Ground state properties of a fully frustrated quantum spin system

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We find that ground states of the quantum Heisenberg antiferromagnet on the geometrically frustrated pyrochlore checkerboard lattice are singlets and can be expressed in terms of positive matrices. The magnetization at zero external field vanishes for each frustrated tetrahedral unit separately and there is an upper bound of 1/8 in natural units on the susceptibility both for the ground state and at finite temperature. These results are the first exact ones in this field and generalize to some other lattices; the approach is also of interest for other spin systems.

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Geometrically frustrated spin systems are known to have many interesting properties that are quite unlike those of conventional magnetic systems or spin glasses [1]. Most results are for classical systems. The first frustrated system, for which the richness of classical ground states was noted, was the triangular lattice [2]. Subsequently, the pyrochlore lattice, which consists of tetrahedra that share sites, was identified as a lattice on which the frustration effects are especially strong [3]. Unusual low-energy properties – in particular the absence of ordering at any temperature, was predicted both for discrete [3] and continuous [4] classical spin sytems. The ground state and low energy properties of the classical pyrochlore antiferromagnet – whose quantum version is focus of this letter – has been extensivly studied in [5]. Both the interest and difficulty in studying frustrated spin systems stem from the large ground-state degeneracy, which precludes most perturbative approaches.

As is the case for most other strong interacting systems in more than one-dimension, very little is known exactly about the ground states of frustrated quantum spin systems. Most of the present knowledge has been obtained by numerics or clever approximations. Quantum fluctuations have been studied in the limts of large-S [6], where a tendency towards lifting the ground-state degeneracy in favor of an ordered state ("quantum order by disorder") was detected. In the opposite limit – $S = 1/2$, where quantum fluctuations are much stronger – the pyrochlore afm has been identified as a candidate for a quantum disordered magnet ("quantum spin liquid") [7], and it has also been discussed in terms of an RVB approach [8]. However, there are no exact results against which to test the reliability of the results in this limit. In contrast to this, for conventional – bipartite – antiferromagnetic spin systems it is well known, for example, that the energy levels are ordered in a natural way according to spin, starting from spin zero [9]. Geometrically frustrated systems are not bipartite and thus this otherwise quite general theorem does not apply.

In this letter, we find some first exact results for the fully frustrated quantum antiferromagnet on a pyrochlore checkerboard, with the help of the reflection symmetry of this two dimensional lattice. We shall establish rigorously that there is always a ground state with total spin zero. Furthermore, in the periodic case all ground states (if there is more than one) have total spin zero, the spin-expectation vanishes for each frustrated unit (this is the quantum analog of the "*ice rule*" for the Ising system), and there are concrete upper bounds on the susceptibility.

Geometric frustration occurs typically for spin systems with interactions that favor anti-alignment and involve fully connected units of three or more spins, that can obviously not all be mutually anti-aligned. The kagome lattice is an example of frustrated spin systems with site-sharing triangular units, the pyrochlore lattice and its two-dimensional version, the pyrochlore checkerboard are examples with site-sharing tetrahedra. We shall focus on the latter; it is a two dimensional array of site-sharing tetrahedra, whose projection onto a plane is a square lattice with two extra diagonal bonds on every other square. (The regular pyrochlore lattice is a three-dimensional array of site-sharing tetrahedra.) The tetrahedra – or squares with extra diagonal bonds – are the frustrated units and will henceforth be called *boxes*.

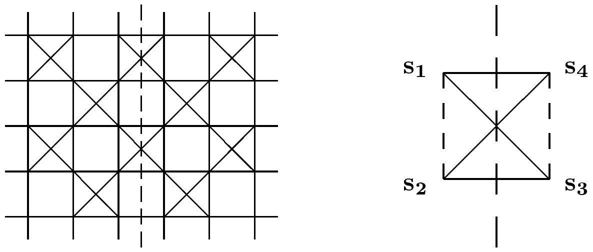

FIG. 1. (a) pyrochlore checkerboard, reflection symmetric about dashed line (b) frustrated unit with crossing bonds

