Exact diagonalization for a 16-site spin-1/2 pyrochlore cluster

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Abstract

We find exact solutions to the Hamiltonian of a 16-site spin-1/2 pyrochlore cluster with nearest neighbour exchange interactions. The methods of group theory (symmetry) are used to completely block-diagonalize the Hamiltonian, yielding precise details about symmetry of the eigenstates, in particular those components which are spin ice states, in order to evaluate the spin ice density at finite temperature. At low enough temperatures, a ‘perturbed’ spin ice phase, where the ‘2-in-2-out’ ice rule is largely obeyed, is clearly outlined within the four parameter space of the general model of exchange interactions. The quantum spin ice phase is expected to exist within these boundaries.

Keywords: pyrochlores, spin ice, exact diagonalization

1. Introduction

The ideal spin ice was proposed by Philip Anderson in 1956 [1] in a study of finite entropy, short-range ordered states in magnetic spinels. In pyrochlores and spinels (which share the same crystallographic symmetry group, Fd3m), the ‘spin ice rule’ is that the four Ising-like spins on a tetrahedron must be oriented so that two of them point toward the centre of each tetrahedron, and two point away, the so-called ‘two-in-two-out’ configuration. Given that the tetrahedra share spins on each vertex, the number of pure spin ice states (those states for which the spin ice rule is satisfied on every tetrahedron) is constrained by the geometry of the system and is approximately 1.5N/2 [2]. The spin ice rule is precisely analogous to the ice rule for water ice, which governs the positions of the hydrogen atoms, and which results in a highly degenerate ground state in water ice, producing a residual entropy at zero temperature, as shown by Pauling in 1935 [2]; spin ice is expected to have the same property [1]. This classical spin ice state occurs in several pyrochlores, including Ho2Ti2O7 and Dy2Ti2O7 [3, 4]. The elementary excitations of spin ice are magnetic monopoles, which occur when the spin ice rule is violated and behave in accordance with Maxwell’s equations [5–7].

In contrast to classical spin ice materials, a quantum spin ice occurs when there are quantum fluctuations of the degenerate spin ice states, even at zero temperature, which may lead to the formation of a quantum spin liquid [8–10]. Examples of candidate quantum spin ice pyrochlores include Tb2Ti2O7 [11, 12], Yb2Ti2O7 [13–16], Ce2Zr2O7 [17], and several Pr oxides [18–20].

In this article we investigate a quantum spin Hamiltonian for the pyrochlore lattice using exact diagonalization. This numerical method can be used to evaluate the entire spectrum, as well as finite temperature spin-correlations, for small systems. This method has been applied to spin-1/2 pyrochlore magnets with anti-ferromagnetic interactions [21] in order to investigate spin correlations on a ‘breathing’ pyrochlore...
lattice, which occurs when the tetrahedra of two different orientations have two different sets of coupling constants. In this work, we study the phase space of the most general model of exchange interactions on the pyrochlore lattice in the vicinity of the classical spin ice phase. The general form of the Hamiltonian has four exchange parameters and the methods of group theory are used to block-diagonalize the Hamiltonian in order to reduce the computational effort required to investigate this 4-parameter space. One of our aims is to determine the position of candidate quantum spin ice materials within this space.

2. Perturbed spin ice

2.1. The Hamiltonian

In pyrochlore magnets, the spins occupy the 16d Wyckoff position of the crystallographic space group $Fd\bar{3}m$, which are the vertices of a network of corner-sharing tetrahedra. The site symmetry is $D_{3d}$ and there are four inequivalent sites (which we number $s = 1, 2, 3, 4$) which are distinguished by the direction of their 3-fold symmetry axes. The $D_{3d}$ site symmetry lifts the $2f+1$-fold degeneracy of spin $J$ magnetic ions into singlets and doublets. Any well-separated doubly degenerate ground state can be treated as spin-1/2 doublet. There are three different kinds of doublets, one which is non-Kramers (integral $J$) and two which are Kramers (1/2-integral $J$) [22].

