

# Evaluation of low-energy effective Hamiltonian techniques for coupled spin triangles

C. Raghu<sup>1</sup>, Indranil Rudra<sup>1</sup>, S. Ramasesha<sup>1</sup>  
and Diptiman Sen<sup>2</sup>

<sup>1</sup> *Solid State and Structural Chemistry Unit*

<sup>2</sup> *Centre for Theoretical Studies*

*Indian Institute of Science, Bangalore 560012, India*

Motivated by recent work on Heisenberg antiferromagnetic spin systems on various lattices made up of triangles, we examine the low-energy properties of a chain of antiferromagnetically coupled triangles of half-odd-integer spins. We derive the low-energy effective Hamiltonian to second order in the ratio of the coupling  $J_2$  between triangles to the coupling  $J_1$  within each triangle. The effective Hamiltonian contains four states for each triangle which are given by the products of spin-1/2 states with the states of a pseudospin-1/2. We compare the results obtained by exact diagonalization of the effective Hamiltonian with those obtained for the full Hamiltonian using exact diagonalization and the density-matrix renormalization group method. It is found that the effective Hamiltonian is accurate only for the ground state for rather low values of the ratio  $J_2/J_1$  and that too for the spin-1/2 case with linear topology. The chain of spin-1/2 triangles shows interesting properties like spontaneous dimerization and several singlet and triplet excited states lying close to the ground state.

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## I. INTRODUCTION

Low-dimensional quantum spin systems with frustration (i.e., competing antiferromagnetic interactions) have been studied extensively in recent years. A particularly interesting class of such systems have lattice structures built out of triangles of spins. Many such systems have been realized experimentally in one and two dimensions, such as the sawtooth chain<sup>1</sup> and the Kagome lattice<sup>2</sup>. A variety of theoretical techniques, both analytical and numerical, are available to study the relevant models<sup>3-6</sup>. Due to large quantum fluctuations, such systems often have several unusual properties. For instance, there may be a gap between a singlet ground state and the nonsinglet excited states; this leads to a magnetic susceptibility which vanishes exponentially at low temperatures. Secondly, the two-spin correlation function may decay rapidly with distance indicating that the magnetic correlation length is not much bigger than the lattice spacing. Finally, there is sometimes no gap to a large number of singlet excited states; this produces an interesting structure for the specific heat at low temperature. The absence of a singlet excitation gap also suggests that there may be a nonmagnetic long-range order, but the nature of this order is not well-understood.

Classically, these systems often have an enormous ground state degeneracy arising from local degrees of freedom which cost no energy; this leads to an extensive entropy at zero temperature. Quantum mechanically, this degeneracy is lifted, but one might still expect a remnant of the classical degeneracy in the form of a large number of low-energy excitations. It is therefore useful to develop simple ways of understanding the low-energy quantum excitations. In this paper, we will critically examine one such way which is to consider the system as being made out of triangles which are weakly coupled to each other<sup>7-9</sup>. We first find the ground states of a single triangle assuming all the three couplings to be antiferromagnetic with a strength  $J_1$ . We then use degenerate perturbation theory to study what happens when different triangles are coupled to each other with antiferromagnetic bonds of strength  $J_2$ , where  $J_2 \ll J_1$ . Since experimental systems typically have  $J_2 = J_1$ , we must finally study how accurate the perturbation theory is when the ratio  $J_2/J_1$  approaches 1.

In Sec. II, we discuss the low-energy states of a single triangle of equal spins coupled antiferromagnetically to each other. If the spin at each site is an integer, then there is a unique ground state for the triangle and the problem is not very interesting. But if the site spin  $S$  is a half-odd-integer, then the ground state has a four-fold degeneracy. This is because the ground state has total spin-1/2, and there is an additional factor of two which can be interpreted as the chirality. The chirality can be expressed as the eigenvalue of a pseudospin operator which also has spin-1/2. We then discuss the low-energy effective Hamiltonian (LEH) for a system in which each pair of triangles is coupled to each other by not more than one antiferromagnetic bond. To first order in  $J_2/J_1$ , the spins and pseudospins of two triangles get coupled to each other. To second order, up to three triangles can get coupled to each other. We have obtained the first order LEH (LEH1) for all values of  $S$ , and the second order LEH (LEH2) for  $S$  equal to 1/2 and 3/2. We have not gone beyond second order because the LEHs rapidly becomes more complicated and longer range as we go to higher orders. For small systems, we numerically diagonalize the LEH1 and LEH2 to obtain the energies of the ground state and the first excited state as functions of  $J_2/J_1$ .

