

## Induced Random Fields in the $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ Quantum Ising Magnet in a Transverse Magnetic Field

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The  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  magnetic material in a transverse magnetic field  $B_x\hat{x}$  perpendicular to the Ising spin direction has long been used to study tunable quantum phase transitions in a random disordered system. We show that the  $B_x$ -induced magnetization along the  $\hat{x}$  direction, combined with the local random dilution-induced destruction of crystalline symmetries, generates, via the predominant dipolar interactions between  $\text{Ho}^{3+}$  ions, *random fields* along the Ising  $\hat{z}$  direction. This identifies  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  in  $B_x$  as a new random field Ising system. The random fields explain the rapid decrease of the critical temperature in the diluted ferromagnetic regime and the smearing of the nonlinear susceptibility at the spin-glass transition with increasing  $B_x$  and render the  $B_x$ -induced quantum criticality in  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  likely inaccessible.

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Quantum phase transitions (QPTs) occur near absolute zero temperature and are driven by quantum mechanical fluctuations associated with the Heisenberg uncertainty principle and not by thermal fluctuations as in classical phase transitions [1,2]. The transverse field Ising model (TFIM) [3,4] with Hamiltonian  $\mathcal{H}_{\text{TFIM}} = -\frac{1}{2}\sum_{(i,j)}J_{ij}\sigma_i^z\sigma_j^z - \Gamma\sum_i\sigma_i^x$ , where  $\sigma_i^\mu$  ( $\mu = x, y, z$ ) are Pauli matrices, is the simplest theoretical model that exhibits a QPT [1,4,5]. The field  $\Gamma$  transverse to the Ising  $\hat{z}$  direction causes quantum tunneling between the spin-up and spin-down eigenstates of  $\sigma_i^z$ . These spin fluctuations decrease the critical temperature  $T_c$  at which the spins develop either conventional long-range order or, for random ferromagnetic and antiferromagnetic  $J_{ij}$ , a spin-glass (SG) state with randomly frozen spins below  $T_g$ . At a critical field  $\Gamma_c$ ,  $T_c$  or  $T_g$  vanishes, and a quantum phase transition between either a long-range ordered or SG state and a quantum paramagnet (PM) ensues.

The phenomenology of both the disorder-free and the random TFIM has been extensively investigated in the  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  magnet with a magnetic field  $B_x$  applied transverse to the  $\text{Ho}^{3+}$  Ising spin direction [6–8], which is parallel to the  $c$  axis of the body-centered tetragonal structure of  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  [9]. Crystal field effects give an Ising ground state doublet  $|\Phi_0^\pm\rangle$  and a first excited singlet  $|\Phi_e\rangle$  at approximately 9 K above the ground doublet [9]. For  $x = 1$ ,  $\text{LiHoF}_4$  is a dipole-coupled ferromagnet (FM) with  $T_c = 1.53$  K [7,10]. Random disorder is generated by replacing the magnetic  $\text{Ho}^{3+}$  ions by nonmagnetic  $\text{Y}^{3+}$ . Quantum mechanical (spin flip) fluctuations are induced by  $B_x$  which admixes  $|\Phi_e\rangle$  with  $|\Phi_0^\pm\rangle$ , splitting the latter, hence producing an effective TFIM with  $\Gamma = \Gamma(B_x)$  ( $\Gamma \propto B_x^2$  for small  $B_x$ ) [10].

Two experimental puzzles pertaining to the effect of  $B_x$  on the FM ( $0.25 < x < 1.0$ ) and the SG ( $x < 0.25$ ) phases