The tetrahedra alternate between two orientations, often called ‘A’ and ‘B’. Each spin is shared between an $A$ and a $B$ tetrahedron, and there are four tetrahedra of each type within a face-centred cubic cell. Classical spin states can be represented as kets of the form $|±±±±\rangle$ where the quantization axis of each spin lies along the direction of its 3-fold symmetry axis, which points toward or away from the centres of the tetrahedra which share the spin. For example, the ‘all-in-all-out’ state $|+++.\rangle$ has all spins pointing out of one set of tetrahedra (say the ‘A’ set) and into the other set. Two spins located on the same tetrahedron (of either orientation) are nearest neighbours.

We consider a general nearest-neighbour exchange interaction for spin-1/2 spins,

$$H_{ex} = \sum_{(i,j)} J_{ij}^{\mu\nu} S_i^\mu S_j^\nu,$$

where the sum over $(i,j)$ runs over pairs of nearest-neighbour spins and $S_i^\mu = (S_i^x, S_i^y, S_i^z)$ is the spin operator for the $i$th site. $J_{ij}^{\mu\nu}$ are exchange constants which are constrained by the space group symmetry of the crystal. In pyrochlore magnets there are only four independent parameters. It is convenient to express the Hamiltonian as

$$H = J_1 X_1 + J_2 X_2 + J_3 X_3 + J_4 X_4,$$

where $J_a$ are the exchange constants and

$$X_1 = -\frac{1}{3} \sum_{(i,j)} S_i^x S_j^x,$$

$$X_2 = -\frac{\sqrt{2}}{3} \sum_{(i,j)} [\Lambda_{xy}(S_i^x S_j^y + S_j^x S_i^y) + \text{h.c.}],$$

$$X_3 = \frac{1}{3} \sum_{(i,j)} [\Lambda_{yz}(S_i^y S_j^z + S_j^y S_i^z) + \text{h.c.}],$$

$$X_4 = -\frac{1}{6} \sum_{(i,j)} (S_i^+ S_j^- + \text{h.c.}).$$

In these expressions, the spin operators $\vec{S}_i$ are given in terms of a set of local axes, such that the local $z$ axes are the 3-fold symmetry axes described above (see [23, 24] for more details). $\Lambda_{ab}$ are phases which depend on the site numbers: $\Lambda_{12} = \Lambda_{34} = 1$ and $\Lambda_{13} = \Lambda_{24} = \Lambda_{15} = e \equiv \exp(i\frac{2\pi}{3})$, and $S_{\pm} = S_x \pm iS_y$. Note that the Hamiltonian (2) is the most general form compatible with the symmetry of the lattice for magnetic site doublets that are spin $S = 1/2$ states (i.e. one of the Kramers-type doublets). The general form of the Hamiltonian will be slightly different for the other kinds of doublets. In the special case when $J_2 = J_3 = J_4 = 0$ all states of the form $|±±±±\rangle$ (i.e. the basis kets) are eigenstates of $H$. If $J_1 > 0$ then the ground state will be the doubly degenerate ‘all-in-all-out’ states, $|+++.\rangle$ and $|--.\rangle$, otherwise the ground state is the set of highly degenerate ‘two-in-two-out’ classical spin ice states. We examine perturbations around the classical spin ice state by finding exact numerical solutions for $H$ for $J_1 < 0$ and the other exchange constants small ($|J_{2,3,4}| \ll |J_1|$), with the aim of determining the range for which the ice rule is satisfied as the system evolves into a quantum spin ice.

2.2. Finite temperature

To study the system at finite temperature we consider the density function,

$$\rho = \frac{1}{Z} \exp(-H/T) = \frac{1}{Z} \sum_i \exp(-E_i/T) |\psi_i\rangle \langle \psi_i|$$

where $|\psi_i\rangle$ are the eigenstates of $H$ with eigenvalues $E_i$ and

$$Z = \sum_i \exp(-E_i/T)$$

is the partition function.

