





Effect of intrinsic quantum fluctuations on the phase diagram of anisotropic dipolar magnets

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The rare-earth material LiHoF_4 is believed to be an experimental realization of the celebrated (dipolar) Ising model and, upon the inclusion of a transverse field B_x , an archetypal quantum Ising model. Moreover, by substituting the magnetic Ho ions by nonmagnetic Y ions, disorder can be introduced into the system, giving rise to a dipolar disordered magnet and at high disorders to a spin glass. Indeed, this material has been scrutinized experimentally, numerically, and theoretically over many decades with the aim of understanding various collective magnetic phenomena. One of the to-date open questions is the discrepancy between the experimental and theoretical $B_x - T$ phase diagram at low fields and high temperatures. Here we propose a mechanism, backed by numerical results, that highlights the importance of quantum fluctuations induced by the off-diagonal dipolar terms, in determining the critical temperature of anisotropic dipolar magnets in the presence and in the absence of a transverse field. We thus show that the description as a simple Ising system is insufficient to quantitatively describe the full phase diagram of LiHoF_4 , for the pure as well as for the dilute system.

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Introduction. Anisotropic dipolar magnets, realized in both single molecule magnets and rare-earth magnetic insulators, are at the forefront of quantum research. The large anisotropy barrier allows their use as nanomagnets, with possible applications in the operation of qubits and memory bits at reduced sizes [1–3]. In lattice form, anisotropic dipolar magnets typically have very small exchange interactions, allowing for efficient induction of quantum fluctuations by applied transverse fields. Thus, anisotropic dipolar magnets are perceived as experimental models for the transverse field Ising model. These unique characteristics motivated intense study of quantum phenomena in these materials, including quantum phase transitions [4–7], quantum annealing [6,8,9], domain wall dynamics [10,11], and high- Q nonlinear dynamics [12].

One of the most studied anisotropic dipolar magnets is LiHoF_4 [5,8,9,13]. Below the Curie temperature of $T_c = 1.53$ K LiHoF_4 orders ferromagnetically due to the dipolar interaction between Ho^{3+} ions combined with its lattice structure [14]. By the inclusion of an external transverse field the transition temperature is suppressed, until eventually it is converted to a quantum phase transition at $B_x \approx 4.9$ T [4]. Additionally, disorder can be introduced by randomly substituting some of the magnetic Ho^{3+} ions with nonmagnetic Y^{3+} ions, resulting in $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$, which presents a rich phase diagram—including a spin-glass phase at low concentrations ($x \lesssim 0.25$ – 0.3) [15–17]—the result of the interplay of interactions, disorder, and quantum fluctuations [18–24].

The $B_x - T$ phase diagram of LiHoF_4 is indeed in qualitative agreement with that of the transverse field Ising model, but a quantitatively correct description has proven enduringly elusive, specifically at the high-temperature, low-field regime,

where thermal, rather than quantum, fluctuations are dominant [23,25,26].

The significance of off-diagonal terms of the dipolar interaction is well appreciated in the presence of disorder and a transverse field, as they break \mathbb{Z}_2 symmetry and transform spatial disorder to an effective random longitudinal field, making this material one of the few magnetic realizations of the random field Ising model [19–23]. In this Letter we establish the importance of off-diagonal dipolar (ODD) interaction terms to the quantitative description of the phase diagram of $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ at $0.4 < x \leq 1$, which may thus provide insights to open questions in the field. We show that even for the pure system, and in the absence of a transverse field, the classical Ising model, which does not take these terms into account, provides an insufficient description of the system. The reason for this is that ODD interactions give rise to quantum fluctuations which markedly affect the phase diagram. These fluctuations are induced when ODD terms exert internal transverse fields that lower the energy of the Ho^{3+} ions on which they are exerted. We argue such fields are more prevalent in the paramagnetic (PM) phase than in the ferromagnetic (FM) phase, thus favoring the former.