In Sec. III, we use both exact diagonalization for small systems and the density-matrix renormalization group method (DMRG) for larger systems to obtain the low-lying energies for the complete Hamiltonian. The DMRG is currently the most accurate numerical method known for obtaining the low-energy properties of quantum systems in one-dimension<sup>10</sup>. We briefly describe the DMRG procedure for our system, and then compare the energies obtained from the LEHs and DMRG. Depending on the quantities of interest, we find that the results from the LEHs start deviating significantly from the DMRG values once  $J_2/J_1$  exceeds 0.2 – 0.4. This is true even for the LEH2 which is significantly more accurate than the LEH1 for small values of  $J_2/J_1$ .

We also examine a simple system of five triangles coupled to each other in such a way as to form a small fragment of the two-dimensional Kagome lattice; we find that the LEHs starts deviating from the

exact results at even smaller values of  $J_2/J_1$ . Thus the LEH approach becomes less accurate the more two-dimensional the geometry is, i.e., the larger the coordination number is for each triangle.

## II. LOW-ENERGY HAMILTONIANS FOR A CHAIN OF TRIANGLES

We are interested in the system of spins shown in Fig. 1. The sites are labeled as  $(n, a)$ , where  $n$  labels the triangle and  $a = 1, 2, 3$  labels the three sites of a triangle as indicated. The spin at each site has the value  $S$ , and all the couplings are antiferromagnetic. The Hamiltonian is

$$\hat{H} = \sum_n \left[ J_1 ( \hat{\mathbf{S}}_{n,1} \cdot \hat{\mathbf{S}}_{n,2} + \hat{\mathbf{S}}_{n,2} \cdot \hat{\mathbf{S}}_{n,3} + \hat{\mathbf{S}}_{n,3} \cdot \hat{\mathbf{S}}_{n,1} ) + J_2 \hat{\mathbf{S}}_{n,2} \cdot \hat{\mathbf{S}}_{n+1,3} \right]. \quad (1)$$

It is convenient to set  $J_1 = 1$  and consider only the parameter  $J_2$ .

Let us first examine a single triangle and drop the label  $n$ . The Hamiltonian  $\hat{h}_\Delta$  is proportional to the square of the total spin operator, ignoring a shift in the zero of energy,

$$\hat{h}_\Delta = \hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2 + \hat{\mathbf{S}}_2 \cdot \hat{\mathbf{S}}_3 + \hat{\mathbf{S}}_3 \cdot \hat{\mathbf{S}}_1 = \frac{1}{2} ( \hat{\mathbf{S}}_1 + \hat{\mathbf{S}}_2 + \hat{\mathbf{S}}_3 )^2 - \frac{3}{2} S(S+1). \quad (2)$$

If  $S$  is an integer  $(1, 2, \dots)$ , then the ground state of this triangle is unique and is a singlet. Thus the low-energy sector of the entire system consists of only one state when different triangles are coupled to each other. The situation is much more interesting if  $S$  is a half-odd-integer, i.e.,  $1/2, 3/2, \dots$ . Then the ground state of each triangle is four-fold degenerate because it must have total spin  $1/2$  and there are two ways of obtaining total spin- $1/2$  by combining three equal spins. For instance, sites 2 and 3 can first combine to give total spin  $S_{23}$  equal to either  $S + 1/2$  or  $S - 1/2$ ;  $S_{23}$  can then combine with the first spin to produce a total spin- $1/2$ . Since the total spin operator  $\hat{\mathbf{S}} = \hat{\mathbf{S}}_1 + \hat{\mathbf{S}}_2 + \hat{\mathbf{S}}_3$  commutes with all the six permutations of the three spins, it is clear that the two ground states of the Hamiltonian in (2) (with a given value of the total spin component  $S^z = \pm 1/2$ ) must form a two-dimensional representation of the permutation group. This representation can be made explicit by introducing pseudospin- $1/2$  operators  $\hat{\tau}$  as follows.

