Residual entropy and spin gap in a one-dimensional analog of the pyrochlore antiferromagnet

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We show that the low-energy sector of the S=1/2, antiferromagnetic Heisenberg model on a one-dimensional lattice of coupled tetrahedra consists of $2^N$ replica of the spectrum of the dimerized Heisenberg chain, where $N$ is the number of tetrahedra. This provides a proof of the following properties: i) there is a residual ground-state entropy per spin equal to $2^{1/2}$; ii) there is a singlet-triplet gap as long as the coupling between the tetrahedra is smaller than the internal one. These properties are compared to available results on the pyrochlore lattice.

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It is by now quite clear that the low-energy properties of frustrated magnets can be very different from those of ordinary magnets with long-range order. In particular, after the pioneering work of Majumdar and Ghosh on the zigzag chain [1], it has been shown that several systems have a singlet-triplet gap in the magnetic spectrum. This property is actually not specific of frustrated systems since it is also shared by ladders with an even number of legs [2]. There is an increasing evidence however that frustration can have more specific consequences, like for instance low-lying singlets in the singlet-triplet gap. The first example was again the zigzag chain which, contrary to ladders, has a two-fold degenerate singlet ground-state [1]. But the spectrum can be more complex. For instance, there is a clear numerical evidence that the Heisenberg antiferromagnet on the Kagome lattice has an exponential number of low lying singlets below the first triplet excitation [3]. If analytical arguments have been put forward to explain this property in terms of coupled triangles [4], an analytical proof that this is the case is still lacking. Such properties are actually not limited to low-dimensional systems. In particular, the Heisenberg model on the pyrochlore lattice is a well known example of 3D frustrated model without long-range order at the classical level [5], and the S=1/2, quantum version is believed to have a very short correlation length and a singlet-triplet gap in the ground-state [6]. There is also some indication that there are low-lying singlet states in the singlet-triplet gap [7], but again there is no proof that this is indeed the case.

In this paper, we show that these properties actually occur in a one-dimensional analog of the pyrochlore lattice, namely a one-dimensional array of coupled tetrahedra. The discussion can actually be carried out for the slightly more general situation of a one dimensional system of alternating spins and triangles (see Fig. 1) defined by the Hamiltonian:

$$\mathcal{H} = J_1 \sum_i (\vec{S}_{2,i} \cdot \vec{S}_{3,i} + \vec{S}_{3,i} \cdot \vec{S}_{4,i} + \vec{S}_{4,i} \cdot \vec{S}_{2,i}) + J_2 \sum_i (\vec{S}_{2,i} + \vec{S}_{3,i} + \vec{S}_{4,i}) + J_3 \sum_i (\vec{S}_{2,i} + \vec{S}_{3,i} + \vec{S}_{4,i}) \cdot \vec{S}_{1,i+1}$$

(1)

where $\vec{S}_{n,i}$ are spin 1/2 operators. Similar models involving more than one spin at a given site have been studied [8–11], but to our knowledge none of them dealt with triangles, and the properties that are described below are very specific to that case.

FIG. 1. System of alternating spins (1, i) and triangles $T_i = \{(2, i), (3, i), (4, i)\}$. The notations for the couplings are: $J_1$ for bonds inside triangles and $J_2$ (respectively $J_3$) between a triangle $T_i$ and its left (right) spin neighbor $S_{1,i}$ ($S_{1,i+1}$).

The most important step toward a solution of this Hamiltonian is to realize that the spins of the triangle of any unit cell $i$ enter the Hamiltonian only through their sum $\vec{T}_i = \vec{S}_{2,i} + \vec{S}_{3,i} + \vec{S}_{4,i}$ since the first term can be rewritten $(J/2) \sum_i (\vec{T}_i^2 - 9/4)$. This observation has two consequences. First of all, it means that the total spin of the triangle of any unit cell, which can take the degenerate value 3/2 or the twofold degenerate value 1/2, is a good quantum number. Secondly, it shows that the additional quantum number needed to specify the states in the subspace of total spin 1/2 - for instance the chirality - does not enter the Hamiltonian.

So the eigenvalues of the problem of Eq. (1) are the same as those of the following family of effective Hamil-
tonians $\mathcal{H}(\{T_i\})$: \[
\mathcal{H}(\{T_i\}) = \sum_i (J_2 \vec{S}_{i,1} \vec{T}_i + J_3 \vec{T}_i \vec{S}_{i,1,i+1}) + (J_1/2) \sum_i (\vec{T}_i^2 - 9/4) \tag{2}
\]
where $\vec{T}_i^2 = T_i(T_i + 1)$ and $T_i = 1/2$ or 3/2, the degeneracy of the eigenvalues of $\mathcal{H}(\{T_i\})$ for the original problem being equal to $2^{n_{1/2}}$, where $n_{1/2}$ is the number of triangles with total spin 1/2.