of  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  have long been known. First, in the FM regime, while the mean-field argument that  $T_c(x) \propto x$  for the PM to FM transition is well satisfied for  $0.25 < x < 1.0$  [8], the rate at which  $T_c(B_x)$  is depressed by  $B_x$  becomes progressively faster than mean-field theory predicts as  $x$  is reduced [11]. This implies that, compared with the energy scale for FM order set by  $T_c(B_x = 0)$ ,  $B_x$  becomes ever more efficient at destroying FM order the lower  $x$  is [11]. Second, for  $B_x = 0$ ,  $\text{LiHo}_{0.167}\text{Y}_{0.833}\text{F}_4$  displays a conventional SG transition, with a nonlinear magnetic susceptibility  $\chi_3$  diverging at  $T_g$  as  $\chi_3(T) \propto (T - T_g)^{-\gamma}$  [12]. However,  $\chi_3(T)$  becomes less singular as  $B_x$  is increased from  $B_x = 0$ , with no indication that a QPT between the PM and SG states occurs as  $T \rightarrow 0$  [6,13]. It has recently been suggested that for dipole-coupled  $\text{Ho}^{3+}$  ions nonzero  $B_x$  generates both longitudinal (along the Ising  $\hat{z}$  direction) [14] and transverse [15,16] random fields (RFs) that either renormalize the critical transverse field [14,15] or even destroy the SG transition [16]. In this Letter, we examine the quantitative merit of this hypothesis by comparing results from numerical and analytical calculations with experimental results on  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ . We obtain compelling evidence that RFs are manifestly at play in  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  and explain the above two long-standing paradoxes.

We first show that the low-energy effective theory of  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  for  $x < 1$  and  $B_x > 0$  is a TFIM with additional  $B_x$ -induced RFs. We start with the Hamiltonian  $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{dip}}$  expressed in terms of the angular momentum operator  $\mathbf{J}$  of  $\text{Ho}^{3+}$  ( $J = 8, L = 6, S = 2$ ). The single ion part  $\mathcal{H}_0 = \sum_i[\mathcal{H}_{\text{cf}}(\mathbf{J}_i) + \mathcal{H}_Z(\mathbf{J}_i)]$  consists of the crystal field Hamiltonian  $\mathcal{H}_{\text{cf}}(\mathbf{J}_i)$  of  $\text{Ho}^{3+}$  in the  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  environment [9,10] and the Zeeman field term  $\mathcal{H}_Z = -g\mu_B(\mathbf{J}_i \cdot \mathbf{B})$ , with  $\mathbf{B}$  the magnetic field.  $g = 5/4$  is the  $\text{Ho}^{3+}$  Landé factor, and  $\mu_B$  is the

Bohr magneton. The interactions between ions are dominated by long-range magnetic dipolar interactions  $\mathcal{H}_{\text{dip}}$  [7,10,17],  $\mathcal{H}_{\text{dip}} = (g^2\mu_B^2/2)\sum_{(i,j)}\epsilon_i\epsilon_j[\mathbf{J}_i \cdot \mathbf{J}_j - 3(\mathbf{J}_i \cdot \mathbf{r}_{ij}\mathbf{J}_j \cdot \mathbf{r}_{ij})r_{ij}^{-2}]r_{ij}^{-3}$ , where  $\mathbf{r}_i$  are the crystalline positions occupied by either a magnetic  $\text{Ho}^{3+}$  ion ( $\epsilon_i = 1$ ) or a nonmagnetic  $\text{Y}^{3+}$  ion ( $\epsilon_i = 0$ ), and  $r_{ij} = |\mathbf{r}_j - \mathbf{r}_i|$  is the inter-ion distance.

The two lowest energy eigenstates  $|\Psi_i^+(B_x)\rangle$  and  $|\Psi_i^-(B_x)\rangle$  of  $\mathcal{H}_0$  are sufficiently below  $|\Phi_e\rangle$  that the latter can be ignored at temperatures near and below  $T_c(x=1) \sim 1.5$  K [10]. This allows us to recast  $\mathcal{H}$  in terms of an effective Hamiltonian  $\mathcal{H}_{\text{eff}}$  with  $S = 1/2$  pseudospin operators that act in the restricted low-energy subspace spanned by the  $\prod_i |\Psi_i^{\sigma_i}(B_x)\rangle$  ( $\sigma_i = \pm$ ) eigenstates of  $\mathcal{H}_0$ . In this subspace,  $\Gamma(B_x) \equiv (1/2)[\langle\Psi^+|\mathcal{H}_0|\Psi^+\rangle - \langle\Psi^-|\mathcal{H}_0|\Psi^-\rangle]$ . The projected  $J_i^\mu$  ( $\mu = x, y, z$ ) operator is written as:  $J_i^\mu = \sum_\nu C_{\mu\nu}(B_x)\sigma_i^\nu + C_{\mu 0}\mathbb{1}$  [10]. The  $|+\rangle$  and  $|-\rangle$  eigenstates of  $\sigma_i^z$  are written in terms of  $|\Psi_i^\pm\rangle$  such that  $J_i^z = C_{zz}\sigma_i^z$  [10]. For  $B_x = 0$ , only  $C_{zz} \neq 0$  and decreases slightly with increasing  $B_x$ , while the other  $C_{\mu\nu}(B_x)$  parameters and  $\Gamma(B_x)$  increase with  $B_x$ , starting from zero at  $B_x = 0$ . By straightforward manipulations replacing  $J_i^\mu$  in terms of  $\sum_\nu C_{\mu\nu}\sigma_i^\nu$  in  $\mathcal{H}_{\text{dip}}$ , one finds that the terms with largest  $C_{\mu\nu}(B_x)$  in  $\mathcal{H}_{\text{eff}}$ , including the transverse field term  $\Gamma\sigma_i^x$ , are

