

Quantum corrections to the Ising interactions in $\text{LiY}_{1-x}\text{Ho}_x\text{F}_4$

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We systematically derive an effective Hamiltonian for the dipolar magnet $\text{LiY}_{1-x}\text{Ho}_x\text{F}_4$, including quantum corrections which arise from the transverse dipolar and hyperfine interactions. These corrections are derived using a generalised Schrieffer-Wolff transformation to leading order in the small parameters given by the ratio of the interaction energies to the energy of the first excited electronic state of the Holmium ions. The resulting low-energy Hamiltonian involves two-level systems, corresponding to the low-lying electronic states of the Holmiums, which are coupled to one another and to the Holmium nuclei. It differs from that obtained by treating the electronic states of the Holmium as a spin-1/2 with an anisotropic g-factor. It includes effective on-site transverse fields, and both pairwise and three-body interactions among the dipoles and nuclei. We explain the origins of the terms, and give numerical values for their strengths.

I. INTRODUCTION

The rare-earth compound $\text{LiY}_{1-x}\text{Ho}_x\text{F}_4$ has often been described as a model quantum magnet, and as such has been studied for over three decades^{1,2,3,4,5,6}. The basic model for this material is a diluted Ising model with dipole interactions, and depending on the dilution the low-temperature phase is expected to be either a ferromagnet or a spin-glass⁷. Applying a strong transverse field to the material introduces quantum fluctuations which can lead to domain wall tunnelling in the ferromagnet⁸, and to quantum melting of the ferromagnet³ and the spin-glass^{9,10,11,12,13}. Even in the absence of an applied transverse field, however, quantum tunnelling of magnetic dipoles can be observed in $\text{LiY}_{1-x}\text{Ho}_x\text{F}_4$ ^{4,14}. Furthermore, samples with a Holmium concentration of 4.5% do not appear to behave as a spin-glass on cooling, as expected for a classical Ising model. Instead they show unexplained “anti-glass” properties which have been attributed to quantum mechanics.⁵ This attribution is supported by theoretical predictions of the static susceptibility, which agree with experiments only once quantum corrections are included⁶.

Although quantum effects are implicated in several phenomena in $\text{LiY}_{1-x}\text{Ho}_x\text{F}_4$ in the absence of an applied transverse field, there is no derivation in the literature of an effective low-energy Hamiltonian which contains them. Here we derive such a Hamiltonian, projecting out the high-energy electronic states to obtain a theory describing the low-lying electronic doublet and the nuclei. We shall find a Hamiltonian which has a different form from that appropriate in a strong applied field¹⁵, and whose electronic part differs from the two-level model previously proposed for the zero-field case⁶.

II. MODEL

The magnetic degrees of freedom in $\text{LiY}_{1-x}\text{Ho}_x\text{F}_4$ are the f electrons on the Ho^{3+} ions. The strong spin-orbit coupling of the ions leads to a well-defined $J = 8$ for

the ions, and an associated dipole moment $\mu = g_L\mu_B$, with the Landé g-factor $g_L = 5/4$. The $2J + 1 = 17$ -fold degeneracy of the free ion is broken by the crystal-field Hamiltonian, leaving a degenerate ground-state doublet. All matrix elements of J_x and J_y are zero within this ground-state subspace, but there are non-zero matrix elements for J_z . This is the source of the strong Ising anisotropy in the interactions and response to an applied field.

The interactions between Holmium ions are dipolar, giving an interaction Hamiltonian

$$H_{\text{int}} = \frac{1}{2} \sum_{i,j} \frac{\mu_0 \mu_B^2 g_L^2}{4\pi r_{ij}^3} (\mathbf{J}_i \cdot \mathbf{J}_j - 3(\hat{\mathbf{r}}_{ij} \cdot \mathbf{J}_i)(\hat{\mathbf{r}}_{ij} \cdot \mathbf{J}_j)). \quad (1)$$