We will assume henceforth that all the site spins are identical and have the half-odd-integer value  $S$ . To obtain a total spin- $1/2$  for a triangle of three spins, the total spin of any two sites should be one of the integers  $S \pm 1/2$ . Under an exchange of the two spins 2 and 3, a state with their total spin  $S_{23}$  transforms by the phase  $(-1)^{S_{23}+1}$ ; thus it is symmetric or antisymmetric depending on whether  $S_{23}$  is odd or even. Let us define the pseudospin operator  $\hat{\tau}^x$  such that the symmetric states have the eigenvalue of  $\hat{\tau}^x = 1$ , while the antisymmetric states have the eigenvalue of  $\hat{\tau}^x = -1$ . Similarly, we can introduce an operator  $\hat{\tau}^y$  such that states which are symmetric (antisymmetric) with respect to exchange of spins 1 and 2 have eigenvalues of  $(-\hat{\tau}^x + \sqrt{3}\hat{\tau}^y)/2$  equal to  $1$  ( $-1$ ). For spins 1 and 3, the operator  $(-\hat{\tau}^x - \sqrt{3}\hat{\tau}^y)/2$  will have the same property. We define the third pseudospin operator  $\hat{\tau}^z = -(i/2)[\hat{\tau}^x, \hat{\tau}^y]$ . From these statements, it follows that within the space of the four ground states of a triangle, we have the operator identities

$$\begin{aligned} \hat{\mathbf{S}}_2 \cdot \hat{\mathbf{S}}_3 &= A - \frac{(-1)^{S+1/2}}{4} (2S+1) \hat{\tau}^x, \\ \hat{\mathbf{S}}_3 \cdot \hat{\mathbf{S}}_1 &= A - \frac{(-1)^{S+1/2}}{4} (2S+1) \left[ -\frac{1}{2} \hat{\tau}^x - \frac{\sqrt{3}}{2} \hat{\tau}^y \right], \\ \hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2 &= A - \frac{(-1)^{S+1/2}}{4} (2S+1) \left[ -\frac{1}{2} \hat{\tau}^x + \frac{\sqrt{3}}{2} \hat{\tau}^y \right], \end{aligned} \quad (3)$$

where  $A = -S^2/2 - S/2 + 1/8$ . On taking the commutator of any two of the equations in (3), we find that the three-spin chirality operator is given by

$$\hat{\mathbf{S}}_1 \cdot \hat{\mathbf{S}}_2 \times \hat{\mathbf{S}}_3 = \frac{\sqrt{3}}{4} \left( S + \frac{1}{2} \right)^2 \hat{\tau}^z . \quad (4)$$

Before proceeding further, we should mention that the pseudospin operators have been discussed earlier in Refs. 7 and 8 for the spin-1/2 case. In those papers, the two ground states (with a given value of  $S^z$ ) are written in terms of a spin wave running around the triangle with momenta  $\pm 2\pi/3$ ; these are called right and left moving respectively, and they are eigenstates of the operator  $\hat{\tau}^z$ . We have instead chosen to describe the states in terms of two of the spins forming total spin  $S + 1/2$  or  $S - 1/2$ . The two descriptions are clearly related to each other by an unitary transformation.

To continue, the Wigner-Eckart theorem says that the matrix elements of any of the site spin operators  $\hat{\mathbf{S}}_a$  (where  $a = 1, 2, 3$ ) between the four ground states of a triangle must be proportional to the matrix elements of the total spin operator  $\hat{\mathbf{s}}$ . The proportionality factors must be independent of the spin component (i.e.,  $x, y$  or  $z$ ), but they will involve the pseudospin operators. Let us introduce the operators

$$\hat{\tau}_a = \cos \frac{2\pi}{3}(1-a) \hat{\tau}^x + \sin \frac{2\pi}{3}(1-a) \hat{\tau}^y , \quad (5)$$

where  $a$  can take the values 1, 2, 3. Using the spin wave functions, we can then show that the matrix elements of  $\hat{\mathbf{S}}_a$  and  $\hat{\mathbf{s}}$  are related as

$$\langle \sigma, \tau | \hat{\mathbf{S}}_a | \sigma', \tau' \rangle = \frac{1}{3} \langle \sigma | \hat{\mathbf{s}} | \sigma' \rangle \left[ \delta_{\tau, \tau'} + (-1)^{S+1/2} (2S+1) \langle \tau | \hat{\tau}_a | \tau' \rangle \right] , \quad (6)$$

where  $\sigma, \sigma'$  are the eigenstates of  $\hat{\sigma}^z$  and  $\tau, \tau'$  are the eigenstates of  $\hat{\tau}^z$ .