The twofold degeneracy associated with each triangle in a doublet state can actually be understood in a more direct way. Let us consider the eigenstates of the problem obtained by considering only one spin in the triangle of unit cell $i$, say $\vec{S}_{2,i}$. Then the wave functions obtained as the product of the eigenstates of that new problem with the singlet constructed out of $\vec{S}_{3,i}$ and $\vec{S}_{4,i}$ are eigenstates of the original problem since all the extra couplings connect a single spin to both ends of that singlet. Besides, $\vec{S}_{3,i}$ and $\vec{S}_{4,i}$ forming a singlet, these eigenstates correspond to $T_i = 1/2$. Finally, the wave functions obtained by putting the singlet on the three possible bonds of one triangle are not linearly independent but generate a space of dimension 2, as for a single triangle.

So the problem has now been split into different sectors corresponding to the values of the $T_i$’s. For clarity, we will continue the discussion in two specific situations: i) Alternating spins 1/2 and triangles, where the analysis is particularly straightforward; ii) Coupled tetrahedra, which is physically more relevant as an analog of the pyrochlore antiferromagnet.

I. ALTERNATING SPINS 1/2 AND TRIANGLES

This case corresponds to $J_2 = J_3$. For clarity, we rename the parameters $J_1 = J$ and $J_2 = J_3 = J'$. So we are dealing in this section with triangles of strength $J$ coupled to spins 1/2 with 3 exchange integrals of strength $J'$. In the limit where $J'/J$ goes to zero, the spectrum of each sector $T_i$ is completely degenerate, and the energy is given by the sum of the energies of the triangles $E(\{T_i\}) = -(3/2)(N - \sum_i T_i)$, where $N$ is the number of unit cells. In that limit, the lowest energy states are those obtained when all the triangles are in a doublet. Perturbative arguments show that this will remain true up to a certain value of the ratio $J'/J$. To study that problem quantitatively, we have performed exact diagonalizations of finite clusters for the Hamiltonians of Eq. (2). The results are given in Fig. 3.

Finite size analysis. It is well known that the ground state energy per site of the spin 1/2 Heisenberg model scales like $\varepsilon_g = e_\infty - A/L^2$ where $e_\infty = 1/4 - \ln 2$. So the ground state energy per site in units of $J$ for $\mathcal{H}(\{T_i = 1/2\})$, which is nothing but the standard Heisenberg model with coupling $J'$, is constant up to a constant of order $J$, scales like $\varepsilon_g^{(0)} = -3/16 + (J'/2J)e_\infty - A/L^2$ where $L = 4N$ is the total number of sites of the system. Let us focus now on $\varepsilon_g^{(1)}$, the GS per site of $\mathcal{H}(\{T_{i\neq i_0} = 1/2, T_{i_0} = 3/2\})$, and let us denote by $\delta_\infty$ the thermodynamic limit of the energy difference between the GS energy of the spin 1/2 Heisenberg model and the GS energy of the same model with one spin 1/2 replaced by a spin 3/2. Since there are no 1/L corrections for $\varepsilon_g^{(0)}$, one should expect the following scaling for $\varepsilon_g^{(1)}$, \[
\varepsilon_g^{(1)} = \varepsilon_g^{(0)} + \Delta_\infty/L + \mathcal{O}(1/L^2) \tag{3}
\]
where $\Delta_\infty = (3 - (J'/J)\delta_\infty)/2$ is the thermodynamic limit of the energy difference between the GS of $\mathcal{H}(\{T_{i\neq i_0} = 1/2, T_{i_0} = 3/2\})$ and $\mathcal{H}(\{T_i = 1/2\})$. Numerical simulations up to $L = 28$ with one spin 3/2 show that the scaling (3) is very well verified and allowed us to extract the gap (see Fig. 2) which, as expected, is linear in $J'$, and $\delta_\infty \simeq 5/2$.

![FIG. 2. Main figure. Ground state energy per site of $\mathcal{H}(\{T_{i\neq i_0} = 1/2, T_{i_0} = 3/2\})$ for a $L = 16$ cluster (i.e. 4 tetrahedra) for different values of the number of triangles $T_i = 3/2$: (●) $n_{3/2} = 0$, (■) $n_{3/2} = 1$, (▲) $n_{3/2} = 2$ neighboring triangles, (★) $n_{3/2} = 3$, (▼) $n_{3/2} = 4$. Inset. Thermodynamic limit of the energy difference between the GS of $\mathcal{H}(\{T_{i\neq i_0} = 1/2, T_{i_0} = 3/2\})$ and $\mathcal{H}(\{T_i = 1/2\})$. Up to $J'/J \approx 1.2$ the low energy physics of the model is given by the spin 1/2 Heisenberg model.](image_url)

Discussion. It is then clear that the ground state remains in the sector $\{T_i = 1/2\}$ up to $J'/J \simeq 1.2J$, and in particular for the isotropic point $J' = J$. When this is the case, the low energy physics is given by the Hamiltonian of Eq. (2) with all $T_i$’s equal 1/2, which, as we saw, is nothing but the one-dimensional, spin 1/2 Heisenberg model with coupling $J'$. The only difference is that we now deal with $2^N$ replica of the spectrum. All properties of interest can be deduced from this mapping. In particular, there is no singlet-triplet gap in the spectrum, the elementary excitations are deconfined spinons, and there is a residual entropy per spin equal to $2^J/4$. 