$$\begin{aligned} \mathcal{H}_{\text{eff}} = & \frac{(g\mu_B)^2}{2} C_{zz}^2 \sum_{(i,j)} \epsilon_i \epsilon_j L_{ij}^{zz} \sigma_i^z \sigma_j^z \\ & - \Gamma \sum_j \sigma_i^x + (g\mu_B)^2 C_{zz} \left\{ C_{x0} \sum_{(i,j)} \epsilon_i \epsilon_j L_{ij}^{xz} \sigma_i^z \sigma_j^z \right. \\ & \left. + C_{xx} \sum_{(i,j)} \epsilon_i \epsilon_j L_{ij}^{xz} \sigma_i^x \sigma_j^z \right\}, \end{aligned} \quad (1)$$

where  $L_{ij}^{ab} = [\delta_{ab} - 3r_{ij}^a r_{ij}^b / r_{ij}^2] r_{ij}^{-3}$ . We see from Eq. (1) that a longitudinal RF term  $\propto \sigma_i^z$  along  $\hat{z}$  and an off-diagonal (bilinear)  $\propto \sigma_i^x \sigma_j^z$  coupling are induced by  $B_x \neq 0$  ( $C_{x0} = C_{xx} = 0$  for  $B_x = 0$ ). For pure  $\text{LiHoF}_4$  ( $x = 1$ , all  $\epsilon_i = 1$ ), lattice symmetries enforce  $\sum_j L_{ij}^{xz} = 0$ , causing the term linear in  $\sigma_i^z$  and the Boltzmann thermal average  $\langle \sigma_i^x \sigma_j^z \rangle$  to vanish for  $B_x > 0$ . However, for  $x < 1$  and  $B_x > 0$ , a RF  $\propto \sigma_i^z$  emerges. With the time-reversal symmetry broken by  $B_x$  ( $\langle J_i^x \rangle > 0$ ), the bilinear  $\sigma_i^x \sigma_j^z$  also provides a ‘‘mean-field’’ contribution to the longitudinal RFs,  $\langle \sigma_i^x \rangle \sigma_j^z$ , as well as transverse RFs,  $\langle \sigma_i^z \rangle \sigma_j^x$ . We find that  $C_{xx}/C_{x0} \lesssim 1$  for all  $B_x > 0$  so that the leading correction to  $H_{\text{TFIM}}$  is indeed a (correlated) RF term,  $\sim \sum_i h_i^z \sigma_i^z$ , with  $h_i^z \propto C_{x0} C_{zz} \sum_j \epsilon_j L_{ij}^{xz}$ .