Each basis state $|±±±±\rangle$ is a unique, classical spin configuration on the eight tetrahedra (A-type and B-type) of our 16-site system. For each state, the number of all-in-all-out tetrahedra is $N_{AA}$, the number of two-in-two-out (ice rule) tetrahedra is $N_{22}$, and the number of three-in-one-out or one-in-three-out tetrahedra is $N_{13}/N_{31}$, where $N_{AA} + N_{22} + N_{13}/N_{31} = 8$. A spin ice state satisfies the spin ice rule on all tetrahedra and will have $N_{22} = 8$; such states include states
that are superpositions of classical spin ice states. Thus the total density of each configuration is

\[ n_{\text{config}} = \sum_j \left( \frac{N_{\text{config},j}}{8} \right) |u_j\rangle \rho |u_j\rangle \]  

(5)

where \( |u_j\rangle \) are the basis kets.

2.3. Method

Generally, the Hamiltonian can be block-diagonalized into sectors classified by the irreducible representations (IR’s) of the underlying space group. These will be labelled by a \( k \)-vector to which will belong various different IR’s. In a finite system with periodic boundary conditions only a small number of \( k \)-vectors will occur. In order to fully exploit the symmetry of the system, the shape of the finite system should be chosen so that the point group symmetry is preserved along with a set of translations. Periodic boundary conditions are assumed. The smallest unit that preserves the point group symmetry is a single tetrahedron with four spins; here there are no translations and the only \( k \)-vector is \( k = (0, 0, 0) \), the \( \Gamma \)-point of the Brillouin zone. The next smallest unit has 16 spins arranged on four tetrahedra inside a cubic cell; here the IR’s belong to the \( \Gamma \)-point or the \( X \)-point, \( k = \frac{\pi}{a} (1, 0, 0) \).

The symmetry group of the sixteen-site system contains 192 elements with 14 IR’s (10 belonging to the \( \Gamma \)-point and 4 to the \( X \)-point), of which all but four are degenerate. Thus the \( 2^{16} \times 2^{16} \) Hamiltonian matrix can be block-diagonalized by an appropriate unitary transformation which is found by explicitly constructing the set of symmetrized states belonging to each IR (see appendix A). Among the \( 2^{16} \) basis kets, only 90 are pure spin ice states; linear superpositions of these form 90 symmetrized spin ice states which occur in 12 of the 14 IR’s.

3. Results

We solved the block-diagonalized \( H \) numerically for \( J_1 = -1 \) and computed the densities \( n_{\text{AAAO}}, n_{\text{2220}} \) and \( n_{31/13} \) (equation (5)) for select planes within the parameter space of \( J_2, J_3 \) and \( J_4 \). The results are shown in figures 1–6 at two different temperatures. Figures 1 and 2 show the two-in-two-out density \( n_{2220} \). A well-defined, finite region in the parameter space where the ice rule is largely satisfied is evident. Outside this region, the number of tetrahedra satisfying the ice rule is between roughly 50% and 80%.

Figures 3 and 4 show the three-in-one-out/one-in-three-out density \( n_{31/13} \) using the same parameter ranges, temperatures, and overall scale as the two-in-two-out results discussed above. Figures 5 and 6 show the all-in-all-out density \( n_{\text{AAAO}} \) for the same parameter ranges and temperatures, but a different colour scheme has been used because the maximum density is only 10% and otherwise negligible for much of the parameter space. Hence the plots of \( n_{31/13} \) mirror those of \( n_{2220} \), with a well-defined region centred at the origin where the density vanishes, and a maximum value of approximately 50% outside this region.

The origin point of the figures, corresponding to \( J_2 = J_3 = J_4 = 0 \) and \( J_1 = -1 \) can be solved analytically, yielding a 90-fold degenerate pure, classical spin ice ground state with energy \(-4/3\) and a 2680-fold degenerate first excited state with energy \(-1\). The spacing between energy levels is \(1/3\), up to the highest energy all-in-all-out state with energy \(4\). Generally, the energy of a pure, classical spin ice state is \(-N/12\), where \(N\) is the number of sites and \(N/2\) is the total number of tetrahedra, while the energy of the highest level is \(N/4\), and the level spacing is \(1/3\). At \(T = 0.05\), the value of \(n_{2220}\) is 0.99, i.e. the degenerate ground state carries nearly all of the weight at this temperature, while at \(T = 0.1\) the value of \(n_{2220}\) is reduced to 0.84. At \(T = 0.1\), the first excited state, with its large degeneracy, contributes the same weight as the ground state.
state, and the second excited state, with an even larger degeneracy of 8934, also has a non-negligible contribution to $n_{2I2O}$.