Results from previous studies, using various Monte Carlo (MC) techniques [25,27] and mean-field analyses [4,28], show a persistent discrepancy with experimental results for the $B_x - T$ phase diagram [4,23,26,29]. Namely, when the theoretical results are fitted to the experimental results, either the zero-field critical temperature or the low-temperature–high-field regime can be made compatible with experiment, but not both. If the former is chosen, then even at small fields the $T_c(B_x)$ dependence is not theoretically well reproduced, and, if the latter, then the critical temperatures at low and intermediate transverse fields are significantly overestimated.

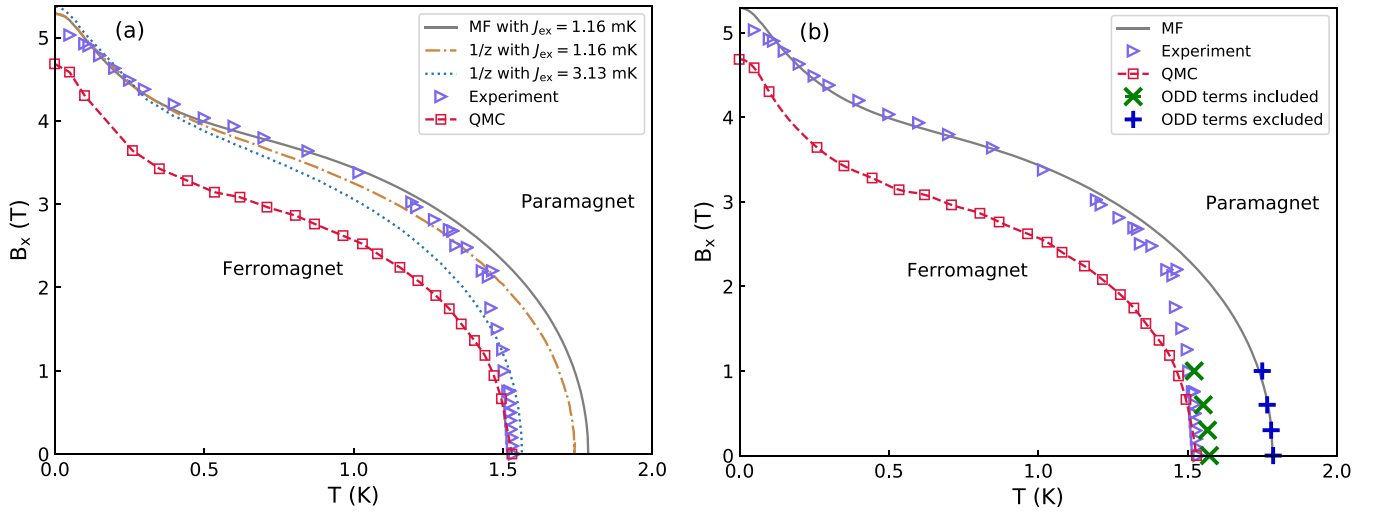


FIG. 1. The full phase diagram of LiHoF₄ as a function of temperature and applied transverse field. (a) A compilation of previous numerical works. Open squares are quantum Monte Carlo (QMC) results [27]. The dotted line is $1/z$ calculation with $J_{\text{ex}} = 3.13$ mK and the dot-dashed line is the same calculation with $J_{\text{ex}} = 1.16$ mK [28]. The solid line is a mean-field calculation [29] which uses the latter exchange value. Triangles represent results from several different experiments [4,26,29]. An apparent tradeoff is observed between theoretical predictions that match the experimental results at low temperatures but completely fail at the low-field regime, and ones that give correct zero-field T_c but fail to predict the correct $T_c(B_x)$ dependence and give a poor match at the intermediate B_x region. (b) Results of this Letter overlaid on top of previous theoretical and experimental results. The green Xs are the numerical results of this Letter with off-diagonal dipolar terms included and blue plus signs are for numerical results where they are excluded. Both use $J_{\text{ex}} = 1.16$ mK.

