

## LiHo<sub>x</sub>Y<sub>1-x</sub>F<sub>4</sub> as a random-field Ising ferromagnet

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(Received 2 November 2006; revised manuscript received 29 October 2007; published 7 January 2008)

As a result of the interplay between the intrinsic off-diagonal terms of the dipolar interaction and an applied transverse field  $H_t$ , the diluted LiHo<sub>x</sub>Y<sub>1-x</sub>F<sub>4</sub> system at  $x > 0.5$  is equivalent to a ferromagnet in a longitudinal random field (RF). At low  $H_t$  the quantum fluctuations between the Ising-like doublet states are negligible, while the effective induced RF is appreciable. This results in a practically exact equivalence to the classical RF Ising model. By tuning  $H_t$ , the applied longitudinal field, and the dilution, the Ising model can be realized in the presence of an effective RF, transverse field, and constant longitudinal field, all independently controlled. The experimental consequences for  $D=1,2,3$  dimensions are discussed.

DOI: 10.1103/PhysRevB.77.020401

PACS number(s): 75.10.Nr, 75.10.Hk, 75.10.Jm

Since its seminal discussion by Imry and Ma,<sup>1</sup> the Ising model in a random longitudinal magnetic field was found to have many interesting realizations in nature and has been a subject of intensive theoretical and experimental investigation for both short-range and dipolar ferromagnetic (FM) interactions (see Refs. 2–5 and references therein). In particular, experiments are mostly done on diluted antiferromagnets (DAFMs),<sup>6</sup> which were shown to be equivalent in their static critical behavior to the random-field Ising model (RFIM).<sup>6,7</sup> In the DAFMs the order parameter is given by the staggered magnetization and a constant applied longitudinal field plays the role of a tunable random field (RF). However, the straightforward description of the RFIM in a FM system has not found an experimental realization, due to the difficulty of applying a local random magnetic field.

Interestingly, the diluted LiHo<sub>x</sub>Y<sub>1-x</sub>F<sub>4</sub> system provides the possibility of producing effective local magnetic fields in the longitudinal direction, due to the interplay of the off-diagonal terms of the dipolar interactions and an applied constant magnetic field  $H_t$  in the transverse direction.<sup>8–11</sup> Importantly, as is further explained below, due to the strong hyperfine interactions in this system, for  $H_t < 0.5$  T the applied transverse field does generate an effective longitudinal RF due to fluctuations between each Ising ground-state (GS) doublet and its corresponding excited state,<sup>10,12</sup> but the coupling between the two Ising doublet states is negligible.<sup>9</sup> Furthermore, despite the presence of some correlations between the RFs and their origin in the long-range dipolar interaction, we show below that on length scales larger than the intervacancy distance the behavior of the system is equivalent to that of the uncorrelated RF model. Therefore, despite the presence of a finite  $H_t$  and finite correlations on short scales, LiHo<sub>x</sub>Y<sub>1-x</sub>F<sub>4</sub> at  $H_t < 0.5$  T is a perfect realization of the classical RFIM with a tunable random magnetic field.

As  $H_t$  is enhanced, the effective transverse field becomes appreciable. Thus, the Ising model Hamiltonian

$$H = \sum_{ij} J_{ij} \tau_i^z \tau_j^z + \sum_i \gamma_i \tau_i^z + \Delta \sum_i \tau_i^x + h_{\parallel} \sum_i \tau_i^z \quad (1)$$

can be realized in the LiHo<sub>x</sub>Y<sub>1-x</sub>F<sub>4</sub> system with an effective local RF term  $\gamma_i$ , an effective transverse (quantum) term  $\Delta$ , and an effective constant field in the longitudinal direction

$h_{\parallel}$ . In the regime considered in this Rapid Communication, where  $x > 0.5$ , the system is FM at low  $T, H_t$ ,<sup>13,14</sup> and  $J_{ij}$  correspond to the long-range dipolar interaction. In comparison to the DAFM systems, the LiHo<sub>x</sub>Y<sub>1-x</sub>F<sub>4</sub> system is different, since in the former the effective interaction is short range, and advantageous, since in the DAFM system one can realize neither an effective longitudinal field in the staggered magnetization nor an appreciable quantum term. Importantly, the three effective fields  $\Delta, h_{\parallel}$ , and  $\gamma \equiv \sqrt{\text{Var}(\gamma_i)}$  can be independently controlled by tuning  $H_t$ , the applied magnetic field in the longitudinal direction  $H_z$ , and the dilution  $x$ . Specifically, for  $H_t \ll 1$  T and appreciable dilution  $\gamma \gg \Delta$ , where  $\Delta \gg \gamma$  for  $H_t > 1.5$  T and  $\bar{x} \equiv 1-x \ll 1$ .