We can now derive the LEHs when two or more triangles are coupled together as in Eq. (1). We write that Hamiltonian as  $\hat{H} = \hat{H}_0 + \hat{V}$ , where  $\hat{H}_0$  consists of the interactions within the triangles as in Eq. (2), while  $\hat{V}$  consists of the interactions between triangles and is proportional to  $J_2$ . The LEH is a perturbative expansion in the parameter  $J_2$ . For  $J_2 = 0$ , the low-energy sector for  $N$  triangles contains  $4^N$  states, all of which have the same energy  $E_0 = N e_0$ , where

$$e_0 = \frac{3}{8} - \frac{3}{2} S(S+1) . \quad (7)$$

is the ground state energy of (2). Following Ref. 9, let us denote the different low-energy states of the system as  $p_i$  and the high-energy states of the system as  $q_\alpha$  (these are states in which at least one of the triangles is in a state with total spin  $\geq 3/2$ ). The high-energy states have energies  $E_\alpha$  according to the exactly solvable Hamiltonian  $\hat{H}_0$ . Then the LEH1 is given by degenerate perturbation theory,

$$\hat{H}_{eff}^{(1)} = \sum_{ij} |p_i\rangle \langle p_i | \hat{V} | p_j \rangle \langle p_j | . \quad (8)$$

The LEH2 is given by

$$\hat{H}_{eff}^{(2)} = \sum_{ij} \sum_{\alpha} |p_i\rangle \frac{\langle p_i | \hat{V} | q_\alpha \rangle \langle q_\alpha | \hat{V} | p_j \rangle}{E_0 - E_\alpha} \langle p_j | . \quad (9)$$

The total effective Hamiltonian to this order is given by

$$\hat{H}_{LEH2} = E_0 + \hat{H}_{eff}^{(1)} + \hat{H}_{eff}^{(2)} . \quad (10)$$

The LEH1 can now be directly read off from Eq. (6) after adding the triangle label  $n$ ; thus the total spin operator of triangle  $n$  is denoted as  $\hat{\mathbf{s}}_n$  and the pseudospin operator as  $\hat{\tau}_n$ . In general, if site number

$a$  of triangle  $l$  is connected to site number  $b$  of triangle  $m$  by a bond of strength  $J_2$  (see Fig. 2), the contribution of that bond to the LEH1 is given by

$$\hat{h}_{eff}^{(1)} = \frac{J_2}{9} \hat{\mathbf{s}}_l \cdot \hat{\mathbf{s}}_m \left[ 1 + (-1)^{S+1/2} (2S+1) \hat{\tau}_l^a \right] \left[ 1 + (-1)^{S+1/2} (2S+1) \hat{\tau}_m^b \right]. \quad (11)$$

For the particular form of couplings shown in Fig. 1, the total LEH1 takes the form

$$\hat{H}_{LEH1} = -N e_0 + \frac{J_2}{9} \sum_n \hat{\mathbf{s}}_n \cdot \hat{\mathbf{s}}_{n+1} \left[ 1 + (-1)^{S+1/2} (2S+1) \hat{\tau}_n^2 \right] \left[ 1 + (-1)^{S+1/2} (2S+1) \hat{\tau}_{n+1}^3 \right]. \quad (12)$$

The derivation of the LEH2 in (9) requires a much longer calculation since we have to first compute matrix elements of the form  $\langle q_\alpha | \hat{\mathbf{S}}_a | p_i \rangle$  for all the states within each triangle, and we then have to take products of these to obtain the matrix elements of  $\hat{\mathbf{s}}_{n,2} \cdot \hat{\mathbf{s}}_{n+1,3}$ . The second-order terms in the LEH can arise from either (i) a single bond connecting site  $a$  of triangle  $l$  to site  $b$  of triangle  $m$ , or (ii) a bond connecting site  $a$  of triangle  $l$  to site  $b$  of triangle  $m$  and another bond connecting site  $c$  of triangle  $m$  to site  $d$  of triangle  $n$ , where  $a, b, c, d$  take values from 1, 2, 3 (here  $b$  may or may not be equal to  $c$ ). The notation for these two types is shown in Fig. 2.