The crudest way to obtain a low-energy effective Hamiltonian from (1) is to truncate the electronic state-space to the ground-state doublet on each Holmium ion. It is possible to choose a basis for the doublet in which J_z has no off-diagonal matrix elements, while the diagonal matrix elements are α and $-\alpha$. With this choice of basis, truncating leads to the dipolar Ising model

$$H_{\text{int}} = \frac{1}{2} \sum_{i,j} \frac{\alpha^2 \mu_0 \mu_B^2 g_L^2}{4\pi r_{ij}^3} (1 - 3(\hat{\mathbf{r}}_{ij} \cdot \hat{\mathbf{k}})^2) \sigma_i^z \sigma_j^z, \quad (2)$$

where σ^x, σ^y and σ^z are the usual Pauli matrices. As discussed in previous studies of this system^{3,4,10,15} the contact hyperfine interaction between the Holmium nuclei and the electronic degrees of freedom also plays an important role, as it is typically of similar strength to the dipolar interaction. Under the simple two-state truncation scheme described above the hyperfine interaction also takes a simple Ising form,

$$H_{\text{hyp}} = \sum_i A_J \mathbf{I}_i \cdot \mathbf{J}_i \quad (3)$$

$$\longrightarrow \alpha \sum_i A_J I_i^z \sigma_i^z, \quad (4)$$

as do other couplings to the Holmium moments.

The procedure of projecting into the low-energy subspace of the single-ion Hamiltonian has been applied in the presence of an applied transverse field.¹⁵ The field changes the low-energy states, leading to finite matrix elements for J_x and J_y within the low-energy doublet. This leads to corrections to the Ising forms (2,4) that are pair interactions involving at least one of $\sigma^x, \sigma^y, I^x, I^y$, and to effective field terms.

To obtain quantum terms in the absence of an applied transverse field we must go beyond a simple projection onto the low-lying single-ion states. We thus now consider the lowest three levels of the crystal-field Hamiltonian, denoting the two states of the doublet as $|\uparrow\rangle, |\downarrow\rangle$ and the first excited state, with energy Δ , as $|\Gamma\rangle$. The parameters in such a three-level model are Δ and the matrix elements of the angular momentum operators, and can be obtained from a numerical diagonalisation of the full crystal field Hamiltonian. With an appropriate choice of basis this gives,¹⁵

$$\langle\uparrow|J_z|\uparrow\rangle = -\langle\downarrow|J_z|\downarrow\rangle = 5.52 = \alpha, \quad (5)$$

$$\langle\downarrow|J_x|\Gamma\rangle = \langle\uparrow|J_x|\Gamma\rangle = 2.4 = \rho, \quad (6)$$

$$\langle\downarrow|J_y|\Gamma\rangle = -\langle\uparrow|J_y|\Gamma\rangle = i\rho, \quad (7)$$

$$\Delta = 10.8\text{K}. \quad (8)$$

All other matrix elements of angular momentum among the three states vanish.

III. DERIVATION OF THE EFFECTIVE HAMILTONIAN

Quantum corrections to the Ising Hamiltonian (2,4) appear once one includes the state at $|\Gamma\rangle$ because the interaction terms (1) and (3) couple $|\Gamma\rangle$ to the electronic ground-state doublet. The typical nearest-neighbour dipolar interaction energy scale is $\approx 300\text{mK}$ and $A_J \approx 38\text{mK}$ ^{4,15}. Since these interaction scales are small compared with Δ the couplings to the $|\Gamma\rangle$ states can be treated in perturbation theory, and an effective Ising model obtained using the standard Schrieffer-Wolff procedure¹⁶. The resulting model will include quantum terms arising from virtual transitions between the ground-state doublet and the $|\Gamma\rangle$ state.

