LiHo$_x$Y$_{1-x}$F$_4$ in nonzero $B_x$, as suggested by some experiments [69]. Finally, we note that the presence of induced random fields has been invoked to interpret the transverse field and temperature dependence (e.g. scaling behavior) of the magnetic susceptibility in LiHo$_x$Y$_{1-x}$F$_4$ ($x = 0.44$). In particular, these random fields have been postulated to give rise to Griffiths’ singularity effects in the paramagnetic regime at temperatures just above $T_c$ [69]. It would be useful to explore this claim further. In fact, a systematic experimental investigation of the fascinating physics of random field systems in the diluted ferromagnetic regime of LiHo$_x$Y$_{1-x}$F$_4$ and subject to a nonzero transverse field $B_x$ is probably warranted.

3. LiHo$_x$Y$_{1-x}$F$_4$ – spin glass vs antiglass

3.1. Spin glass vs antiglass physics in LiHo$_x$Y$_{1-x}$F$_4$

The previous section discussed the properties of LiHo$_x$Y$_{1-x}$F$_4$ in nonzero transverse field $B_x$. We now turn to the question of the existence of a spin glass phase in LiHo$_x$Y$_{1-x}$F$_4$ for $x \lesssim 0.25$ and in zero transverse field ($B_x = 0$).

The $x - T$ phase diagram of LiHo$_x$Y$_{1-x}$F$_4$ in the limit of large dilution ($x \lesssim 0.25$) has been and remains a subject of much controversy. Theoretical studies of the dilute Ising dipolar model predict that a spin-glass state may be favored over ferromagnetic order at high dilution [24, 70]. Since there is no percolation threshold for long range dipolar couplings, a thermodynamic spin glass state is expected to persist all the way to $x = 0^+$ [70]. Furthermore, a study of the quantum corrections to the Ising model arising from interaction-induced virtual crystal field fluctuations finds that these quantum effects should be small in LiHo$_x$Y$_{1-x}$F$_4$ [42]. This is in contrast to the Tb$_2$Ti$_2$O$_7$ and Tb$_2$Sn$_2$O$_7$ pyrochlore Ising magnets [71, 72] and, therefore, are unlikely to destroy the spin glass phase in LiHo$_x$Y$_{1-x}$F$_4$ at finite Ho$^{3+}$ concentration. Consequently, in light of the accumulated evidence for a thermodynamic spin glass phase transition in the three-dimensional Edwards-Anderson (EA) Ising spin glass model [49], one would also expect such a
transition in LiHo$_x$Y$_{1-x}$F$_4$ for $0 < x \ll 1$ and $B_z = 0$ since the EA model and dipolar systems should be in the same universality class for a spin glass transition [73].

Measurements of the imaginary part of the magnetic susceptibility, $\chi''$ in LiHo$_{0.045}$Y$_{0.955}$F$_4$ by Reich et al. [74, 75] was the first to challenge the above theoretical picture of a spin glass phase down to $x = 0^+$. In a typical glassy system, the frequency distribution of $\chi''$ broadens upon lowering the temperature [48]. In contrast, the authors of Refs. [74, 75] found a distribution that *narrowed* as the temperature is reduced, indicative of an overall speed-up of the dynamics and an “antiglass” (quantum disordered) ground state. The measurement was later repeated [76] to lower temperatures (50 mK), still showing an evident narrowing of the spectrum. The manifestation of dissipationless dynamics of a broad inhomogeneous distribution of quantum oscillators within the inferred antiglass regime was further demonstrated through (i) hole burning in the AC susceptibility spectrum [76] and, (ii) perhaps most interestingly, via the observation of persistent oscillations for up to 30 seconds after an external AC field is turned off [76]. It has been argued that this antiglass behaviour may originate from a quantum mechanical entanglement of the magnetic dipole moments and the formation of a sort of random-singlet state [77]. However, as mentioned above, the quantum effect corrections to the otherwise perfect Ising nature of the body-centered cubic lattice and did not investigate the spin glass freezing [24]. The traditional way to identify the glass transition has been to calculate the Edwards-Anderson overlap between two replicas, #1 and #2, $q = 1/N \sum_i \sigma_i^1 \sigma_i^2$, and to determine the temperature at which the corresponding Binder ratios, $g = 1 - [(q^4)^2 / 3(q^2)^4]$, for different system sizes intersect. However, several Monte Carlo studies [26, 27] have been unable to detect a spin glass transition on the basis of such intersection, or other finite-size scaling properties of $q$ (see Ref. [78]). These “first generation” Monte Carlo simulations of diluted dipole-coupled Ising spins did therefore suggest that the absence of a thermodynamic spin glass transition in LiHo$_x$Y$_{1-x}$F$_4$ at small $x$ could possibly have a *classical* origin. However, as we discuss in the following section, more recent experimental and numerical studies reveal a different picture.
The left panel shows a plot of the imaginary part of the AC susceptibility, $\chi''$, as a function of frequency, $f$, normalized to its maximum value $\chi''_{\text{max}}$ at frequency $f_{\text{max}}$, and for temperatures between 77 mK and 260 mK. The figure shows a broadening of the spectrum below and above the peak frequency $f_{\text{max}}$ at which $\chi''(f)$ peaks (adapted from Ref. [79]). The right panel shows the real part of the AC susceptibility, $\chi'(f)$, as a function of temperature. One notes the general good agreement with the $\lim_{f\to 0} \chi''(f)$ data set from Ref. [75] down to a temperature $\sim 180$ mK as well as with the DC susceptibility data determined via classical Monte Carlo [27]. The figure, adapted from Ref. [79], illustrates the disagreement with the results of Jönsson et al. in Ref. [52] as well as with the $T^{-0.75}$ behaviour reported in Ref. [77] and argued to support an entangled collective singlet ground state.