Let us first consider the simplest situation in which all the site spins are equal to  $S = 1/2$ . The contribution of type (i) to the LEH2 is then given by

$$\hat{h}_{eff,(i)}^{(2)} = -\frac{J_2^2}{54} \left[ (1 - \hat{\tau}_{l,a} \hat{\tau}_{m,b}) (3 + 4\hat{\mathbf{s}}_l \cdot \hat{\mathbf{s}}_m) + (1 + \hat{\tau}_{l,a})(1 + \hat{\tau}_{m,b}) \right], \quad (13)$$

where we have used the notation of Eq. (5). The contribution of type (ii) is

$$\begin{aligned} \hat{h}_{eff,(ii)}^{(2)} = & -\frac{4J_2^2}{243} (1 - 2\hat{\tau}_{l,a})(1 - 2\hat{\tau}_{n,d}) \left[ \left( \cos \frac{2\pi}{3}(b-c) + \hat{\tau}_{m,-b-c} \right) \hat{\mathbf{s}}_l \cdot \hat{\mathbf{s}}_n \right. \\ & \left. + \sin \frac{2\pi}{3}(b-c) \hat{\tau}_m^z \hat{\mathbf{s}}_l \cdot \hat{\mathbf{s}}_m \times \hat{\mathbf{s}}_n \right]. \end{aligned} \quad (14)$$

Putting all this together for the system in Fig. 1, we find that for  $S = 1/2$ , the total LEH2 is given by

$$\begin{aligned} \hat{H}_{LEH2} = & -\frac{3N}{4} + \frac{J_2}{9} \sum_n \hat{\mathbf{s}}_n \cdot \hat{\mathbf{s}}_{n+1} (1 - 2\hat{\tau}_{n,2}) (1 - 2\hat{\tau}_{n+1,3}) \\ & -\frac{J_2^2}{54} \sum_n \left[ (1 - \hat{\tau}_{n,2} \hat{\tau}_{n+1,3}) (3 + 4\hat{\mathbf{s}}_n \cdot \hat{\mathbf{s}}_{n+1}) + (1 + \hat{\tau}_{n,2}) (1 + \hat{\tau}_{n+1,3}) \right] \\ & -\frac{4J_2^2}{243} \sum_n (1 - 2\hat{\tau}_{n,2}) (1 - 2\hat{\tau}_{n+1,3}) \left[ \left(-\frac{1}{2} + \hat{\tau}_{n+1,1}\right) \hat{\mathbf{s}}_n \cdot \hat{\mathbf{s}}_{n+2} - \frac{\sqrt{3}}{2} \hat{\tau}_{n+1}^z \hat{\mathbf{s}}_n \cdot \hat{\mathbf{s}}_{n+1} \times \hat{\mathbf{s}}_{n+2} \right]. \end{aligned} \quad (15)$$

If the site spins are equal to  $S = 3/2$ , the contribution of type (i) to the LEH2 is given by

$$\begin{aligned} \hat{h}_{eff,(i)}^{(2)} = & -\frac{J_2^2}{27} \left[ 56 + 42\hat{\mathbf{s}}_l \cdot \hat{\mathbf{s}}_m + (\hat{\tau}_{l,a} + \hat{\tau}_{m,b}) (-1 + 8\hat{\mathbf{s}}_l \cdot \hat{\mathbf{s}}_m) \right. \\ & \left. - \hat{\tau}_{l,a} \hat{\tau}_{m,b} (4 + 8\hat{\mathbf{s}}_l \cdot \hat{\mathbf{s}}_m) \right]. \end{aligned} \quad (16)$$

The contribution of type (ii) is

$$\hat{h}_{eff,(ii)}^{(2)} = -\frac{4J_2^2}{243} (1 + 4\hat{\tau}_{l,a})(1 + 4\hat{\tau}_{n,d}) \left[ \left( 7 \cos \frac{2\pi}{3}(b-c) - 2 \hat{\tau}_{m,-b-c} \right) \hat{\mathbf{s}}_l \cdot \hat{\mathbf{s}}_n + 4 \sin \frac{2\pi}{3}(b-c) \hat{\tau}_m^z \hat{\mathbf{s}}_l \cdot \hat{\mathbf{s}}_m \times \hat{\mathbf{s}}_n \right]. \quad (17)$$