The fitting is done by tuning the single free parameter representing the nearest-neighbor antiferromagnetic interaction strength, where higher values correspond to lower critical temperatures. This apparent tradeoff can be clearly seen in Fig. 1(a).

Employing classical MC simulations with variable single-spin magnetic moments, we find that the inclusion of ODD terms in the effective Hamiltonian allows for fitting T_c at zero field using the same exchange parameter that accurately fits the data at low temperatures and high transverse fields. At the same time, we find better agreement with experimental results for the long unexplained weak dependence of T_c on the transverse field at small fields, and the linear dependence of T_c on Ho³⁺ concentration in the absence of a transverse field.

Theoretical considerations. Off-diagonal terms of the dipolar interaction have been known to give rise to many interesting phenomena in the case of the diluted LiHo_xY_{1-x}F₄ in presence of an *external* transverse field, where they do not cancel by symmetry [19–22,30]. We argue that similar effects, arising from *internal* transverse fields exerted by the single-ion expectation values $\langle J_i^z \rangle$ on the x angular momentum component J_j^x through terms of the form $V_{ij}^{zx} \langle J_i^z \rangle J_j^x$, make a significant impact on the phase diagram even in the undiluted case. The reason is that these ODD terms have a distinctly different contribution in the FM phase, where they are more likely to cancel by symmetry, than in the paramagnetic phase, where they are less likely to do so. For example, a pair of spins that lie along the a axis of the crystal will exert a transverse field on spins located between them, above or below the axis connecting the two spins, if the two spins have opposite orientations. Said field acts to lower the energy of the spin on which it acts regardless of its state, thereby energetically favoring the antialigned configuration of its two neighboring spins.

See illustrations in Fig. 2. This interaction thus constitutes a disorder-enhancing mechanism which acts to decrease the critical temperature. It requires the existence of three spins and correlation between two of them—an important aspect which will be discussed further below.

Another effect of the transverse fields is to decrease the absolute value of $\langle J^z \rangle$ for the two lowest single-ion electronic states, by mixing them with the higher electronic states. This also contributes to the reduction of T_c just by reducing the dominant zz dipolar term proportional to $\langle J^z \rangle^2$. This mechanism of the correlation induced enhancement of the transverse field, by its nature, is not likely to be captured in any sort of mean-field-like analysis as it depends on the spatial fluctuations of the states. We posit that including the ODD terms is necessary to explain the previously mentioned discrepancies between theory and experiment.

Numerical details. In order to examine the effect of ODD terms on the phase diagram of LiHo_xY_{1-x}F₄ we perform Monte Carlo simulations using an effective Hamiltonian derived building upon the work of Chakraborty *et al.* [27], but keeping the ODD terms. In this way we get an effective Hamiltonian:

$$H_{\text{eff}} = \sum_i V_C(\hat{\mathbf{J}}_i) - g_L \mu_B \sum_i \hat{\mathbf{B}}_i \cdot \hat{\mathbf{J}}_i \quad (1)$$

where

$$\begin{aligned} \hat{B}_i^x &= B_x - g_L \mu_B \sum_{j \neq i} V_{ij}^{zx} \hat{J}_j^z, \\ \hat{B}_i^y &= -g_L \mu_B \sum_{j \neq i} V_{ij}^{zy} \hat{J}_j^z, \\ \hat{B}_i^z &= -\frac{1}{2} g_L \mu_B \sum_{j \neq i} V_{ij}^{zz} \hat{J}_j^z - \frac{J_{\text{ex}}}{2g_L \mu_B} \sum_{j \in \text{NN}} \hat{J}_j^z \end{aligned} \quad (2)$$