We now investigate the effect of the RFs on the PM to FM transition. To do so, we make a mean-field (MF) approximation to  $\mathcal{H}$  and consider the one-particle MF Hamiltonian  $\mathcal{H}_i^{\text{MF}}$  for an arbitrary site  $\mathbf{r}_i$  occupied by a  $\text{Ho}^{3+}$  moment:  $\mathcal{H}_i^{\text{MF}} = \mathcal{H}_{\text{cf}} - g\mu_B(\mathbf{h}_i^{\text{MF}} \cdot \mathbf{J}_i) - g\mu_B J_i^x B_x$ .  $\mathbf{h}_i^{\text{MF}}$  is the MF acting on magnetic moment  $\mathbf{J}_i$ ,

$\mathbf{h}_i^{\text{MF}} = g\mu_B \sum_j \epsilon_j [3(\mathbf{r}_{ij} \cdot \mathbf{M}_j) \mathbf{r}_{ij} / r_{ij}^2 - \mathbf{M}_j] r_{ij}^{-3}$ , with the self-consistent MF equation  $\mathbf{M}_i = \langle \mathbf{J}_i \rangle$ , with  $\mathbf{M}_i = \text{Tr}(\rho_{\text{MF}} \mathbf{J}_i) / \text{Tr}(\rho_{\text{MF}})$  and  $\rho_{\text{MF}} = \exp(-\beta H_i^{\text{MF}})$ , with  $\beta \equiv 1/(k_B T)$ . The crystal field Hamiltonian  $H_{\text{cf}}$  for  $\text{Ho}^{3+}$  expressed in terms of the components  $J_i^\mu$  is taken from the 4 crystal field parameter  $\mathcal{H}_{\text{cf}}$  of Ref. [9]. Slightly different choices of  $\mathcal{H}_{\text{cf}}$  [9,10] do not qualitatively affect the results. We diagonalize  $\mathcal{H}_i^{\text{MF}}$  for each  $i$  and update the  $\mathbf{M}_i$ 's using the newly obtained eigenstates. We continue iterating until convergence is reached at the  $n$ th iteration, defined by the convergence criterion  $\sum_i (\mathbf{M}_i^{(n)} - \mathbf{M}_i^{(n-1)})^2 / \sum_i (\mathbf{M}_i^{(n)})^2 \leq 10^{-6}$ . To simplify the calculations and speed up the convergence, we keep only the diagonal  $L_{ij}^{zz}$  and the off-diagonal  $L_{ij}^{xz}$  terms in  $\mathcal{H}_{\text{dip}}$ , since no qualitatively new physics arises when keeping the other diagonal ( $L_{ij}^{xx}$  and  $L_{ij}^{yy}$ ) and off-diagonal ( $L_{ij}^{xy}$ ,  $L_{ij}^{yz}$ ) terms in  $\mathcal{H}_{\text{dip}}$ . For a given  $B_x$ , we compute the temperature dependence of the magnetization  $M_z = [\sum_i M_i^z]_d$  along the  $\hat{z}$  direction [18]. Here  $[\dots]_d$  signifies an average over the bimodal lattice occupancy probability distribution  $P(\epsilon_i) = x\delta(\epsilon_i - 1) + (1-x)\delta(\epsilon_i)$ . The transition temperature  $T_c(B_x)$  is determined by the temperature at which  $M_z(T)$  sharply rises [18]. We consider a system of linear size  $L = 4$ , with the total number of sites  $N_0 = 4L^3$  with  $N = xN_0$  sites occupied by  $\text{Ho}^{3+}$ , and perform disorder averages over 50 different diluted samples. We use the Ewald summation method to define infinite-range dipolar interactions. For  $B_x = 0$ , we find that, as found experimentally, the decrease of  $T_c$  while reducing  $x$  is proportional to  $x$  [8,11]. To investigate the role of RFs, we compare  $T_c(B_x)$  obtained when both the  $L_{ij}^{xz}$  and  $L_{ij}^{zz}$  terms in  $\mathcal{H}_{\text{dip}}$  are kept (open symbols) with the  $T_c(B_x)$  found when only the  $L_{ij}^{zz}$  term is retained (solid symbols). Figure 1 shows that  $T_c(B_x)$  is depressed for small  $B_x$  faster when the off-diagonal  $L_{ij}^{xz}$  term in  $\mathcal{H}_{\text{dip}}$  is present than when only the