This gives an opportunity to study experimentally the RFIM in a FM system, in both the classical and quantum regimes. In particular, the realization of the Hamiltonian (1) in a FM system allows both the study of long-standing questions using direct bulk probes such as magnetization and susceptibility, and an experimental study of models in which in addition to the random field a constant longitudinal field or a quantum term plays a role. Thus, the present work could be a basis for numerous experimental possibilities, a few of which are the following: (i) The thermal and quantum phase transitions (PTs) of the RFIM at  $D=3$  can be studied in the whole  $T, H_t$  phase diagram (see Fig. 1). Experiments studying the classical phase transition in three-dimensional (3D)

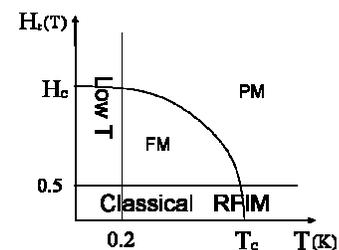


FIG. 1. Schematic picture of the phase diagram at some dilution  $x < 1$ . At  $H_t < 0.5$  T the quantum fluctuations between the Ising doublet states are negligible, while the effective random field is already appreciable. In this regime the system is equivalent to the classical RFIM. At  $T \ll 0.2$  K only the electronuclear ground states are occupied and the quantum phase transition in the presence of the random effective field can be studied.

DAFM systems show that the RF prevents the sample from reaching equilibrium on experimentally accessible time scales.<sup>15,16</sup> The study of the phase transition in diluted dipolar magnets can shed light on this interesting phenomenon, and the effect of the dipolar interaction on the theoretical predictions for the equivalence of the quantum and classical PTs in the RFIM<sup>17,18</sup> can be tested as well.<sup>19</sup> Of particular interest is the ability, resulting from the quantum term, to control the relaxation rate of the system by tuning the effective transverse field. (ii) As a result of the possibility to apply tunable random field and constant field in the longitudinal direction, one could study experimentally the hysteresis in the RFIM as function of  $H_z$ . Theoretical studies of this model in the absence<sup>20</sup> and presence<sup>21</sup> of long-range interactions have shown an intriguing disorder-driven out-of-equilibrium PT, a paradigm for the study of crackling noise in self-organized critical systems. (iii) In  $D=1,2$  dimensions, the predictions of Imry and Ma<sup>1</sup> and Binder<sup>22</sup> for the destruction of long-range FM order<sup>23</sup> and the rounding of the phase transition in 2D can be verified by studying the magnetization as function of the  $H_t$ -dependent effective RF.

All our analysis and results below can be easily generalized to any dipolar Ising magnet, as is explained in detail in Ref. 10. In the following we concentrate on the  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  compound since this enables us to present quantitative results for this system which is of prime experimental interest.

*Realization of the RFIM in the  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ .* Anisotropic dipolar magnets in general, and the  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  compound in particular, are considered to be realizations of the Ising model. As a result of the crystal-field anisotropy the GS is an Ising-like doublet, and all but the longitudinal dipolar interactions are effectively reduced. The application of  $H_t$  induces quantum fluctuations (QFs) between the Ising GSs, resulting in an effective transverse-field Ising model (TFIM). For the undiluted system both the thermal and quantum PT to the PM state were observed.<sup>24</sup> The dominant interaction in the  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  system is dipolar,<sup>25</sup>  $H_{\text{dip}} = \sum_{ij,\alpha\beta} V_{ij}^{\alpha\beta} J_i^\alpha J_j^\beta$ . In pure  $\text{LiHoF}_4$ , the off-diagonal terms of the dipolar interaction—e.g.,  $J_i^x J_j^y$ —are not only effectively reduced, but cancel by symmetry. However, at finite dilution  $x < 1$ , this cancellation does not hold. Furthermore, in Ref. 10 it was shown that in the presence of a constant  $H_t$ , the off-diagonal terms, despite being effectively reduced, are significant since their presence changes the symmetry of the system. The system is no longer symmetric under the transformation  $S_z \rightarrow -S_z$ , and an effective local longitudinal magnetic RF  $\tilde{h}_j$  is generated. In the regime  $H_t \ll \Omega_0 / (\mu_B S)$ ,

$$\tilde{h}_j = \frac{2SH_t}{\Omega_0} \sum_i V_{ji}^{zx}, \quad (2)$$

see Ref. 10. The random local energy is given by  $\tilde{\gamma}_j = \mu_B \tilde{h}_j S$ .  $\Omega_0 \approx 10$  K is the measure of the anisotropy, given by the excitation energy between each of the Ising GSs to its relevant excited state,<sup>10</sup> and  $S \approx 5.5$  is the magnitude of the effective Ho spin.