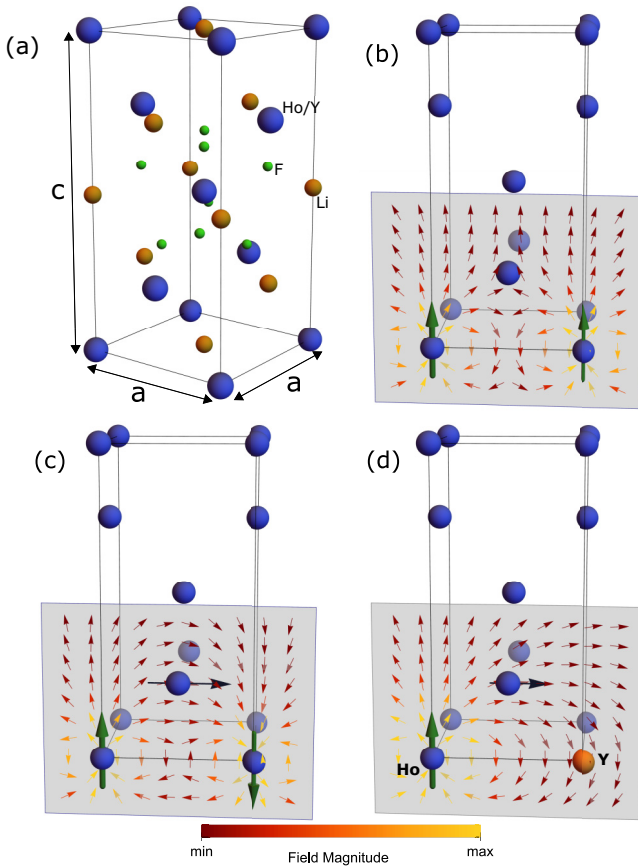


FIG. 2. Crystal and magnetic structures of $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$. (a) Crystal structure of LiHoF_4 . Only the F^- ions nearest to the central Ho^{3+} site are shown. (b) Correlated spins, more abundant in the FM phase, induce a field with vanishing transverse component on their common intermediate neighbor. (c) Spins in opposite orientations, more abundant in the PM phase, induce a nonzero transverse magnetic field on the third spin. (d) When one of the Ho^{3+} ions is replaced by a Y^{3+} ion, the remaining Ho induces a transverse field on its neighbor regardless of its orientation. Thick green arrows indicate dipole moments, small colored arrows show the magnetic field generated by the two magnetic dipoles, and a narrow black arrow qualitatively indicates the magnitude of the transverse component of the magnetic field exerted on the middle ion. The full effect captured in the simulation is a result of the transverse field not just on the middle ion, but on all other ions in the system. For further details on this point see Supplemental Material [31].

act as effective internal fields when taking their expectation values, thereby transforming Eq. (1) to an effective Hamiltonian for the single spins i . The $V_C(\mathbf{J}_i)$ term is a crystal field potential which imposes an Ising easy axis along the c axis of the crystal, with a first excited state at ≈ 10 K above the ground-state doublet [28]. $V_{ij}^{\mu\nu}$ is the magnetic dipole interaction, J_{ex} is the nearest-neighbor exchange interaction coupling constant, $\mu_B = 0.6717 \text{ KT}^{-1}$ is the Bohr magneton, and $g_L = \frac{5}{4}$ is a Landé g factor. \mathbf{J}_i are angular momentum operators of the Ho^{3+} ions. See further details on the derivation of the effective Hamiltonian in the Supplemental Material [31].

Since the Ho^{3+} ions retain their Ising character up to transverse fields well above the critical transverse field [30]

we model a single-ion as a two-state Ising system under an applied field exerted by all other ions in the system, as well as the external field. This applied field not only shifts the energies of the two states, but also modifies the single-ion magnetic moments $\langle J^z \rangle$ associated with them. These are the magnetic moments which in turn exert magnetic fields on other ions. Therefore, each site has two possible states, $|\uparrow\rangle$ and $|\downarrow\rangle$, and each of these has two quantities of interest associated with it: local energy $\langle \uparrow(\downarrow) | H_{\text{single-site}} | \uparrow(\downarrow) \rangle$ and magnetic moment $\langle \uparrow(\downarrow) | J^z | \uparrow(\downarrow) \rangle$ where

$$H_{\text{single-site}} = V_C(\hat{\mathbf{J}}) - g_L \mu_B \mathbf{B} \cdot \hat{\mathbf{J}}. \quad (3)$$

The method of employment of the effective Hamiltonian within the Monte Carlo simulations is detailed in the “Numerical Methods” section of the Supplemental Material [31].