For small values of the constants $J_2$, $J_3$ and $J_4$, to zeroth order in degenerate perturbation theory, the eigenstates of the Hamiltonian will be the symmetrized states belonging to the various IR’s. These symmetrized states are, by construction, superpositions of classical states which all have the same values of $N_{AI/1O}$, $N_{2I2O}$ and $N_{31/13}$. More generally, the eigenstates of $H$ are superpositions of states with different values of $N$’s; nevertheless, the densities $n_{\text{config}}$ remain well-defined and may be calculated using equation (5).

Figure 3. The three-in-one-out/one-in-three-out density $n_{31/13}$ at $T = 0.05$.

(a) $J_1 = -1, J_4 = 0$

(b) $J_1 = -1, J_3 = 0$

(c) $J_1 = -1, J_2 = 0$

(d) $J_1 = -1$

Figure 4. The three-in-one-out/one-in-three-out density $n_{31/13}$ at $T = 0.1$.

(a) $J_1 = -1, J_4 = 0$

(b) $J_1 = -1, J_3 = 0$

(c) $J_1 = -1, J_2 = 0$

(d) $J_1 = -1$

Figure 5. The all-in-all-out density $n_{\text{AiAO}}$ at $T = 0.05$.

(a) $J_1 = -1, J_4 = 0$

(b) $J_1 = -1, J_3 = 0$

(c) $J_1 = -1, J_2 = 0$

(d) $J_1 = -1$

Figure 6. The all-in-all-out density $n_{\text{AiAO}}$ at $T = 0.1$.

The notable feature of our results is the extent of the region in which the ice-rule is obeyed for well-defined, finite ranges of the constants $J_2$, $J_3$ and $J_4$. In appendix B we show the results for a smaller system—a single tetrahedron. We find that in the smaller system the 2-in-2-out density plots are similar to those of the larger system for the parameters $J_2$ and $J_3$, but an unusual finite size effect for the single-tetrahedron occurs when $J_4$ is non-zero. This effect arises when the number of basis states is small compared to the number of IRs, which occurs in the four-site single-tetrahedron problem, but not in the larger sixteen-site problem.

In praseodymium pyrochlores the constant $J_2$ is zero because the term $X_2$ in $H$ is not invariant under time reversal.
for the non-Kramers ground state doublet of the Pr ions [25]. The phase diagram for this model was studied using mean field theory in [20], with the boundaries of the spin ice phase for similar to what we have shown in figures 1(c) and 2(c); the roughly triangular shape of the central region is similar, as are the other phase boundaries appearing outside this region. (The relations between the coupling constants used in [20] and those used in equation (2) are given in appendix C). Outside this region, [20] identifies ‘quantum’ phases associated with spins arranged perpendicular to their local z-axes, which are compatible with the superposition of three-in-one-out/one-in-three-out and two-in-two-out states evident in our results.

We can determine the location in our phase diagrams of quantum spin ice candidates using estimates of coupling constants. For Yb$_2$Ti$_2$O$_7$, the exchange constants were determined by fits to spin wave spectra in several different experiments [13, 16, 26, 27]. These experiments found consistent results for $J_2$, in the range $-3.5$ to $-3.9$ K, and for $J_3$ $\approx 1.4$ to 2.1 K, but with larger discrepancies for $J_4$ $\approx 3.5$ to 6.1 K, and even larger disagreement for $J_5$ $\approx -.9$ to $-5.9$ K. Nevertheless, all of these results correspond to points that lie well outside the central 2-in-2-out zone shown in figures 1 and 2, but with an appreciable 2-in-2-out density (about 50%).

The terbium ions in Tb$_2$Ti$_2$O$_7$ are another example of non-Kramers doublets, but mixing with excited states (within the 2$I+1$ multiplet) and a symmetry-preserving map allows for all four exchange terms to be present in the Hamiltonian. Assuming a perturbative renormalization of the coupling constants, estimates of them obtained from fits to diffuse neutron scattering measurements are: $J_1 = -5.1$ K, $J_2/|J_1| = .04$, $J_3/|J_1| = .02$ and $J_4/|J_1| = .06$ [12], which is near the centre of the region where the 2-in-2-out rule is obeyed, suggesting that this material is a perturbed spin ice.