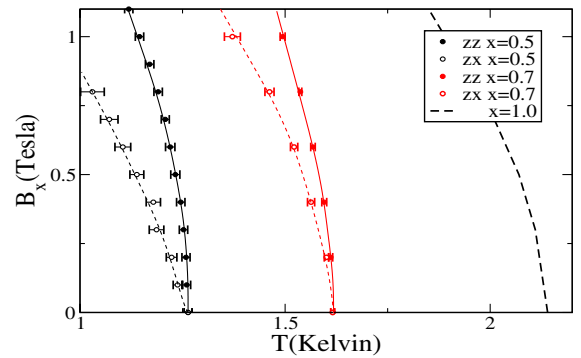


FIG. 1 (color online).  $T_c(B_x)$  from a finite lattice mean-field calculation (see text). For  $x = 0.7$  and  $x = 0.5$ , the solid symbols show  $T_c(B_x)$  when only the  $L_{ij}^{zz}$  dipolar terms that couple  $z$  components of  $\mathbf{J}_i$  are kept. The open symbols show the increased rate of depression of  $T_c(B_x)$  when the off-diagonal dipolar  $L_{ij}^{xz}$  couplings are included. For the pure  $x = 1$  case,  $T_c(B_x)$  is the same for both models since the lattice symmetries eliminate the internal local random fields along  $\hat{z}$ .

Using  $L_{ij}^{zz}$  term is kept. Also, as found experimentally [11], the rate at which  $T_c(B_x)$  is depressed by  $B_x$  increases as  $x$  is lowered. This provides strong evidence that the experimental observation is due to  $B_x$ -induced RFs whose variance increases as  $x$  decreases or  $B_x$  increases.

We now consider the role of RFs at the SG transition. While the nonlinear susceptibility  $\chi_3$  diverges at  $T_g \simeq 0.13$  K in  $\text{LiHo}_{0.167}\text{Y}_{0.833}\text{F}_4$  when  $B_x = 0$ ,  $\chi_3(B_x, T)$  becomes progressively smeared as  $B_x$  is turned on (see top inset in Fig. 3) [6,13]. It has been suggested that random off-diagonal dipolar couplings destroy the SG transition when  $B_x > 0$  [16]. Here we take a more pragmatic approach and ask whether the behavior of  $\chi_3(B_x > 0, T)$  in SG samples of  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  can indeed be interpreted in terms of induced RFs. To investigate this question on  $\chi_3(B_x, T)$ , we introduce a mean-field variant of  $\mathcal{H}_{\text{eff}}$  in Eq. (1) that preserves the crucial physics therein. Our model is a generalization of the Sherrington-Kirkpatrick transverse field Ising SG model [5,19–21] but with additional off-diagonal and RF interactions similar to that in  $\mathcal{H}_{\text{eff}}$ :

$$\tilde{\mathcal{H}} = \frac{1}{2} \sum_{(i,j)} J_{ij} \sigma_i^z \sigma_j^z + \frac{1}{2} \sum_{(i,j)} K_{ij} \sigma_i^x \sigma_j^z - \Gamma \sum_i \sigma_i^x - \sum_i h_i^z \sigma_i^z - h_0^z \sum_i \sigma_i^z. \quad (2)$$

For simplicity, we take infinite-ranged  $J_{ij}$  and  $K_{ij}$  given by independent Gaussian distributions of zero mean and variance  $J^2$  and  $K^2$ , respectively.  $h_i^z$  is a Gaussian RF with zero mean and variance  $\Delta^2$ . Model (2), but with  $K_{ij} = 0$ , was previously used to calculate  $\chi_3$  in quadrupolar glasses [19], which also possess internal RFs [19,22].

We follow the procedure of Ref. [21], employing the imaginary time formalism and the replica trick to derive the (replicated) free energy of the system. To further simplify the calculations, we make a static approximation for the replica-symmetric solution in the PM phase. This allows us to derive self-consistent equations for the  $\alpha$  components of the magnetization  $M_\alpha$  and spin-glass order parameters  $Q_\alpha$  ( $\alpha = x, z$ ). We find  $M_\alpha = (1/2\pi) \int_{-\infty}^{\infty} dx dz e^{-(x^2+z^2)/2} [(H_\alpha/H) \tanh(\beta H)]^p$ , with  $p = 1$ ,  $H_z = [h_0^z + z\sqrt{J^2 Q_z + (K^2/2)Q_x + \Delta^2}]$ ,  $H_x = [\Gamma + x\sqrt{(K^2/2)Q_z}]$ , and  $H^2 = H_x^2 + H_z^2$ . The self-consistent equations for  $Q_x$  and  $Q_z$  are obtained by replacing  $M_\alpha \rightarrow Q_\alpha$  above and setting  $p = 2$ .  $\chi_3$  is obtained from  $\chi_3 = \frac{1}{6} \partial^3 M_z / (\partial h_0^z)^3$  and by solving numerically the resulting four coupled self-consistent equations. Figures 2(a) and 2(b) show the  $\Gamma$  dependence of  $\chi_3$  for various temperatures  $T$  in models either with only  $h_i^z$  random fields [Fig. 2(a)] or with only random off-diagonal  $K_{ij}$  couplings [Fig. 2(b)].