Results. Figure 1(b) shows the $B_x - T$ phase diagram of LiHoF_4 . Our results, with ODD terms included and excluded, both use the exchange parameter $J_{\text{ex}} = 1.16 \text{ mK}$ suggested in Refs. [28,29] which corresponds to the fitting at low temperatures and high transverse fields. It is easy to see that the simulation with ODD terms excluded corresponds to the mean-field calculation, while the results with ODD terms included are in close agreement with the experimental results, for zero and small transverse fields.

Thus, the inclusion of the ODD terms results in good agreement with experiment at zero transverse field without the need to choose J_{ex} that is in clear disagreement with experiment at lower temperatures and higher transverse fields. For finite but small transverse fields we find that the decrease in T_c due to the ODD terms is maintained. We emphasize that our simulation is a classical MC simulation, but one that allows for varying magnetic moments due to the influence of transverse fields. Therefore, the $T_c(B_x)$ dependence in our results is a consequence of the renormalization of individual magnetic moments due to the quantum coupling of each of the Ising doublet states to higher excited electronic states, as opposed to quantum fluctuations between the two Ising doublet states. Hence, our model is expected to be valid at low fields $B_x \lesssim 1 \text{ T}$ where quantum fluctuations are small [30].

Another facet of the incomplete quantitative understanding of this material has to do with the phase diagram of $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ as a function of the Ho^{3+} concentration x at zero applied field. At moderate to high concentrations ($x \gtrsim 0.4$) experiments show a linear dependence of T_c on x , in agreement with the mean-field prediction [15,29,37], whereas the available numerical work seems to indicate a steeper decline of T_c as x is reduced [17,38].

We note here that the inclusion of the ODD terms in the effective low-energy Hamiltonian of the system leads to an effective three-spin interaction, proportional to the anticorrelation of two spins and the existence of the third. We thus expect this term to depend strongly on Ho concentration, allowing for its distinction from the excess antiferromagnetic exchange used to parametrize the system, and for better agreement with the experimental $x - T$ phase diagram. In Fig. 3 we present our results for T_c as function of Ho concentration x in the presence of ODD terms and exchange parameter $J_{\text{ex}} = 1.16 \text{ mK}$, and in the absence of ODD terms and $J_{\text{ex}} = 3.91 \text{ mK}$. Indeed, the results with ODD terms included show milder reduction of T_c with decreased concentration, in

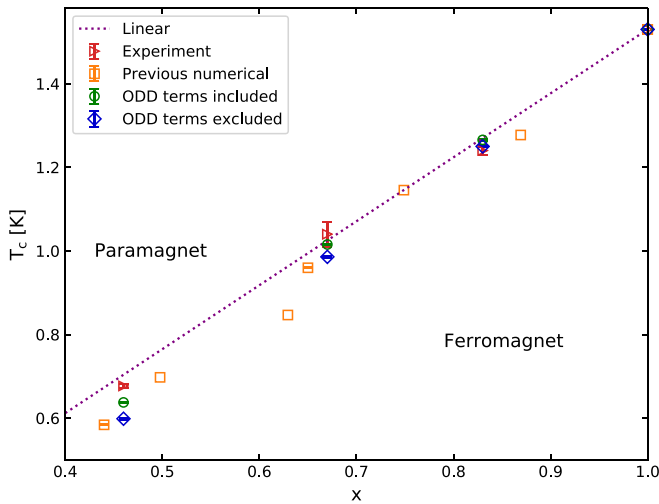


FIG. 3. Transition temperature, T_c , vs Ho^{3+} concentration, x , phase diagram from different sources: experimental from Ref. [29] (triangles), and numerical from Refs. [17,38] (squares) and this Letter with ODD terms included (circles) and ODD terms excluded (diamonds). A dotted line shows the mean-field prediction (linear). Numerical results from this Letter are scaled so that they agree with the experimental $T_c(x = 1) = 1.53$ K. The results with ODD terms included use $J_{\text{ex}} = 1.16$ mK, while the results with ODD terms excluded use $J_{\text{ex}} = 3.91$ mK, as suggested, e.g., in Ref. [25].