Importantly,  $\tilde{h}_j$ , Eq. (2), is independent of the directions of the spins at sites  $i$  and therefore equivalent for the spin-glass (SG) and FM regimes. However, the energy gain due to the

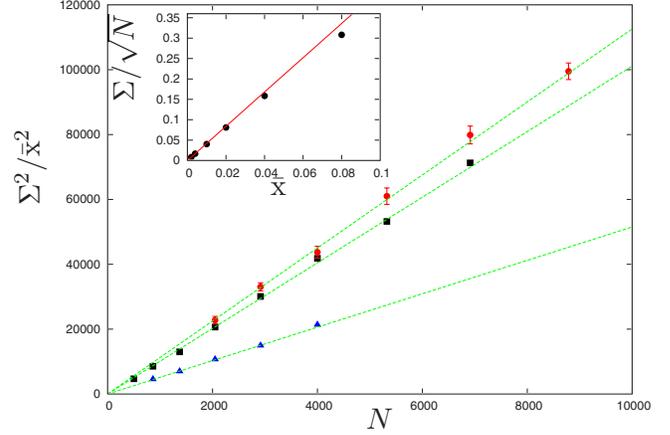


FIG. 2. (Color online)  $\Sigma^2 / \bar{x}^2 \equiv (E_{IM} / cV_0 \bar{x})^2$  in three dimensions is plotted as function of  $N=4L^3$  for dilutions  $x=0.6$  (blue triangles),  $0.96$  (black squares), and  $0.996$  (red circles).  $L$  is the length of the cube in unit cells. Averaging is done over  $10^4$  samples for each data point. Where not drawn, error bars are smaller than the point size. Inset:  $|\Sigma| / \sqrt{N}$  as function of  $\bar{x}$  for  $N=4000$ .  $|\Sigma| = \eta_0 \bar{x} \sqrt{N}$  for  $\bar{x} \ll 1$ , with  $\eta_0 = 4.12$ .

effective RF does depend on the spin configuration. Thus, while the randomness in the energy gain in the SG regime is a result of both the random position and the random orientation of the spins, in the FM regime only the former are random. Furthermore, the RFs  $\tilde{h}_j$  are not uncorrelated. For  $\bar{x} \ll 1$  the correlations of  $\tilde{h}_j$  at distances smaller than the typical intervacancy distance are large. However, as is shown explicitly below, for all dilutions, for distances larger than the intervacancy distance, the typical energy gain in flipping a domain  $E_{IM} = \sum_j \tilde{\gamma}_j$  obeys the Imry-Ma behavior. In particular  $\text{Var}(E_{IM}) \propto N$ , where  $N$  is the number of sites in a domain.

Let us now calculate explicitly  $\Sigma^2 \equiv (\sum_{ij} V_{ij}^{zx} / V_0)^2 = (1/cV_0)^2 E_{IM}^2$ , where  $c \equiv 2\mu_B H_t S^2 / \Omega_0$  and  $V_0$  is the magnitude of the dipolar interaction at distance of one unit cell along the  $x$  direction. First, note that for a single vacancy  $\bar{i}$ , as a consequence of lattice symmetry,  $\sum_j V_{ij}^{zx} = 0$ . This results in  $\sum_{ij} V_{ij}^{zx} = \sum_{\bar{i}\bar{j}} V_{\bar{i}\bar{j}}^{zx}$ , where  $i, j$  run over occupied sites and  $\bar{i}, \bar{j}$  over the unoccupied sites. Consider first the case of  $\bar{x} \ll 1$ . Assuming that the interactions causing the RF,  $V_{ij}^{zx}$ , are nearest neighbor only, then in leading order  $E_{IM}^2$  is proportional to the number of nearest-neighbor vacancy pairs and therefore to  $\bar{x}^2$ . In analogy to the case of a dipolar interaction in a random system<sup>26</sup> it is expected that *for the random interaction leading to the effective RF* the dipolar and short-range interactions would be equivalent. Thus, we expect  $\text{Var}(E_{IM}) \propto N$  for all dilutions and  $\text{Var}(E_{IM}) \propto \bar{x}^2 N$  for  $\bar{x} \ll 1$ , where correlations of three or more vacant spins can be neglected.