4. Summary

Useful physical insight, in addition to computational advantages, can be gained by exploiting the high symmetry underlying pyrochlore magnets. Here we have combined this approach with numerical methods to determine precisely how the spin ice contribution to the total density function varies with the coupling constants of the most general model for nearest-neighbour spin-spin interactions. A well-defined region centred at the origin of this parameter space is the location of a phase which can be thought of as a perturbed spin ice—a phase where the 2-in-2-out ice rule is largely obeyed, with small fluctuations due to the transverse terms $X_2$, $X_3$ and $X_4$ in the nearest neighbour exchange Hamiltonian. Beyond this region, where transverse fluctuations are larger, boundaries between other phases are also evident, suggestive of competing quantum phases.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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Appendix A. Block Diagonalization

The pyrochlore crystal structure has space group symmetry $Fd\bar{3}m$ (No. 227), with octahedral point group symmetry $O_h$ and fcc lattice translations $n_1\vec{t}_1 + n_2\vec{t}_2 + n_3\vec{t}_3$, where $\vec{t}_1 = a(0,1/2,1/2)$, $\vec{t}_2 = a(1/2,0,1/2)$ and $\vec{t}_3 = a(1/2,1/2,0)$ and $n_1$, $n_2$ and $n_3$ are integers.

By considering the cubic conventional cell with periodic boundary conditions, the symmetry group used to block-diagonalize the Hamiltonian is $O_h \times \{1, t_1, t_2, t_3\}$ (where the point group $O_h$ contains non-symorphic elements), which has 192 group elements and 14 IRs. The unitary transformation that block-diagonalizes the Hamiltonian is found by generating the complete set of symmetrized, orthogonal basis states belonging to each IR. Ninety of these states will be spin ice states (superpositions of classical spin ice states that satisfy the ice-rule on every tetrahedron), which appear in nearly all the representations of the group. In addition to these spin ice states, there are many more states where the ice rule is satisfied on some of the tetrahedra that also contribute to the two-in-two-out density. These states occur in every block.

Table 1 lists the dimension (degeneracy) of each IR, as well as the size of the blocks of the Hamiltonian and the number of symmetrized spin ice states in each IR. The last column in table 1 contains a sample of our data for a specific set of coupling constants. The ground state occurs in the $A_{1g}$ block with energy $-1.43$ (in units of $J_1$). All the blocks containing spin ice states have similar smallest values, while those with no spin ice states have lowest energy values of approximately $-1.19$; there is a gap of approximately 0.2 between the spin ice states and the first set of excited states for the set of coupling constants used.
Table 1. Block-diagonalization of $H$ for 16 sites. The first column lists the IR’s of the symmetry group, the second column lists their dimension (degeneracy), the third column gives the size of each block (i.e. the number of basis states that belong to each dimension of each IR), and the fourth column lists the number of spin ice states belonging to each block. The last column lists the lowest energy eigenvalue of each block, calculated using $J_1 = -1$, $J_2 = 0.02$, and $J_3 = J_4 = 0.1$.

| IR  | Dimension | Block size | Number of spin ice states | Lowest energy in each block |
|-----|-----------|------------|---------------------------|----------------------------|
| $A_{1g}$ | 1 | 383 | 4 | $-1.43673$ |
| $A_{2g}$ | 1 | 371 | 0 | $-1.19478$ |
| $A_{1u}$ | 1 | 335 | 2 | $-1.38473$ |
| $A_{2u}$ | 1 | 335 | 0 | $-1.19904$ |
| $E_g$ | 2 | 774 | 4 | $-1.40312$ |
| $E_u$ | 2 | 682 | 2 | $-1.39776$ |
| $T_{1g}$ | 3 | 1081 | 2 | $-1.38476$ |
| $T_{2g}$ | 3 | 1085 | 3 | $-1.40894$ |
| $T_{1u}$ | 3 | 957 | 0 | $-1.17351$ |
| $T_{2u}$ | 3 | 957 | 1 | $-1.38197$ |
| $X_1$ | 0 | 2038 | 2 | $-1.38315$ |
| $X_2$ | 0 | 2042 | 4 | $-1.40764$ |
| $X_3$ | 0 | 2038 | 1 | $-1.39129$ |
| $X_4$ | 0 | 2042 | 2 | $-1.39200$ |