### 3.2. Recent developments in spin glass physics in LiHo$_x$Y$_{1-x}$F$_4$

Since much of the evidence for the antiglass phase is based on the scaling of the imaginary part of the AC susceptibility, the reproducibility of these measurements is very important. Recent measurements by Quilliam et al. [79] contradict the earlier results [74, 75, 76] and find that the frequency distribution of $\chi''$ broadens as the temperature is reduced also in the high dilution limit ($x = 4.5\%$) as shown in the left panel of Fig. 2. Furthermore, the quasi-static susceptibility, $\chi_0$, reported in Ref. [79] does not show the unusual $T^{-0.75}$ divergence found earlier [77]. We note, perplexingly, as noted in Refs. [27, 79], that the quasi-static susceptibility $\chi_0$ reported by Ghosh et al. in Ref. [77] does not agree with the previous results [74, 75] by the same group on, presumably, the very same sample. Meanwhile, the experimental $\chi_0$ data of Quilliam et al. [79] agree reasonably well with the early results of Refs. [74, 75] down to a temperature of approximately 180 mK, as well as with the DC susceptibility found in a classical Monte Carlo simulation of Eq. (2) with $B_x = 0$ [27], as shown in the right panel of Fig. 2. The work of Ref. [79] would appear to give a strong indication that there is conventional spin glass freezing in the high dilution limit of LiHo$_x$Y$_{1-x}$F$_4$. Furthermore, the specific heat data reported in Ref. [80] do not display any of the unusual features that were previously interpreted as a telltale signature of entangled magnetic moments within a quantum disordered antiglass state [77]. Note that the specific heat determined via classical Monte Carlo simulations [27] does not agree particularly well with either set of experimental data. This discrepancy may be in part due to the difficulty in taking into account accurately of the hyperfine contribution to the experimental specific heat data.

The reason for the difference between the AC susceptibility and specific heat experimental results of Refs. [74, 75, 76, 77] and Refs. [79, 80] remains an open question. Due to the very slow dynamics, all these measurements are technically very demanding. However, we believe...
that it would be important to settle the question of the nature of the high dilution phase in LiHo$_x$Y$_{1-x}$F$_4$. Perhaps the study of other highly diluted Ising magnetic materials could help shed some light on the physics at play in LiHo$_x$Y$_{1-x}$F$_4$. In this context, a study of the highly diluted Ising spin ice materials $(\text{Dy}_x\text{Y}_{1-x})_2\text{Ti}_2\text{O}_7$ and $(\text{Ho}_x\text{Y}_{1-x})_2\text{Ti}_2\text{O}_7$ [81] as well as the proposed Ho$_x$Y$_{1-x}(\text{OH})_3$ and Dy$_x$Y$_{1-x}(\text{OH})_3$ TFIM materials [38] to compare with LiHo$_x$Y$_{1-x}$F$_4$ could prove interesting and instructive.