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II. COUPLED TETRAHEDRA

This case corresponds to \( J_2 = J_1 \). For clarity we use the notations \( J_2 = J_1 = J \) and \( J_3 = J' \) in the following. So we are now dealing with tetrahedra of strength \( J \) coupled by bonds of strength \( J' \). Let us start from the limit \( J' = 0 \). In that case, the groundstate is obtained by putting each tetrahedron in its groundstate. Since the ground state of a tetrahedron is twofold degenerate with energy \(-3/2J\) and singlet-triplet gap of \( J \), the groundstate is \( 2^N \)-fold degenerate with energy \(-3/2NJ\). The low lying excited states correspond to putting one tetrahedron in its first excited state with an energy cost of \( J \). With respect to the general classification of the states proposed at the beginning of the paper, the situation is slightly more complicated than in the previous case. First of all, the groundstate manifold in the limit \( J' = 0 \), of dimension \( 2^N \), contains only some of the eigenstates of \( H(T_i = 1/2) \), the Hilbert space of that Hamiltonian being of dimension \( 2^{3N} \). Besides, the manifold of the first excitation of energy \( J \) contains states corresponding to different effective Hamiltonians. This comes from the fact that the triplet excitation of a tetrahedron is threefold degenerate, and that only two of them can be constructed with a given triangle being a doublet, the other one corresponding to the groundstate of a spin 1/2 coupled to the spin 3/2 state of a triangle. So it is no longer clear a priori that the low energy sector can be described by only one of the effective Hamiltonians of Eq. (2).

To address this point, we have again resorted to exact diagonalizations of finite clusters, and we have determined which effective Hamiltonian gives the ground-state and the first triplet excited state as a function of \( J' \). The results are given in Fig. 3. As in the previous case, the groundstate is given by \( H(T_i = 1/2) \) regardless of the size of the system up to very large values of \( J' \), and in particular beyond the isotropic limit \( J' = J \). For the first excited state, the situation is slightly more involved since \( H(T_i = 1/2) \) is now equivalent to the dimerized Heisenberg model and has a gap if \( J' \neq J \) [13]. For small enough \( J' \), the first excited state is also given by \( H(T_i = 1/2) \) for all sizes. However, this is no longer true if \( J' \) is beyond a certain value \( J'(N) \) which increases with the size of the system. Since we are interested in the properties of the system in the thermodynamic limit, we have performed a finite size analysis of the first excited state of \( H(T_i = 1/2) \) and of the GS of \( H \). (see results in Fig. 3).

Finite size analysis. Contrary to the situation discussed in the previous section, there are exponential corrections to the finite size energies if \( J' \neq J \) because there is then gap in the spectrum of \( H(T_i = 1/2) \). Namely, with the same notations as in section I,

\[
\varepsilon^{(1)}_L = \varepsilon^{(0)}_\infty + \Delta_\infty/L - (A/L^2)e^{-L/\xi}
\]

Such a scaling for clusters up to \( L = 28 \) with one spin 3/2 in the system allows the determination of \( \Delta_\infty(T_i = 1/2, T_{i0} = 3/2) \) which remains positive up to very large values of \( J' \) (at least 2J). Let us turn to the case of the first excited state of \( H(T_i = 1/2) \). Again, a scaling of the type \( \Delta_L = \Delta_\infty + (A/L)e^{-L/\xi} \) is expected with \( \xi \) diverging and \( \Delta_\infty(T_i = 1/2) = 0 \) at the isotropic point \( J' = J \). Since the systems contains only spin 1/2 is that sector we performed diagonalizations up to \( L = 48 \) to calculate \( \Delta_\infty \). The results (see Fig. 3) clearly agree with what was expected even if the precise behavior of \( \Delta_\infty \sim \delta^{2/3} \) [12] (up to logarithmic corrections) as the dimerization \( \delta = (1-J'/J)/(1+J'/J) \) goes to zero is difficult to extract from numerical simulations.