The hamiltonian of a quantum Heisenberg antiferromagnet on a general lattice is (in natural units)

$$H_{\rm AF} = \sum_{\langle i,j\rangle} \mathbf{s_i} \cdot \mathbf{s_j}, \tag{1}$$

where the sum is over bonds $\langle i,j\rangle$ that connect sites i and j and $\mathbf{s} = (s^1, s^2, s^3)$ are spin operators in the spin-s representation, where s can be anything. For the checkerboard lattice the hamiltonian is up to a constant equal to half the sum of the total spin squared of all boxes (labelled by x)

$$H = \frac{1}{2}\sum_x (\mathbf{s_1}+\mathbf{s_2}+\mathbf{s_3}+\mathbf{s_4})_x^2. \qquad (2)$$

A 3×3 checkerboard with periodic boundary conditions, i.e., with four independent sites, provides the simplest example. It has a hamiltonian that is (up to a constant) the total spin squared of one box and the energy levels, degeneracies, and eigenstates follow from the decompostion of $[s]^{\otimes 4}$ into components of total spin; all ground states have total spin zero and there are $2s+1$ of them.

A checkerboard lattice of arbitrary size, with or without periodic boundary conditions but with an even number of independent sites, has the property that it can be split into two equal parts that are mirror images of one another about a line that cuts bonds, as indicated in figure 1, and that contains no sites. We shall now show that such a system has at least one spin-zero ground state. It is actually not important, for the following argument, what the lattice looks like on the left or right; these sublattices neither need to be checkerboards nor do they have to be purely antiferromagnetic (as long as total spin is a good quantum number). What is impotant is, that the whole system is reflection symmetric about the line that separates left and right and that the crossing bonds are of checkerboard type. (For a system with periodic boundary conditions in one direction there will actually be two such lines, but we emphasize that PBC is not needed here even though it is needed in the usual reflection positivity applications; see [10] and references therein.) A key observation is that these crossing bonds (solid lines in figure 1(b)) form antiferromagnetic bonds $\mathbf{S_L}\cdot\mathbf{S_R}$ between pairs of spins $\mathbf{S_L}=\mathbf{s_1}+\mathbf{s_2}$ and $\mathbf{S_R}=\mathbf{s_3}+\mathbf{s_4}$ of each box on the symmetry line.

The hamiltonian is $H=H_L+H_R+H_C$, where H_L and H_R act solely in the Hilbert spaces of the left, respectively right, subsystem and H_C contains the crossing bonds. For the checkerboard $H_C=\sum_y((\mathbf{s_1}+\mathbf{s_2})\cdot(\mathbf{s_3}+\mathbf{s_4}))_y$, with the sum over boxes y that are bisected by the symmetry line. H_L and H_R are completely arbitrary as long as they commute with the total spin operator. We will, however, assume here that they are real in the S^3 basis. Any state of the system can be written in terms of a matrix c as

$$\psi = \sum_{\alpha,\beta} c_{\alpha\beta}\psi^L_\alpha \otimes (\psi^R_\beta)_{\rm rot}, \qquad (3)$$

where the ψ^L_α form a real orthonormal basis of S^3 eigenstates for the left subsystem and the $(\psi^R_\beta)_{\rm rot}$ are the corresponding states for the right subsystem, but rotated by an angle π around the 2-direction in spin space. This rotation takes $\uparrow$ into $\downarrow$, $\downarrow$ into $-\uparrow$, and more generally $|s,m\rangle$ into $(-)^{s-m}|s,-m\rangle$. It reverses the signs of the operators S^1 and S^3, while it keeps S^2 unchanged. The eigenvalue problem $H\psi=E\psi$ is now a matrix equation

$$h_L c + c(h_R)^T - \sum_{i=1}^3\sum_y t_y^{(i)} c (t_y^{(i)})^T = Ec, \qquad (4)$$

where $(h_L)_{\alpha\beta}$ and $(h_R)_{\alpha\beta}$ are real, symmetric matrix elements of the corrosponding terms in the hamiltonian and the $t_y^{(i)}$ are the *real* matrices defined for the spin operators $\mathbf{s_1}$ and $\mathbf{s_2}$ in box y by $t^{(1,3)}_{\alpha\beta}=\langle\psi^L_\alpha|s_1^{(1,3)}+s_2^{(1,3)}|\psi^L_\beta\rangle$ and $t^{(2)}_{\alpha\beta}=i\langle\psi^L_\alpha|s_1^{(2)}+s_2^{(2)}|\psi^L_\beta\rangle$. Note the overall minus sign of the crossing term in (4): replacing $\mathbf{s_1}+\mathbf{s_2}$ by $\mathbf{s_3}+\mathbf{s_4}$ and ψ^L by $(\psi^R)_{\rm rot}$ gives a change of sign for directions 1 and 3, while the i in the definition of $t^{(2)}$ gives the minus sign for direction 2.