In Fig. 2 we plot  $\text{Var}(\Sigma) / \bar{x}^2$  for cubes in 3D as function of  $N=4L^3$  for  $x=0.6, 0.96, 0.996$ . Here and below lengths are given in terms of lattice unit cells. The dilutions were chosen to include the cases where  $\bar{x} \ll 1$ , and effective fields are strongly correlated on small distances,  $x=0.6$ , where correlations due to the relative positions of three or more vacancies are significant, and  $x=0.96$  in between. For each  $x$  we

obtain a good linear fit  $\text{Var}(\Sigma)/\bar{x}^2 = [\eta(x)]^2 N$ , which corresponds to  $\text{Var}(E_{IM}) = [\eta(x)c\bar{x}V_0]^2 N$ . In the inset we plot  $\sqrt{\text{Var}(\Sigma)}/N$  as a function of  $\bar{x}$  for a 3D cube with  $N=4000$ , showing  $\lim_{\bar{x} \rightarrow 0} \eta = \eta_0$  with  $\eta_0 = 4.12$ .

We therefore conclude that for distances larger than the intervacancy distance the system is equivalent to a RFIM, with an effective *uncorrelated* RF

$$\gamma_j = \eta_j \frac{2\mu_B H_t S^2}{\Omega_0} V_0 \bar{x}, \quad (3)$$

$\eta_j$  being a random number with  $\langle \eta_j \rangle = 0$  and  $\text{Var}(\eta_j) = \eta^2/x$ . Similar results (not shown) were obtained for  $D=1$ . Interestingly,  $\gamma_j$  in Eq. (3) is similar to the RF obtained in the SG regime,<sup>10</sup> with  $\bar{x}$  replacing  $x$  and the constant prefactor being different for the two regimes due to the different spin configuration.

While our results are general to any dipolar Ising magnet, we now use the specifics of the LiHo<sub>x</sub>Y<sub>1-x</sub>F<sub>4</sub> system to show that despite the presence of  $H_t$  the analogy to the classical RFIM is possible. The Ising-like anisotropy is dictated by a crystal-field Hamiltonian having large  $J_z^2$  and  $J_z^4$  terms.<sup>25</sup> In addition, a large  $(J_+^4 + J_-^4)$  term results in a mixing of free-ion states with  $\Delta J_z = \pm 4$ . The electronic GSs are an Ising-like doublet, denoted  $|\uparrow\rangle, |\downarrow\rangle$ , belonging to the  $J_z = (\pm 7, \pm 3, \mp 1, \mp 5)$  multiplets. Although the full splitting of the  $2J+1=17$  states is  $\Omega \approx 600$  K, the first excited state  $|\Gamma\rangle$ , belonging to the  $J_z = (6, 2, -2, -6)$  multiplet, is only  $\Omega_0 = 10$  K higher than the GS doublet. The electronic Hamiltonian allows a coupling between the electronic GSs which is second order in  $H_t$ . However, the low-energy physics of the LiHo<sub>x</sub>Y<sub>1-x</sub>F<sub>4</sub> system is strongly affected by the strong hyperfine (hf) interaction between the Ho electronic angular momentum and its nuclear spin.<sup>9,27</sup> In particular, each electronic GS is split to  $2I+1=8$  nearly equidistant states with  $I_z = -7/2, \dots, 7/2$  and a separation of 205 mK between them.<sup>28</sup> Thus, the relevant Ising-like single-spin GS doublet is of electronuclear type, having a definite and opposite nuclear spin states: i.e.,  $a \equiv |\uparrow, -7/2\rangle$  and  $\bar{a} \equiv |\downarrow, 7/2\rangle$ . The tunneling between the states  $a$  and  $\bar{a}$  requires the flipping of the nuclear spin, resulting in a very weak coupling at low fields [less than  $10^{-4}$  K at  $H_t < 1$  T (Ref. 27)]. Only when  $|\Gamma\rangle$  becomes well hybridized with the electronic GSs  $|\uparrow\rangle, |\downarrow\rangle$ , will QFs between the states  $a$  and  $\bar{a}$  become appreciable. This occurs at  $H_t \approx \Omega_0/(\mu_B S) \approx 1.5$  T [see a detailed plot of  $\Delta(H_t)$  in Fig. 2 of Ref. 9].