Discussion. The results are quite clear: For large systems, \( J'_c \) is larger than \( J \), namely \( J'_c \approx 1.15J \). Then to understand the low energy properties of the model in the parameter range \( J' \leq J \) and in the thermodynamic limit, one can use \( H(T_i = 1/2) \) as an effective Hamiltonian. This means that the low energy physics can be described by the dimerized, spin 1/2 Heisenberg chain with alternating exchange integrals \( J \) and \( J' \). This model has been extensively studied in the context of spin-Peierls systems [13]. The main property, already used to perform the finite-scaling, is that there is a singlet-triplet gap in the system as long as \( J' < J \), and that this energy gap closes at the point \( J' = J \). Besides, the elementary excitations are boundstates of spinons due to the confinement potential introduced by the dimerization [13]. Regarding the properties of the original problem, one should not forget that all these states are degenerate, and in particular that there is, as in the previous case, a residual entropy per spin equal to \( 2^{1/4} \).
III. DISCUSSION

The main motivation in undertaking the present study was to shed some light on the properties of the pyrochlore antiferromagnet. This system is a three-dimensional structure that can be thought of as an array of coupled tetrahedra. The current situation as far as a theoretical understanding of that model is concerned is the following: There is a clear evidence that the spin-spin correlation functions are extremely short ranged, and there is some numerical evidence that there is a singlet-triplet gap in the spectrum \[ J' < J \]. Both properties are indeed satisfied by the present 1D model of coupled tetrahedra in the parameter range \( J' < J \). In particular, since the ground states can be written as products of local singlets, the spin-spin correlation functions indeed decay very fast. The fact that the gap closes when \( J' = J \) is not really inconsistent with the case of the pyrochlore since there is an important difference in the way tetrahedra are coupled in both cases: Pairs of tetrahedra are never coupled through more than one spin on each of them in the pyrochlore structure whereas one of the tetrahedra is coupled through 3 spins in the present model. So \( J' = J \) in the present case somewhat corresponds to a stronger coupling than for the pyrochlore.

More importantly, the presence of a residual entropy per spin in the model of the present paper suggests that, if there is a singlet-triplet gap in the spectrum, there should indeed be low-lying singlets within this gap. This should be particularly easy to detect if one goes away from the standard pyrochlore and considers a dimerized version of the same model with weakly coupled tetrahedra since the gap becomes larger in that limit while at the same time the splitting between the singlet states decreases. Work is in progress along these lines.

To summarize, we have proposed and solved a one-dimensional analog of the pyrochlore antiferromagnet and proved that it exhibits at least some of the exotic physics one can hope to find in very frustrated magnets, namely: A singlet-triplet gap, and a lot of low-lying singlets. Given the relative simplicity of the model, it is the authors hope that some compound can be synthesized with this kind of structure. In any case, the very simple picture of quite unusual properties provided by this model is expected to serve as a useful guide in the search of new experimental realizations of very frustrated magnets.

The numerical simulations were performed on the Cray supercomputers of the IDRIS (Orsay, France).

[1] C. K. Majumdar and D. Ghosh, J. Math. Phys. 10, 1388 (1969).
[2] For a review, see E. Dagotto and T. M. Rice, Science 271, 618 (1996).
[3] P. Lecheminant, B. Bernu, C. Lhuillier, L. Pierre and P. Sindzingre, Phys. Rev. B 56, 2521 (1997).
[4] C. Waldtmann, H.-U. Everts, B. Bernu, C. Lhuillier, P. Sindzingre, P. Lecheminant, L. Pierre, Eur. Phys. J. B 2, 501 (1998).
[5] F. Mila, Phys. Rev. Lett. 81, 2356 (1998).
[6] A. V. Chubukov, Phys. Rev. Lett. 69, 832 (1992); A. B. Harris, C. Kallin and A. J. Berlinsky, Phys. Rev. B 45, 2899 (1992); P. Chandra, P. Coleman and I. Ritchey, J. Phys. I (France) 3, 591 (1993).
[7] B. Canals and C. Lacroix, Phys. Rev. Lett. 80, 2933 (1998).
[8] A. K. Kolezhuk and H.-J. Mikeska, Phys. Rev. B 56, R11380 (1997).
[9] H. Niggemann, G. Uimin and J. Zittartz, J. Phys.: Condens. Matter 9 (1997) 9031-9042.
[10] K. Takano, K. Kubo, H. Sakamoto, J. Phys.: Condens. Matter 8 (1996) 6405-6411.
[11] T. Tonegawa, T. Hikihara, T. Nishino, M. Kaburagi, S. Miyashita, H.-J. Mikeska, J. Magn. Magn. Matter. (Proc. Int. Conf. Magnetism, Cairns, 1997).
[12] M. C. Cross and D. S. Fisher, Phys. Rev. B 19, 402 (1979).
[13] I. Affleck, Dynamical properties of unconventional magnetic systems (A. T. Skjeltorp and D. Sherrington, Kluwer Academic, Dordrecht, Boston 1998).