Consider now the energy expectation in terms of c:

$$\langle\psi|H|\psi\rangle = {\rm tr}\, cc^\dagger h_L + {\rm tr}\, c^\dagger c h_R - \sum_{i,y}{\rm tr}\, c^\dagger t_y^{(i)} c (t_y^{(i)})^\dagger. \quad (5)$$

Since H is left-right symmetric and by assumption real, we find that for an eigenstate of H with coefficient matrix c, there is also an eigenstate with matrix $c^\dagger$ and, by linearity, with $c+c^\dagger$ and $i(c-c^\dagger)$. Without loss of generality we may, therefore, write eigenstates of H in terms of Hermitean $c=c^\dagger$. We shall also take ψ to be normalized: $\langle\psi|\psi\rangle={\rm tr}\, c^\dagger c=1$. Following [11], let us write the trace in the last term of (5) in the diagonal basis of c: $-{\rm tr}\, c^\dagger t_y^{(i)} c (t_y^{(i)})^\dagger = -\sum_{k,l} c_k c_l |(t_y^{(i)})_{kl}|^2$. This expression clearly does not increase if we replace all the c_k by their absolute values $|c_k|$, i.e., if we replace the matrix c by the positive semidefinite matrix $|c|=\sqrt{c^2}$. The first two terms in (5) and the norm of ψ remain unchanged under this operation. We conclude that if c is a ground state than so is $|c|$. Since $c=c^+-c^-$ and $|c|=c^++c^-$, with p.s.d. c^+ and c^-, we may, in fact, chose a basis of ground states with p.s.d. coefficient matrices.

Next, we will show that the state ψ_0 with the unit matrix as coefficient matrix (in the S^3 eigenbasis) has total spin zero. Since the overlap of ψ_0 with a state with matrix c is simply the trace of c, which is neccessarily non-zero for states with a p.s.d. matrix, and because spin is a good quantum number of the problem, this will imply that there is a least one ground state with total spin zero. First consider a spin-1/2 system. In the S^3 eigenbasis every site has then either spin up or down. The state with unit coefficient matrix is a tensor product of singlets on corresponding pairs of sites $i\in L$, $i'\in R$ of the two sublattices:

$$\psi_0 = \bigotimes_{i\in L}(\uparrow(\uparrow)_{\rm rot} + \downarrow(\downarrow)_{\rm rot})_{ii'} = \bigotimes_{i\in L}(\uparrow\downarrow - \downarrow\uparrow)_{ii'}. \quad (6)$$

The analogous state for a system with arbitrary spins,

$$\psi_0 = \bigotimes_{i\in L}\sum_{m=-s}^{s}\left((-)^{s-m}|s,m\rangle\otimes|s,-m\rangle\right)_{ii'}, \qquad (7)$$

is also a tensor product of spin-zero states.

Finally, we would like to show that the projection onto the spin zero part of a state with p.s.d. coefficient matrix preserves its positivity. This is only of academic interest here, but it is non-trivial and may very well be important for other physical questions. To find the projection onto spin zero we need to decompose the whole Hilbert space into tensor products of the spin components $[j]_k \otimes [j']_{k'}$ of the two subsystems; here k, k' are additional quantum numbers that distinguish multiple multiplets with the same spin j. Only tensor products with $j = j'$ can have a spin zero subspace, which is unique, in fact, and generated by the spin zero state $\sum_{m=-j}^{j} |j,m,k\rangle \otimes (-)^{j-m}|j,-m,k'\rangle$. Noting that $(-)^{j-m}|j,-m,k'\rangle$ is precisely the spin-rotated state $(|j,m,k'\rangle)_{\text{rot}}$, we convince ourselves that the projection onto spin zero amounts to a partial trace over m in a suitably parametrized matrix c. This operation preserves positive semidefiniteness, so we actually proved that the checkerboard has at least one ground state that has both total spin zero *and* a p.s.d. coefficient matrix c.