Comparing Fig. 2 with the experimental  $\chi_3^{\text{expt}}(B_x)$  (top inset in Fig. 3), one finds that the dependence of  $\chi_3$  upon  $\Gamma$  in Fig. 2(a) does not show a decrease in magnitude as  $T$  is

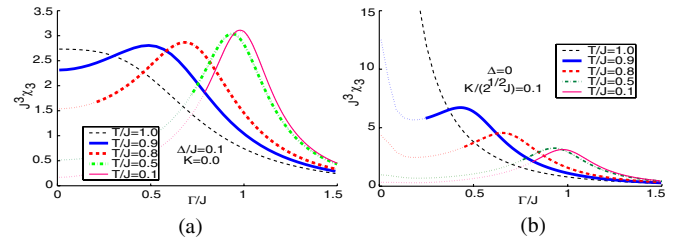


FIG. 2 (color online). (a)  $\chi_3$  vs  $\Gamma$  with  $K = 0$ . (b)  $\chi_3$  vs  $\Gamma$  with  $\Delta = 0$ . The change of line style at low  $\Gamma$  indicates the limit of stability of the replica-symmetric paramagnetic solution, as determined following the standard procedure [21].

decreased. Also, while  $\chi_3$  shows a decreasing amplitude with decreasing  $T$  in Fig. 2(b), it does not reveal a rapid sharpening as  $T$  is increased. It turns out that the key physics ingredient missing in these calculations is the underlying *microscopic* dependence of  $J$ ,  $\Delta$ , and  $K$  upon  $B_x$  via the  $C_{\mu\nu}(B_x)$  transformation coefficients. Physically, the built-in  $B_x$  dependence of the  $C_{\mu\nu}$  ensures that the  $B_x$  scale at which  $\chi_3$  is quenched by RFs is not trivially tied to the scale at which the  $T = 0$  SG-PM crossover occurs, as it is in  $\mathcal{H}$  with  $J_{ij}$ ,  $K_{ij}$ ,  $\Gamma$ , and  $h_i^z$  independent of  $B_x$  (cf. Fig. 2). The widths  $J(B_x)$ ,  $\Delta(B_x)$ , and  $K(B_x)$  are obtained by calculating the disorder average of the first, third, and fourth lattice sums in Eq. (1), respectively. We have  $K^2 = 4(g\mu_B)^4 [C_{zz}(B_x)C_{xx}(B_x)]^2 \times [(1/N_0) \sum_{(i,j)} \epsilon_i \epsilon_j (L_{ij}^{xz})^2]_d$  and  $\Delta^2 = (g\mu_B)^4 \times [C_{zz}(B_x)C_{x0}(B_x)]^2 [(1/N_0) \sum_i \epsilon_i (\sum_{j \neq i} \epsilon_j L_{ij}^{xz})^2]_d$ . To make further contact between calculations and experimental data, we note that hyperfine interactions, which are important in Ho-based materials [7,10], lead to a renormalization of the critical  $B_x$ ,  $B_x^c$ , when  $T_c$  or  $T_g$  is less than the hyperfine energy scale [10,15]. To obtain a relation be-