To derive the effective Hamiltonian we begin by writing the three-level model as $H = H_0 + H_T$, where H_0 contains terms which do not couple the doublet to $|\Gamma\rangle$, and H_T contains those which do. Measuring energy from the electronic ground-state doublet, H_0 contains the crystal field term $V_c = \Delta \sum_i |\Gamma_i\rangle\langle\Gamma_i|$ and the Ising parts of the interactions, while H_T contains the parts of the interactions which involve J_x and J_y . Including the hyperfine interaction the Hamiltonian can be split as

$$H_0 = V_c + \frac{1}{2} \sum_{i \neq j} J_{ij}^{zz} J_i^z J_j^z + \sum_i A_J I_i^z J_i^z, \quad (9)$$

$$H_T = \frac{1}{2} \sum'_{i \neq j, \nu, \mu} J_{ij}^{\nu\mu} J_i^\nu J_j^\mu + A_J \sum_i (I_i^x J_i^x + I_i^y J_i^y), \quad (10)$$

where $\nu, \mu = x, y, z$, but the prime on the sum indicates that we must exclude all terms that have $\nu = \mu = z$.

We then seek a unitary transformation $H \rightarrow e^S H e^{-S}$ which decouples the electronic doublet from the $|\Gamma\rangle$ state to first order in H_T . Such a transformation obeys

$$[S, H_0] = -H_T. \quad (11)$$

An S which approximately satisfies (11) can be constructed from H_T using projection operators,

$$S = \sum'_{i \neq j, \nu, \mu} \frac{J_{ij}^{\nu\mu} (\Gamma_i^\nu \Gamma_j^\mu J_i^\nu J_j^\mu P_i P_j - P_i P_j J_i^\nu J_j^\mu \Gamma_i^\nu \Gamma_j^\mu)}{2\epsilon_{\nu\mu} \Delta} + \frac{A_J}{\Delta} \sum_i \Gamma_i (I_i^x J_i^x + I_i^y J_i^y) P_i - \frac{A_J}{\Delta} \sum_i P_i (I_i^x J_i^x + I_i^y J_i^y) \Gamma_i, \quad (12)$$

$$P_i = |\uparrow_i\rangle\langle\uparrow_i| + |\downarrow_i\rangle\langle\downarrow_i|, \quad (13)$$

$$\Gamma_i^\nu = \begin{cases} |\Gamma\rangle\langle\Gamma| & \text{if } \nu = x, y \\ P_i & \text{if } \nu = z \end{cases}, \quad (14)$$

$$\epsilon_{\nu\mu} = \begin{cases} 2 & \text{if } \nu = \mu \\ 1 & \text{if } \nu \neq \mu. \end{cases} \quad (15)$$

The form (12) for S actually obeys the relation $[S, V_c] = -H_T$, and although we have successfully eliminated the linear term in H_T the remainder of the commutator $[S, H_0 - V_c]$ generates new couplings between the doublet and $|\Gamma\rangle$. However these tunnelling terms are of order (interaction)²/ Δ , and as they can only couple between the ground states and $|\Gamma\rangle$, they can only contribute to the effective low-energy Hamiltonian in higher order perturbation theory. Therefore they do not contribute to the effective low-energy Hamiltonian to leading order in (interaction)²/ Δ , and vanish when we project onto the Ising basis at the end of the Schrieffer-Wolff procedure.

Having eliminated H_T , the effective two-state Hamiltonian is given to lowest order in (interactions)²/ Δ by¹⁶

$$H_{\text{eff}} = \prod_i P_i (H_0 + \frac{1}{2} [S, H_T]) \prod_i P_i. \quad (16)$$

This form extends the Ising interaction Hamiltonian by including second-order processes in which H_T causes virtual transitions from the electronic doublet to $|\Gamma\rangle$ and then back again.

IV. EFFECTIVE HAMILTONIAN

Substituting the form (12) for S into Eq. 16 and discarding irrelevant energy shifts we find that the effective Hamiltonian takes the form

$$H_{\text{eff}} = H_0 + H_D + H_{\text{DTB}} + H_N + H_{\text{ND}}. \quad (17)$$

This Hamiltonian operates in the space with sixteen states for each ion: two low-lying electronic states, each with the eight nuclear states. The various contributions to H_{eff} are labelled according to the interactions that give rise to them.