Table 2. Block-diagonalization of $H$ for four sites. The columns are the same as in Table 1.

| IR  | Dimension | Block size | Number of spin ice states |
|-----|-----------|------------|---------------------------|
| $A_{1g}$ | 1 | 1 | 1 |
| $A_{2g}$ | 1 | 0 | 0 |
| $A_{1u}$ | 1 | 0 | 0 |
| $A_{2u}$ | 1 | 0 | 0 |
| $E_g$ | 2 | 3 | 1 |
| $E_u$ | 2 | 0 | 0 |
| $T_{1g}$ | 3 | 2 | 1 |
| $T_{2g}$ | 3 | 1 | 0 |
| $T_{1u}$ | 3 | 0 | 0 |
| $T_{2u}$ | 3 | 0 | 0 |

Appendix B. Single Tetrahedron

For comparison, we present results for a single tetrahedron. Since periodic boundary conditions are applied, the system contains two tetrahedra and so all interactions for a single tetrahedron occur twice (effectively doubling the eigenvalues). With a Hamiltonian of size $2^4$, this problem is easy to solve numerically, and block-diagonalization yields analytic results for some sectors. Table 2 lists the IR’s for the symmetry group of a single tetrahedron, their dimensions and block size, as well as the number of spin ice states for each IR. There are only 6 spin ice states altogether. The 2-in-2-out density $n_{220}$, the three-in-one-out/one-in-three-out density $n_{31/13}$ and the all-in-all-out density $n_{440}$ (equation (5)) for a single tetrahedron are shown in figures 7 and 8, 9 and 10, at $T = 0.05$ and $T = 0.1$.

Symmetry and finite-size effects play a large role in the single tetrahedron problem, which are especially evident when $J_4 \neq 0$. To analyse this problem, we consider the four terms in $H$ separately. As discussed in section 2.1, the operator $X_1$ has only diagonal matrix elements. Because the operator $X_2$ contains single raising or lowering operators it must mix spin ice states with other states; likewise $X_3$ will have the same effect. However, $X_4$ may connect symmetrized spin ice states to either non-spin ice states or to other symmetrized spin ice states.

As we have discussed, the general Hamiltonian can be block-diagonalized into blocks with symmetrized states belonging to the different IR’s. It turns out that in the single-tetrahedron problem, after block-diagonalization, $X_4$ has only
Figure 9. The three-in-one-out/on-in-three-out density $n_{31/13}$ for a single tetrahedron at $T = 0.05$.

Figure 10. The three-in-one-out/one-in-three-out density $n_{31/13}$ for a single tetrahedron at $T = 0.1$ for select cuts within the parameter space of $J_2$, $J_3$, and $J_4$. Diagonal matrix elements, which means that $X_4$ will not mix the symmetrized spin ice states appearing singly in each block with any other state. Therefore, large enough positive or negative values of $J_4$ will produce a spin ice ground state that will cause the value of $n_{2120}$ to be very close to one, hence there is no confinement of the phase where the 2-in-2-out rule is largely obeyed for the parameter $J_4$. This does not occur in the larger (216) system because $X_4$ has non-diagonal matrix elements in each block. However the phase diagram in the space of the parameters $J_2$ and $J_3$ is very similar to the larger system.

Figure 11. The all-in-all-out density $n_{AIAO}$ for a single tetrahedron at $T = 0.05$.

Figure 12. The all-in-all-out density $n_{AIAO}$ for a single tetrahedron at $T = 0.1$.

Appendix C. Exchange constants conversion

The relations between the exchange constants in [13, 20] and those used in (2) are:

$J_1 = -3J_{zz}$ \hspace{2cm} (C1)

$J_2 = 3\sqrt{2}J_{\pm \pm}$ \hspace{2cm} (C2)

$J_3 = 3J_{\pm \pm}$ \hspace{2cm} (C3)

$J_4 = 6J_{\pm}$. \hspace{2cm} (C4)
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