better agreement with the experimental findings of $T_c(x) = x T_c(x = 1)$.

An additional microscopic indication of the effect of ODD terms can be obtained by inspecting the distribution of local transverse fields. Figure 4 shows the distribution of B_x at the end of the simulation, when the system has reached thermodynamic equilibrium, for simulations where ODD terms are included and where they are excluded, yet still considered at the end of the simulation for the calculation of the effective transverse fields. It is clear that, at a given temperature, when ODD terms are included the distribution of B_x becomes wider. This is expected, since when ODD terms are included, configurations that maximize internal transverse fields become more energetically favorable and are thus more abundant at any given temperature.

Lastly, in order to demonstrate the effect of the ODD terms on the full $B_x - T$ phase diagram we pursue a simplified approach, assuming the main difference between the FM and PM phases relevant to the effectiveness of the ODD-induced mechanism is the width of the distribution of local transverse fields. For simplicity, we assume that the FM phase is characterized by a vanishing width of this distribution, while the PM phase is characterized by a finite width h which we take to be 0.4 T, i.e., the field exerted on a spin by its two nearest neighbors along the x or y directions while they are in opposite orientations from each other. Unsurprisingly, this is also approximately the value of the secondary peaks in Fig. 4. In both phases the distribution of local transverse fields is centered around the value of the external field B_x . Hence, on average half of the spins experience a local transverse field $B_x + h$ and the other half experience $B_x - h$. This is true

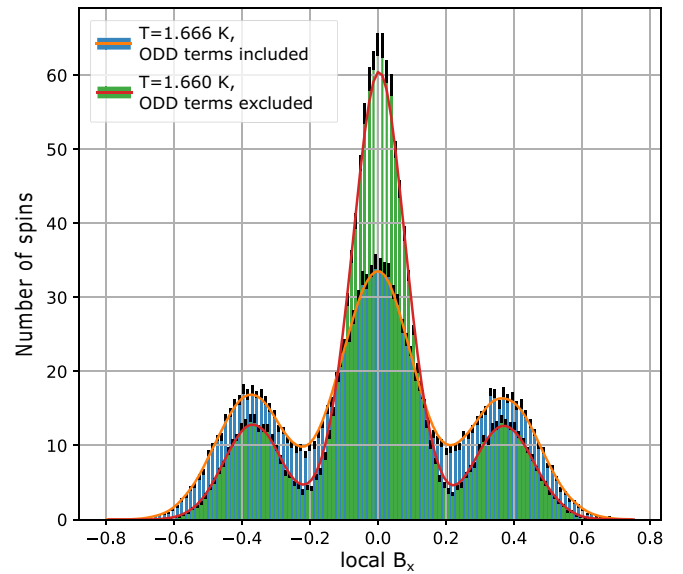


FIG. 4. Distribution of local B_x at thermodynamic equilibrium for system size $L = 7$ with zero external transverse field and at $x = 1$. The temperatures used, around $T = 1.66$ K, are below T_c when ODD terms are excluded and above T_c when ODD terms are included. The black vertical lines at the end of each bar are standard errors. For each distribution a solid smooth line is plotted as a guide to the eye, obtained by convolution of the bin values with a Gaussian function. Where ODD terms are excluded, they are nevertheless considered at the end of the simulation for obtaining these effective transverse fields. One can clearly see that when ODD terms are included, the distribution of B_x is wider than when they are excluded—the peak at $B_x = 0$ is lower, compensated by higher values at the exterior. This is an indication of the ODD induced mechanism at work. Configurations maximizing internal transverse fields become more energetically favorable and thus for any given temperature they are more common.