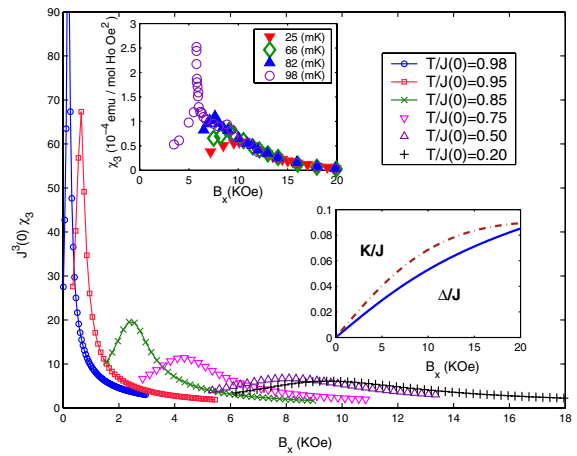


FIG. 3 (color online).  $\chi_3^{\text{theor}}$  vs  $B_x$  for different temperatures given by the model Eq. (2). The top inset shows  $\chi_3^{\text{expt}}$  from Ref. [6]. The parameters  $K$  and  $\Delta$  are computed using  $x = 0.167$  and rescaled by  $\eta = 0.15$  and are shown in the bottom inset (see text).

tween  $\Gamma$  and  $B_x$  in  $\tilde{\mathcal{H}}$  which does not incorporate hyperfine effects, we set  $\Gamma(B_x)/J(B_x) = 1.05(B_x/B_x^c)^{0.35}$ , where  $B_x^c = 1.2$  T is the experimental zero temperature critical field [13]. This ansatz for  $\Gamma(B_x)$  is obtained by matching the critical temperature  $T_g(B_x)$  of  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  with the  $T_g(\Gamma)$  of  $\tilde{\mathcal{H}}$ . For the former, we use  $T_g(B_x) = T_g(0) \times [1 - (B_x/B_x^c)^\phi]$  ( $\phi \approx 1.7$ ) as found experimentally [13]. For  $\tilde{\mathcal{H}}$ , we find  $T_g(\Gamma) \approx T_g(0)\{1 - a[\Gamma/T_g(0)]^\psi\}$  ( $a \approx 0.79$  and  $\psi \approx 4.82$ ) by fitting  $T_g(\Gamma)$  vs  $\Gamma$  with  $K = \Delta = 0$ . To incorporate the role of hyperfine effects on  $h_i^z$  and  $K_{ij}$ , we rescale  $K$  and  $\Delta$  calculated from their microscopic origin in Eq. (1) by a scale factor  $\eta = 0.15$  that positions the peak of  $\chi_3^{\text{theor}}(B_x)$  at  $B_x \sim 5$  kOe for  $T_g(B_x)/T_g(0) = 0.75$ . With an estimate of  $\eta$  available, one could then determine a parametric  $\Gamma(B_x)$  such that, for a given  $T/T_g(B_x = 0)$ , the experimental  $\chi_3^{\text{expt}}$  and the theoretical  $\chi_3^{\text{theor}}$  peak at the same  $B_x$  value for all  $T_g(B_x)/T_g(0)$ . Such a procedure gives results qualitatively very similar to those in Fig. 3.

$\chi_3^{\text{theor}}$  reproduces the overall trend of  $\chi_3^{\text{expt}}$ . This is evidence that RFs are indeed at play when  $B_x > 0$  and at the origin of the experimental  $\chi_3(B_x, T)$  behaviors in SG samples of  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  [6,13]. A noticeable difference between  $\chi_3^{\text{theor}}$  and  $\chi_3^{\text{expt}}$  is that, for fixed  $T/T_g(0)$ ,  $\chi_3^{\text{theor}}$  collapses more rapidly and at smaller  $B_x$  than  $\chi_3^{\text{expt}}$ . This is likely a further manifestation of the renormalization effects of the RFs and random off-diagonal couplings caused by the aforementioned hyperfine interactions. We will report elsewhere results addressing this hyperfine renormalization of  $\mathcal{H}_{\text{eff}}$  and its role on  $\chi_3(B_x, T)$ .