A. Dipolar processes

The first term in (17) arises from the second-order dipolar processes that involve only single pairs of spins, and is

$$H_D = \sum_i (h_i^x \sigma_i^x + h_i^y \sigma_i^y) + \sum_{i \neq j} \Delta_{ij}^{\nu\mu} \sigma_i^\nu \sigma_j^\mu. \quad (18)$$

We see that the pairwise dipolar interaction thus generates both transverse interaction terms and effective magnetic fields. In appendix A we give expressions for the strengths of the parameters h_i and $\Delta^{\nu\mu}$ in terms of the parameters of the underlying three-level model. Using these relations we estimate the characteristic magnitude of these corrections by calculating the contribution from a single nearest-neighbour. This yields $h_x \approx 3\text{mK}$, $h_y \approx 0$ and $\Delta^{\nu\mu} \approx 0.06\text{mK}$, which may be compared to the dipolar Ising energy $J_{NN}^{zz} \approx 300\text{mK}$, and the Ising hyperfine splitting $\alpha A_J \approx 210\text{mK}$. Note that terms such as (18) describe tunnelling between the low-lying electronic states. It is important to stress that this is not sufficient to generate tunnelling between the two electro-nuclear ground states of H_0 ¹⁰.

In addition to the dipolar processes involving only a single pair of Holmium ions there are processes involving three ions. In such a process the interaction between one pair of spins, say i, j , virtually excites the j th spin into $|\Gamma\rangle$, and the dipolar interaction of this spin with the k th spin brings the j th spin back into the Ising doublet. This generates two- and three- body interaction terms in the effective theory,

$$H_{\text{DTB}} = -\frac{\alpha^2 \rho^2}{\Delta} \sum_{i \neq j \neq k} (J_{ik}^{xz} J_{ij}^{xz} + J_{ik}^{yz} J_{ij}^{yz}) \sigma_k^z \sigma_j^z \quad (19)$$

$$- \frac{\alpha^2 \rho^2}{\Delta} \sum_{i \neq j \neq k} (J_{ik}^{xz} J_{ij}^{xz} - J_{ik}^{yz} J_{ij}^{yz}) \sigma_k^z \sigma_j^z \sigma_i^x \quad (20)$$

$$- \frac{\alpha^2 \rho^2}{\Delta} \sum_{i \neq j \neq k} (J_{ik}^{xz} J_{ij}^{yz} + J_{ik}^{yz} J_{ij}^{xz}) \sigma_k^z \sigma_j^z \sigma_i^y. \quad (21)$$

For an isolated group of three spins in which $\{i, j\}$ and $\{i, k\}$ are nearest neighbours we obtain 3 mK for the magnitude of the interaction terms $\sigma_k^z \sigma_j^z$, $\sigma_k^z \sigma_j^z \sigma_i^x$ and $\sigma_k^z \sigma_j^z \sigma_i^y$.

B. Hyperfine processes

The contributions to the effective Hamiltonian from the hyperfine interactions are simpler to deal with, as

they are confined to each site. They are

$$H_N = \frac{\rho^2 A_J^2}{\Delta} \sum_i ((I_i^z - (I_i^+)^2) + ((I_i^+)^2 + (I_i^-)^2) \sigma_i^x) - \frac{\rho^2 A_J^2}{\Delta} \sum_i (i((I_i^+)^2 - (I_i^-)^2) \sigma_i^y - 2I_i^z \sigma_i^z). \quad (22)$$

We see that in second order perturbation theory the hyperfine interaction leads to spin flipping terms which require the z component of the nuclear spin to change by two units, gives a slight correction to the longitudinal hyperfine interaction, and introduces a weak nuclear anisotropy. The factors outside the sum give 0.86mK for the interaction energy.