We do not know how many ground states there are. To determine the spin of any remaining ground states we add an external field to the hamiltonian and study the resulting magnetization. We will see that the spontaneous magnetization of every box on the symmetry line vanishes for all ground states, and thus if we have periodic boundary conditions in at least one direction, the total magnetization vanishes. Since S^3_{tot} is a good quantum number and $S^{\pm}_{\text{tot}}$ commute with the hamiltonian, this will imply that all ground states in such a system have total spin zero. Let us thus modify the original hamiltonian (2) by replacing the term $(s_1^{(3)} + s_2^{(3)} + s_3^{(3)} + s_4^{(3)})_z^2$ for a single box, z, on the symmetry line by $(s_1^{(3)} + s_2^{(3)} + s_3^{(3)} + s_4^{(3)} - b)_z^2$, i.e., effectively adding a field b to the spins in box z and a constant term $\frac{1}{2}b^2$ to the hamiltonian. We want to distribute the resulting b-terms $(s_1^{(3)}+s_2^{(3)}-b/2)^2$, $(s_3^{(3)}+s_4^{(3)}-b/2)^2$, and $2(s_1^{(3)}+s_2^{(3)}-b/2)(s_3^{(3)}+s_4^{(3)}-b/2)$ to H_L, H_R, H_C respectively. We cannot use the spin rotation as before, because the crossing terms in the hamiltonian would no longer be left-right symmetric in the basis (3). To avoid this problem we will, instead, expand eigenstates ψ in the same basis on the left and on the right:

$$\psi = \sum_{\alpha,\beta} \tilde{c}_{\alpha\beta} \psi^L_\alpha \otimes \psi^R_\beta. \tag{8}$$

In this basis the hamiltonian is left-right symmetric and we may assume, as before, that $\tilde{c} = \tilde{c}^\dagger$. Except for the presence of b in box z the energy expectations on the left and right are as before. The energy expectation of the crossing terms of box z in the diagonal basis of $\tilde{c}$ is now

$$\sum_{k,l} \tilde{c}_k \tilde{c}_l \left(|(t_z^{(1)})_{kl}|^2 - |(t_z^{(2)})_{kl}|^2 + |(t_z^{(3)})_{kl} - b/2|^2 \right).$$

This expression clearly does not increase if we replace the c_k by their absolute value $|c_k|$ *and* change the signs of the first and last terms. The sign change can be achieved by simultaneously performing a spin rotation and changing the sign of the field b in the right subsystem. This actually completly removes b from the hamiltonian. We have thus shown that the ground state energies of the systems H_b with and H_0 without the b-terms satisfy the inequality $E_b \geq E_0$. Let $|b\rangle$ be a ground state of H_b and $|0\rangle$ a ground state of H_0. It follows from the variational principle, that $\langle 0|H_b|0\rangle \geq \langle b|H_b|b\rangle = E_b \geq E_0$. Expressed in terms of spin operators this reads $E_0 - 2\langle 0|b(s_1^{(3)}+s_2^{(3)}+s_3^{(3)}+s_4^{(3)})_z|0\rangle + b^2 \geq E_0$. Recalling that we are free to choose both the sign and the magnitude of b we find that the ground state magnetization of box z must be zero:

$$\langle 0|(s_1^{(3)} + s_2^{(3)} + s_3^{(3)} + s_4^{(3)})_z|0\rangle = 0. \tag{9}$$

This quantum analog of the "*ice rule*" is true for any box on the symmetry line and it holds for all three spin components. In a system with periodic boundary conditions and an even number of sites in at least one direction we can choose the symmetry line(s) to intersect any given box, so in such a system the magnetization is zero both for every single box separately and also for the whole system: $\langle 0|S^{(3)}_{\text{tot}}|0\rangle = 0$. As mentioned previously this implies that the total spin is zero for all ground states of such a system.

Let us return to the inequality $E_b \geq E_0$. It implies a bound on the local susceptibility of the system: Let $E(b) \equiv \langle b|H_0 - bS^{(3)}_{\text{box}}|b\rangle$ be the ground state energy of the periodic pyrochlore checkerboard with a single box immersed in an external field b. Recalling $H_b = H_0 - bS^{(3)}_{\text{box}} + \frac{1}{2}b^2$, we see that the above inequality becomes $E(b) + \frac{1}{2}b^2 \geq E(0)$ and, assuming differentiability, implies an upper bound on the susceptibility at zero field for single-box magnetization

$$\chi_{\text{loc}} = -\frac{1}{\lambda} \left. \frac{\partial^2 E(b)}{\partial b^2} \right|_{b=0} \leq \frac{1}{4}, \tag{10}$$

where $\lambda = 4$ is the number of spins in a box. (The susceptibility is given in natural units in which we have absorbed the g-factor and Bohr magneton in the definition of the field b.)