We first consider the low- $T$  low- $H_t$  (LTLH) regime ( $H_t < 1$  T and  $T \ll 0.2$  K). In this regime only the states  $a$  and  $\bar{a}$  are appreciably occupied and the quantum tunneling between these two states is negligible. Thus, the system is effectively a classical Ising model.<sup>9</sup> Importantly, the effective RF (3), originating from fluctuations between each Ising GS and its corresponding excited state,<sup>10,12</sup> can already be made appreciable, since its behavior is linear in  $H_t$ . Thus, in this regime, which is of particular interest for the study of the destruction of long-range order (LRO) by the effective RF in  $D=1,2$  (see below), and the equilibrium and nonequilibrium phenomena in the ordered state in  $D=3$ , the analogy between the LiHo<sub>x</sub>Y<sub>1-x</sub>F<sub>4</sub> system and the RFIM is practically exact.

*Phase transitions in three dimensions.* As a result of the very different dependence of  $\gamma$  and  $\Delta$  on  $H_t$ , the thermal and quantum PTs can be well separated, as is sketched in Fig. 1. Both phase transitions occur outside the LTLH regime. For the classical PT one has to go to  $T > 0.2$  K, since  $T_c \approx x \times 1.54$  K.<sup>13</sup> Therefore, all nuclear states are appreciably occupied near the transition. Still, at low enough transverse field  $H_t \approx 0.5$  T, QFs are small between any of the time reversed states  $|\uparrow, -I_z\rangle, |\downarrow, I_z\rangle$ .<sup>27</sup> Thus, the nuclear spin states can be traced over and the classical PT is equivalent to that of the RFIM. Note that the magnitude of the RF can be tuned by varying the dilution and for each given sample by changing the magnitude of  $H_t$ , as can be inferred from Eq. (3).

Considering the quantum phase transition (QPT) we focus on  $T \ll 0.2$  K, where only the GS doublet is appreciably occupied. However, we still have to go out of the LTLH regime since strong QFs are required. Specifically, in the dilutions of interest the QPT occurs at  $H_t > 3$  T. In Ref. 9 it was shown that QFs between the electronuclear Ising states become appreciable at  $H_t \approx 1.5$  T, and at  $H_t \approx 3$  T the GS has all hf levels well mixed.<sup>27</sup> However, since at  $x > 0.5$  the typical magnitude of the dipolar energy  $V_0 x S^2 \gg 0.2$  K, it is only when the QFs of the *electronic* spins are larger than the interaction that the PT to the PM phase takes place. At  $x=1$  it was argued that the system is equivalent to the TFIM, with the hf interactions effectively renormalizing the transition field.<sup>25</sup> For  $x < 1$  and  $V_0 x S^2 \gg 0.2$  K we expect the same picture to be valid, with the addition of the effective longitudinal RF.

At the QPT  $H_t \approx \Omega_0/(\mu_B S)$ , Eq. (2) is not valid and QFs between the Ising doublet states enhance the effective RF. The largest RFs that can be obtained are of the order of  $V_0 S/\mu_B$ . By changing the dilution  $x$ , one can therefore control the magnitude of the typical random magnetic field  $h$  at the QPT in the regime  $0 < h \lesssim V_0 S/\mu_B$ , equivalent to  $0 < \gamma \lesssim J$ , where  $J$  is the average nearest-neighbor interaction. Thus, for  $\bar{x} \ll 1$  the QPT can be studied in the presence of weak RF, while for a substantial dilution the RF, as well as the fluctuations, are of the order of the interaction. Note that LRO is destroyed when either QF or the RF are of the order of the interaction, and the situation where all three energy scales are of the same order is of both theoretical and experimental interest.