C. Mixed hyperfine-dipolar processes

Finally, the term in the effective Hamiltonian labelled H_{ND} arises due to second-order processes whereby the electrons are virtually excited into the state $|\Gamma\rangle$ by the dipolar interaction and then de-excited by the hyperfine interaction or vice versa:

$$H_{\text{ND}} = -\frac{\alpha \rho^2 A_J}{\Delta} \sum_{i \neq j} J_{ij}^{zx} (I_i^x (1 + \sigma_i^x) + I_i^y \sigma_i^y) \sigma_j^z - \frac{\alpha \rho^2 A_J}{\Delta} \sum_{i \neq j} J_{ij}^{zy} (I_i^y (1 - \sigma_i^x) + I_i^x \sigma_i^y) \sigma_j^z. \quad (23)$$

Note that the nuclei mediate a coupling between electronic states of the form $\sigma^{x,y} \sigma^z$ which does not occur in H_D .

V. DISCUSSION

In recent papers^{10,11,12,13} it was shown that the non-Ising parts of the dipolar coupling provide a route by which an applied transverse field can destroy spin-glass order in $\text{LiY}_{1-x}\text{Ho}_x\text{F}_4$. Although our model contains spontaneous transverse fields we do not expect them to destroy the spin-glass phase in this way, because the model retains time-reversal symmetry. An interesting feature of the effective Hamiltonian is that the spontaneous transverse fields and the two- and three- body interactions are all correlated. Theoretical work on spin models with several forms of random couplings, such as the random transverse-field Ising model¹⁷, generally considers different interaction terms as independent.

The approach given here could be straightforwardly extended to derive an effective Hamiltonian for $\text{LiY}_{1-x}\text{Ho}_x\text{F}_4$ in an applied transverse field, so long as the field strength is sufficiently small for perturbation theory to apply. This generates a variety of new processes at second order. The straightforward process, in which the applied field excites to the $|\Gamma\rangle$ state and back again, leads to an electronic tunnelling term $\propto B^2/\Delta$. But note that

there will also be mixed processes, for example where the applied field causes the transition to $|\Gamma\rangle$ and the dipole interaction causes the transition back into the electronic doublet, which will generate electronic tunnelling terms $\propto B/\Delta$.

We stress that although our Hamiltonian contains terms which generate transitions between the states of the electronic Ising doublet, and further such terms will be introduced by an applied field, these terms alone do not couple the doubly-degenerate electro-nuclear ground states of the Ising single-ion Hamiltonian. To generate quantum fluctuations between the electro-nuclear ground states requires processes which flip the nuclear spin, and hence the splittings of the electro-nuclear ground states will be much smaller than the tunnelling terms connecting the bare electronic states.

Although we do not expect the quantum corrections to formally destroy the ordered phase unless there is an applied field, they can still affect the thermodynamics, changing the susceptibilities and moving the phase boundary. However, we only expect this to occur at very low temperatures. In this context we note that the experimental susceptibility of $\text{LiY}_{1-x}\text{Ho}_x\text{F}_4$ in the 10 – 100 mK temperature regime of the antighlass experiment has been reproduced by a quantum theory of a two-level model⁶. That model is obtained by neglecting the nuclei and treating the Holmium ions as spin-1/2 ions with an anisotropic g-factor, *i.e.* writing $J_i^\mu = g^\mu \sigma_i^\mu$, with $\mu = x, y, z$. We note that the resulting Hamiltonian is very different from that which would be obtained by neglecting the nuclei in our model: there are no spontaneous field terms and the non-Ising interactions decay as $1/r^3$ (here $1/r^6$), while the quoted values for g give the energy scale for the largest non-Ising coupling of $\approx 30\text{mK}$, whereas here we have 3mK .

While we do not expect the small corrections derived here to affect thermodynamics except at very low temperatures, they may be relevant to understanding dynamics at much higher temperatures. Quantum tunnelling of the magnetisation has been observed in the magnetisation relaxation and susceptibility of the dilute compound $\text{LiY}_{0.998}\text{Ho}_{0.002}\text{F}_4$, due to both single-ion⁴ and two-ion processes¹⁴, and it would be interesting to compare the details of these results with the processes given here. An understanding of the single or few-ion electro-nuclear dynamics, based on the Hamiltonian (17), may also help to explain the antighlass experiments⁵. In the high-temperature regime these experiments show a characteristic relaxation time for the magnetisation which is activated, with a barrier similar to the width of the hyperfine multiplet and the nearest-neighbour interaction strength. Extrapolating this behaviour indicates that this particular activated dynamics freezes out on the experimental frequency scale as the temperature is lowered into the antighlass regime. The quantum terms derived here, although small, may perhaps then provide a route to the observed dynamics. The potential significance of quantum electro-nuclear dynamics for the antighlass ex-

periment has been noted by Atsarkin¹⁸, who proposes a specific relaxation mechanism due to interactions of the form (18).