We would like to get more detailed information about the response of the spin system to a global field $\{b_x\}$ in a hamiltonian $H_{\{b_x\}}$ which is identical to (2), except for the terms for the third spin component, which are replaced by $(s_1^{(3)} + s_2^{(3)} + s_3^{(3)} + s_4^{(3)} - b_x)_x^2$. From what we have seen so far, it is apparent that the corresponding ground state energy $E_{\{b_x\}}$ is extremal for $b_x = 0$. With the help of a more sophisticated trace inequality [13], that becomes relevant whenever the matrix c in (3) cannot be diagonalized, one can actually show that $E_{\{b_x\}}$ has an absolute minimum at $b_x = 0$:

$$E_{\{b_x\}} \geq E_0. \tag{11}$$

Note that we had to put the field on the boxes for this result to hold; not every field on the individual spins can be written this way. The special choice $b_x = B/2$ corresponds to a global homogenous field B on all spins. (The factor $1/2$ adjusts for the fact that every spin is shared by two boxes.) If $E(B) = \langle B|H_0 - BS^{(3)}_{\rm tot}|B\rangle$ is the ground state energy of the periodic pyrochlore checkerboard in the external field B, then (11) implies $E(B) + \frac{\Lambda}{16}B^2 \geq E(0)$, and thus an upper bound on the susceptibility per site at zero field (in natural units)

$$\chi = -\frac{1}{\Lambda}\left.\frac{\partial^2 E(B)}{\partial B^2}\right|_{B=0} \leq \frac{1}{8}, \tag{12}$$

where Λ is the number of independent sites, which equals twice the number of boxes.

All these results continue to hold at finite temperature. The analog of (11) holds also for the partition function corresponding to $H_{\{b_x\}}$:

$$Z_{\{b_x\}} \leq Z_0, \tag{13}$$

as can be shown by a straightforward application of lemma 4.1 in section 4 of [10] to the pyrochlore checkerboard. The physically relevant partition function for the periodic pyrochlore checkerboard at finite temperature in a homogenous external field, $Z(B) \equiv \operatorname{tr} e^{-\beta(H_0 - BS^{(3)}_{\rm tot})}$, differs from $Z_{\{b_x\}}$, where $b_x = B/2$, only by a factor corresponding to the constant term in $H_{\{b_x\}}$. Due to (13), the free energy $F(B) = -\beta^{-1}\ln Z(B)$ satisfies

$$F(B) + \frac{\Lambda}{16}B^2 \geq F(0). \tag{14}$$

This implies (i) that the magnetization at zero field is still zero at finite temperature,

$$M_T = -\frac{1}{\Lambda}\left.\frac{\partial F(B)}{\partial B}\right|_{B=0} = 0, \tag{15}$$

and, more interestingly, (ii) the same upper bound for the susceptibility per site at zero field as we had for the ground state:

$$\chi_T = -\frac{1}{\Lambda}\left.\frac{\partial^2 F(B)}{\partial B^2}\right|_{B=0} \leq \frac{1}{8}. \tag{16}$$

The bounds on the susceptibility hold for arbitrary intrinsic spin-s and agree very well with the results of [12] for the classical pyrochlore antiferromagnet in the undiluted case.

It is not essential for our method that only every other square of the pyrochlore checkerboard is a frustrated unit, only the reflection symmmetry and the antiferromagnetic crossing bonds are important. We could, e.g., have diagonal bonds on *every* square, but then the horizontal and/or vertical bonds must have twice the coupling strength. Our results also apply to various 3-dimensional cubic versions of the checkerboard, e.g., with diagonal crossing bonds in every other cube. While the method does not directly work for the 3D pyrochlore lattice because its geometry is too complicated, it has been seen in [5] that classically this system has similar properties to the pyrochlore checkerboard, which is also fully frustrated, and has the added advantage of being more easily visualizeable.

We would like to thank Roderich Moessner for bringing this problem to our attention, for explaining his work on the classical pyrochlore antiferromagnet, and for numerous helpful discussions and suggestions.

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[1] For a collection of articles, see *Magnetic systems with competing interactions: frustrated spin systems*, edited by H. T. Diep (World Scientific, Singapore, 1994); for Ising systems, see: R. Liebmann, *Statistical Mechanics of Periodic Frustrated Ising Systems* (Springer, Berlin, 1986).

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