*Measuring the correlation length in one and two dimensions.* Two is the critical dimension for the stability of the FM phase to the RF,<sup>1,2</sup> as the energy gain and energy cost to flip a domain are both linear in the domain size. However, Binder<sup>22</sup> has shown that due to the roughening of the domain wall, a two-dimensional FM Ising system in a RF has a finite correlation length  $\xi^{2D} \propto \exp(V_0 S^2/\gamma^2)$ . Here we consider first a strictly 2D system, where the plane includes the easy  $z$  axis and is chosen as to have a FM phase at zero transverse field. We first consider an in-plane magnetic field  $H_x$  transverse to the easy axis. For  $H_x \ll \Omega_0/(\mu_B S)$  the effective RF is given by Eq. (3), and therefore, for an appreciable dilution, say,  $0.5 < x < 0.8$ ,  $\xi^{2D} \propto \exp[(\Omega_0/\mu_B H_x)^2]$ . Since  $S \gg 1$ ,  $\xi^{2D} > \exp(S^2)$  in the perturbative regime. As  $H_x$  is increased, QFs between the Ising-like doublet become significant,  $h$  increases, and  $\xi$  decreases. Assuming one can reach the regime

where  $\xi^{2D} \ll L$ , the finite correlation length can be detected by studying  $M$  as function of  $H_x$ ,

$$M \approx M_0(H_x) \frac{\xi^2}{L^2} \sqrt{\frac{L^2}{\xi^2}} \approx M_0(H_x) \frac{\xi}{L}, \quad (4)$$

as the ratio  $M/M_0$  depends linearly on  $\xi$ . Here  $M_0(H_x)$  corresponds to the dependence of the magnetization on  $H_x$  in a FM domain due to single-spin fluctuations, as can be measured in a 3D sample where the system stays FM for the relevant magnetic fields [ $H_t \ll \Omega_0/(\mu_B S)$ ].

Unlike systems such as the DAFM  $\text{RB}_2\text{Co}_x\text{Mg}_{1-x}\text{F}_4$ ,<sup>29</sup> where the two dimensionality is due to the inherent layered structure, for the  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  a realistic system would be quasi-two-dimensional, with a finite width  $d$ . The energy cost to flip a domain is then linear in  $d$ , while the energy gain is  $\propto \sqrt{d}$ . As a result  $\xi$  is enhanced,  $\xi^{2D} \propto \exp[\Omega_0^2 d / (\mu_B H_x)^2]$ , and therefore one has to keep  $d$  small in order to observe a finite  $\xi$ . In comparison to layered systems, the observation of a finite  $\xi$  is more difficult. However, the study of the gradual crossover between two and three dimensions is made possible. Note that the degree of two dimensionality can be studied by measuring the dependence of the RF on the angle of  $H_t$  in the  $xy$  plane. This also allows the continuous change of the RF in a given sample. In particular, for the strict 2D case there is no RF when  $H_t$  is perpendicular to the plane.

Using simple Imry-Ma<sup>1</sup> arguments one can show that for a quasi-1D system parallel to the  $z$  axis and of cross section  $A$ , the finite correlation length is given by  $\xi^{1D} \approx A(\Omega_0/\mu_B H_t)^2$  and the magnetization by

$$M \approx M_0(H_t) \frac{\xi}{L} \sqrt{\frac{L}{\xi}} \approx M_0(H_t) \sqrt{\frac{A}{L}} \frac{\Omega_0}{\mu_B H_t}. \quad (5)$$

Thus, the observation of  $M/M_0 \propto 1/H_t$  in the regime  $\Omega_0 \sqrt{A/L} \ll \mu_B H_t \ll \Omega_0/S$  is the experimental manifestation of the instability of the FM phase in the one-dimensional Ising system to RF, as predicted by Imry and Ma.

As explained in detail in Refs. 10 and 12, our analysis above applies directly to a general anisotropic dipolar system, with a crystal-field Hamiltonian, e.g., of the form  $H_{CF} = DS_z^2$ , and with no hyperfine interactions. Interestingly, in the  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  system, if one neglects the hyperfine interactions, the peculiar form of the crystal-field Hamiltonian which allows transitions between the two electronic ground states in second-order perturbation, leads to different physical results. Most importantly, the effective transverse term becomes appreciable at small  $H_t$  and the RFIM is obtained at the expense of significant quantum fluctuations. Thus, the proper consideration of the hyperfine interactions in the  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  system is not only crucial for obtaining the correct effective fields, but also to obtain the qualitative equivalence of the  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$  system to general diluted anisotropic dipolar magnets.

It is a pleasure to thank Gabriel Aeppli, Amnon Aharony, David Belanger, Nicolas Laflorencie, and Alessandro Silva for useful discussions. This work was supported by NSERC of Canada and PITP.

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