VI. CONCLUSIONS

In this paper we have motivated the need to go beyond the simple dipolar electro-nuclear Ising Hamiltonian commonly used to describe $\text{LiY}_{1-x}\text{Ho}_x\text{F}_4$, and have derived an effective low energy Hamiltonian which includes the quantum corrections caused by the transverse elements of the dipolar and hyperfine interactions. We have given estimates for the typical magnitudes of these correction terms and have shown that they are typically about one percent as strong as the energy scale associated with the Ising interactions. As a result, we do not expect any qualitative changes to the Ising phase diagram, but as we have highlighted in this paper, these quantum corrections can describe a large variety of single and many-body processes which might play significant roles in the observed dynamics. Thus, for low temperatures, our effective Hamiltonian should serve as a good starting point for a microscopic investigation of the dynamical physics of $\text{LiY}_{1-x}\text{Ho}_x\text{F}_4$, and as it can be used across the whole dilution series, should contain the rich low energy physics which characterises the spin glass, free ion, and presumably, the ‘‘antighlass’’ phases of this material.

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APPENDIX A: RELATION OF MICROSCOPIC PARAMETERS TO THE EFFECTIVE INTERACTION AND ON-SITE TERMS IN H_D

The components of the effective magnetic field \mathbf{h}_i , defined in Eq. 18, are related to the original microscopic Hamiltonian by,

$$h_i^x = \frac{\rho^2 \alpha^2}{\Delta} \sum_j ((J_{ij}^{zy})^2 - (J_{ij}^{zx})^2) + \frac{\rho^4}{2\Delta} \sum_j ((J_{ij}^{yy})^2 - (J_{ij}^{xx})^2), \quad (\text{A1})$$

$$h_i^y = -2 \frac{\rho^2 \alpha^2}{\Delta} \sum_j J_{ij}^{zy} J_{ij}^{zx} - \frac{\rho^4}{2\Delta} \sum_j J_{ij}^{xy} (J_{ij}^{xx} + J_{ij}^{yy}) \quad (\text{A2})$$

$$h_i^z = 0. \quad (\text{A3})$$

The components of the effective magnetic field vanish in the undiluted crystal, as expected from the crystal symmetry.

The transverse dipolar interactions between spins are described by the following couplings,

$$\Delta_{ij}^{xx} = \frac{\rho^4 (2(J_{ij}^{xy})^2 - (J_{ij}^{xx})^2 - (J_{ij}^{yy})^2)}{4\Delta}, \quad (\text{A4})$$

$$\Delta_{ij}^{yy} = \frac{-\rho^4 (J_{ij}^{xx} J_{ij}^{yy} + J_{ij}^{xy} J_{ij}^{xy})}{2\Delta}, \quad (\text{A5})$$

$$\Delta_{ij}^{zz} = \frac{\rho^4 (J_{ij}^{xx} J_{ij}^{yy} - J_{ij}^{xy} J_{ij}^{xy})}{2\Delta}, \quad (\text{A6})$$

$$\Delta_{ij}^{xy} = \frac{\rho^4 (J_{ij}^{yy} J_{ij}^{xy} - J_{ij}^{xx} J_{ij}^{xy})}{2\Delta}, \quad (\text{A7})$$

$$\Delta_{ij}^{yx} = \Delta_{ij}^{xy}. \quad (\text{A8})$$

These are the only couplings generated; there are no terms such as $\sigma^x \sigma^z$, since these would break time-reversal symmetry.

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