The failure of previous Monte Carlo simulations [26, 27, 78] to find a thermodynamic spin glass transition in the dipolar model would seem to have been, at least partially, explained recently. As in three-dimensional Edward-Anderson Ising spin glass model [49], the spin glass correlation length $\xi_{sg}$ turns out to be a much better indicator of a spin glass transition than the aforementioned Binder ratio $g$. A recent Monte Carlo study [82] that focuses on $\xi_{sg}$ confirms that this is also the case also for the dipolar Ising model. In Ref. [82], a finite-size crossing of $\xi_{sg}$ divided by the linear system size $L$, $\xi_{sg}/L$, is observed, providing fair evidence for a finite temperature spin-glass transition, in agreement with the most recent AC susceptibility experiments [79]. However, the critical temperature determined by the crossing of $\xi_{sg}/L$ is slightly different for $\xi_{sg}$ along the $a$ and $c$ axes. This indicates that the finite size corrections to scaling are not under control for the system sizes considered and simulations on larger system sizes are therefore needed. A more recent numerical study argues for a quasi-long-range ordered spin glass phase in a system of diluted Ising dipoles [83].

3.3. Open questions regarding the spin glass physics in LiHo$_x$Y$_{1-x}$F$_4$

The single most important open question is the nature of the magnetic state in the limit of high dilution ($x < 0.1$). Is it a traditional spin glass, as suggested by the recent measurements of Quilliam et al. [79] or is it an unusual antiglass spin liquid [74, 75, 76, 77]? One may even ask whether there is a spin glass phase at all for $x \lesssim 0.25$ [51, 52, 53]. It would appear that the only way to resolve this issue is through more experiments since the computational effort to perform Monte Carlo simulations of large systems of magnetic moments interacting with long range $1/r^3$ dipolar interactions is rather prohibitive, and finite-size scaling corrections are significant [82]. The “sample quality” (e.g. impurity, homogeneous Y$^{3+}$ disorder, etc) may be of importance in this highly dilute limit and this issue should be examined carefully. Sample shape and demagnetization effects should not affect the existence of the spin glass phase, but this should nevertheless be investigated. There is a significant qualitative difference between the static and dynamic susceptibility measurements reported by three research groups (Refs. [51, 74, 75, 76, 77], Refs. [52, 53] and Refs. [79, 80]), and a new and independent series of measurements could help shed light on this outstanding question. It appears that the nonlinear susceptibility results of Ref. [52] are being dismissed [51, 79] on the basis of measurements that do not really probe the spin glass critical regime. However, disagreement on this interpretation remains [53]. The hole-burning and persistent oscillations observed by Ghosh et al. [76] are also indicative of a very unusual physical state, yet they have not been reported or reproduced by another group as far as we know. To reiterate, we believe that at this stage, in order for the field to move forward, more experiments that carefully explore the critical regime, notwithstanding the highly technical burden associated with such experiments, are very much needed.

On the numerical side, it would be of interest to go beyond the classical Monte Carlo simulations [26, 27, 82] that have so far been applied to the dilute system. The effects of the hyperfine interactions and off-diagonal interactions between the $\vec{\sigma}_i$ pseudospins are challenging to incorporate in a quantum Monte Carlo simulation since they lead to a sign problem. Nevertheless, it would be important to invest some effort in this problem in order to achieve more quantitative comparisons with experiments.
4. Conclusion
Due to the existence of a well-established two-state Ising model for the rare-earth compound LiHo$_x$Y$_{1-x}$F$_4$, this material constitutes a unique model magnet where theory, experiments and simulations can be compared and contrasted at a quantitative level. Despite the apparent simplicity of the underlying model for the parent compound, the introduction of disorder through dilution and quantum effects through a transverse magnetic field leads to a complex system which continues to challenge experimentalists and theorists alike. The two foremost outstanding questions are whether a thermodynamic spin glass phase survives in the limit of high dilution of the magnetic Ho$^{3+}$ ions and the reason for the unexpected stability of the ferromagnetic ground state of pure LiHoF$_4$ with respect to an applied transverse field. Continued research into this remarkable realization of the Ising model will hopefully continue to contribute pushing forward the forefront of research on such diverse topics as random fields, coherent oscillations and tunable quantum fluctuations.

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