In conclusion, by comparing numerical and analytical results with experimental data, we have obtained compelling evidence that induced RFs are indeed at play and “observed” in  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ . As a result,  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  in a transverse field (TF) is identified as a new RF Ising system. As found in other RF systems, we expect that the nontrivial fixed point of the theory ( $\mathcal{H}_{\text{eff}}$ ) is controlled by a fluctuationless classical zero temperature fixed point. In particular, this is what occurs in a TFIM plus RFs ( $\tilde{\mathcal{H}}$  with  $K_{ij} = 0$ ) [23]. Hence, it would therefore seem that quantum criticality is most likely inaccessible in ferromagnetic  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  samples. For the Ising SG model with RFs along  $\hat{z}$ , recent studies suggest that there is no Almeida-Thouless line [12] and no thermodynamic SG transition [24]. Hence, from the arguments above leading to  $\mathcal{H}_{\text{eff}}$ ,  $B_x$ -induced SG to PM quantum criticality in  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  would also appear likely inexistent. The presence of  $B_x$ -induced RFs and the quenching of quantum criticality is presumably the reason why, unlike in quantum Monte Carlo simulations of TF Ising SG models [20], Griffiths-McCoy singularities [25] have not been reported in  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  [6,8]. While quantum criticality seems unlikely for any  $B_x > 0$  and  $x < 1$ , a new set of interesting questions has nevertheless arisen: Do FM samples of

$\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  with  $B_x > 0$  indeed display classical RF criticality and all the fascinating phenomena of the RF Ising model [26]? Given the scarcity of real RF Ising materials [26], the identification of another such system opens new avenues for future theoretical and experimental investigations.

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- [1] S. Sachdev, *Quantum Phase Transitions* (Cambridge University Press, Cambridge, England, 1999).
  - [2] S.L. Sondhi *et al.*, Rev. Mod. Phys. **69**, 315 (1997).
  - [3] P.G. de Gennes, Solid State Commun. **1**, 132 (1963).
  - [4] R.J. Elliott *et al.*, Phys. Rev. Lett. **25**, 443 (1970).
  - [5] B.K. Chakrabarti *et al.*, *Quantum Ising Phases and Transitions in Transverse Ising Models* (Springer-Verlag, Heidelberg, 1996).
  - [6] W. Wu *et al.*, Phys. Rev. Lett. **71**, 1919 (1993).
  - [7] D. Bitko *et al.*, Phys. Rev. Lett. **77**, 940 (1996).
  - [8] J. Brooke *et al.*, Science **284**, 779 (1999).
  - [9] P.E. Hansen *et al.*, Phys. Rev. B **12**, 5315 (1975).
  - [10] P.B. Chakraborty *et al.*, Phys. Rev. B **70**, 144411 (2004).
  - [11] J. Brooke, Ph.D. thesis, University of Chicago, 2000.
  - [12] J.A. Mydosh, *Spin Glasses: An Experimental Introduction* (Taylor & Francis, London, 1993).
  - [13] W. Wu, Ph.D. thesis, University of Chicago, 1992.
  - [14] <http://flux.aps.org/meetings/YR04/MAR04/baps/abs/S4250008.html>
  - [15] M. Schechter and P.C.E. Stamp, Phys. Rev. Lett. **95**, 267208 (2005).
  - [16] M. Schechter and N. Laflorencie, Phys. Rev. Lett. **97**, 137204 (2006).
  - [17] We ignore a small nearest-neighbor exchange interaction in  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  which does not play an important role in the physics that we are discussing here (see Ref. [10]).
  - [18] Because of finite-size effects, there is a residual net field along the  $\pm\hat{z}$  direction for any given realization of disorder that dictates the direction of  $M_z$  at low temperatures.
  - [19] T.K. Kopec *et al.*, Z. Phys. B **78**, 493 (1990).
  - [20] M. Guo *et al.*, Phys. Rev. B **54**, 3336 (1996).
  - [21] D.-H. Kim and J.-J. Kim, Phys. Rev. B **66**, 054432 (2002).
  - [22] P.C.W. Holdsworth *et al.*, J. Phys. Condens. Matter **3**, 6679 (1991).
  - [23] T. Senthil, Phys. Rev. B **57**, 8375 (1998).
  - [24] A.P. Young and H.G. Katzgraber, Phys. Rev. Lett. **93**, 207203 (2004).
  - [25] R.B. Griffiths, Phys. Rev. Lett. **23**, 17 (1969); B.M. McCoy, *ibid.* **23**, 383 (1969).
  - [26] D.P. Belanger, in *Spin Glasses and Random Fields*, edited by A.P. Young (World Scientific, Singapore, 1998).