even though at $B_x > 0$ the former option is more energetically favorable, because, owing to the crystal's mirror symmetry, any spin which generates a positive local field at some site also generates a negative local field at another site. Thus, we estimate the energy reduction of the PM phase due to the ODD interactions, as $\Delta E(B_x) = E(B_x) - [E(B_x - h) + E(B_x + h)]/2$, where, for simplicity, we take $E(B_x)$ to be the average of the two lowest eigenenergies of (3) with the given B_x . Assuming that the reduction in T_c due to the inclusion of ODD terms, ΔT_c , is proportional to the energy reduction $\Delta E(B_x)$, we find the appropriate factor by demanding $\Delta T_c(B_x = 0) = T_c^{\text{MF}}(0) - T_c^{\text{exp}}(0) \approx 1.79 - 1.53 \approx 0.25$ K. Using this scaling factor we apply a B_x -dependent shift $\Delta T_c(B_x)$ to the mean-field phase boundary to obtain an approximate phase boundary with the effect of ODD terms included, as presented in Fig. 5.

Discussion. We have shown here that the description of anisotropic dipolar systems by the Ising model, and in the presence of a transverse field by the transverse field Ising model, is essentially insufficient. Even for the pure system off-diagonal dipolar terms induce an effective three-spin interaction, enhancing paramagnetic fluctuations and lowering the critical temperature. We have analyzed the effect of the

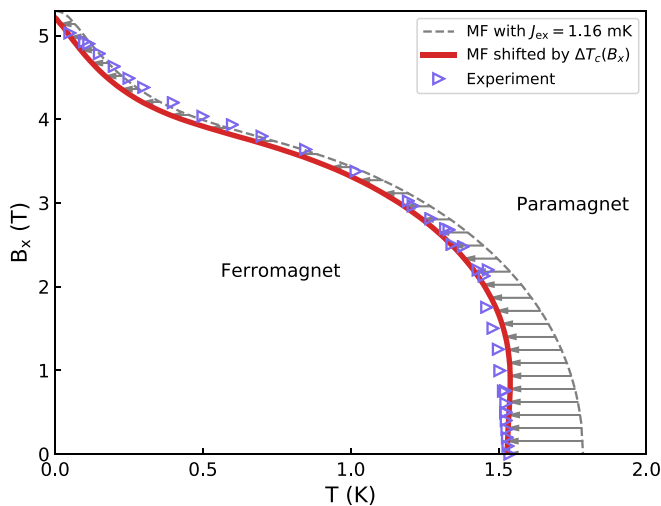


FIG. 5. FM-PM phase transition line in the temperature-transverse field plane, as obtained by simple mean field [29] (dashed gray line) and by including the T_c -reducing effect of ODD terms (solid red line). Horizontal gray arrows indicate the application of $\Delta T_c(B_x)$ to the mean-field result. Triangles denote the same experimental data also shown in Fig. 1.

ODD terms on the relation between critical temperature and both transverse field and dilution, thereby addressing unanswered puzzles regarding discrepancies between theory and experiment. Our results at small fields are obtained with the same exchange parameter used to fit the phase transition at low temperatures and high transverse fields, and produce improved fitting to experimental data at finite transverse fields and as a function of Ho concentration. Furthermore, applying a simplified analysis we have also shown that the critical-temperature-reducing effect of ODD terms is expected to decrease with an increase of the external transverse field, thus further strengthening the proposition that ODD terms play a key role in determining the shape of the phase boundary line. Thus, our results point to the need to include the quantum fluctuations induced by the off-diagonal terms in any theoretical consideration, classical and quantum, of anisotropic dipolar systems. Examples are classical and quantum annealing protocols, and a comprehensive quantum modeling of the system required to establish its full phase diagram.

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