For the system in Fig. 1, the total LEH2 for  $S = 3/2$  is therefore

$$\begin{aligned} \hat{H}_{LEH2} = & -\frac{21N}{4} + \frac{J_2}{9} \sum_n \hat{\mathbf{s}}_n \cdot \hat{\mathbf{s}}_{n+1} (1 + 4\hat{\tau}_{n,2}) (1 + 4\hat{\tau}_{n+1,3}) \\ & -\frac{J_2^2}{27} \sum_n \left[ 56 + 42\hat{\mathbf{s}}_n \cdot \hat{\mathbf{s}}_{n+1} + (\hat{\tau}_{n,2} + \hat{\tau}_{n+1,3})(-1 + 8\hat{\mathbf{s}}_n \cdot \hat{\mathbf{s}}_{n+1}) - \hat{\tau}_{n,2}\hat{\tau}_{n+1,3}(4 + 8\hat{\mathbf{s}}_n \cdot \hat{\mathbf{s}}_{n+1}) \right] \\ & -\frac{4J_2^2}{243} \sum_n (1 + 4\hat{\tau}_{n,2}) (1 + 4\hat{\tau}_{n+1,3}) \left[ \left(-\frac{7}{2} + \hat{\tau}_{n+1,1}\right) \hat{\mathbf{s}}_n \cdot \hat{\mathbf{s}}_{n+2} - 2\sqrt{3} \hat{\tau}_{n+1}^z \hat{\mathbf{s}}_n \cdot \hat{\mathbf{s}}_{n+1} \times \hat{\mathbf{s}}_{n+2} \right]. \end{aligned} \quad (18)$$

We have carried out exact diagonalization studies of the LEH1 and LEH2 for systems up to 10 triangles for both the spin-1/2 and spin-3/2 cases.

### III. DENSITY-MATRIX RENORMALIZATION GROUP STUDY OF THE CHAIN OF TRIANGLES

We have numerically studied the system described by Eq. (1) using exact diagonalization for small systems and the DMRG method for larger systems. The number of triangles is  $N$  and the number of sites is  $3N$ .

For small systems, we have performed exact diagonalization with periodic boundary conditions. For larger systems, we have done DMRG calculations (using the infinite system algorithm<sup>10</sup>) with open boundary conditions. For exact diagonalization, we have gone up to 30 sites (10 triangles). With DMRG, we have gone up to 50 triangles and in some cases up to 100 triangles, after checking that the DMRG and exact results match for 10 triangles for the spin-1/2 system. The number of dominant density matrix eigenstates, corresponding to the  $m$  largest eigenvalues of the density matrix, that we retained at each DMRG iteration was 64. In fact, we varied the value of  $m$  from 64 to 100 and found that  $m = 64$  gives satisfactory results in terms of agreement with exact diagonalization for small systems and good numerical convergence for large systems. The system is grown by adding two new triangles at each iteration; we found that this gives more accurate results than either adding two new sites at each iteration (in which case we would have obtained the triangle structure only after every third iteration) or adding one triangle at each iteration.

In Fig. 3, we show the DMRG values of the ground state energy per site versus  $1/N$  for a few illustrative values of  $J_2$  (in units of  $J_1$ ) for the case in which each site has spin-1/2. After extrapolating to the thermodynamics limit  $N \rightarrow \infty$ , we show the ground state energy per site as a function of  $J_2$  in Fig. 4. In that figure, we also show the numerical results obtained by exact diagonalization of the LEH1 and LEH2 respectively. As expected, the LEH2 is more accurate than the LEH1 up to a larger value of  $J_2$ . However, even the LEH2 becomes fairly inaccurate beyond about  $J_2 = 0.4$ .

A more detailed comparison of the LEH1 and LEH2 with the exact results for chains with up to 10 triangles is given in Table 1. We see that the LEH2 agrees better with the exact results than the LEH1, although the agreement becomes rather poor for large  $J_2$ . We also see that the accuracy of the LEHs is poorer for the gap than it is for the ground state energy. For instance, the LEH2 results for the gap become relatively inaccurate even for  $J_2 = 0.2$ .

Fig. 5 shows the low-lying triplet and singlet gaps above the ground state for the same system at different values of  $J_2$ . The figure compares the results obtained from the LEH2 with those obtained after extrapolating the gaps from exact diagonalization of the full Hamiltonian for different values of  $N$ . Both the triplet (Fig. 5 (a)) and singlet (Fig. 5 (b)) gaps deviate considerably from the "exact" results beyond  $J_2 \sim 0.4$ . In fact, we see from Fig. 5 (a) that the ground state of the LEH2 switches from a singlet to a triplet for  $J_2 > 0.7$ .

In Fig. 6, we show a larger number of  $S = 0$  and  $S = 1$  states which have very small gaps above the ground state. It is possible that some of these actually become degenerate with the ground state in the thermodynamic limit. The lowest spin sector with an appreciable gap (which is likely to remain non-zero in the thermodynamic limit) seems to be  $S = 2$ . However it is possible that there are many more singlet and triplet states lying below the lowest  $S = 2$  state than we have shown in the figure; it is very difficult to find more than a few low-lying states in each spin sector using the DMRG method. For the same reason, we cannot rule out the possibility of a finite gap to a higher singlet or a higher triplet (than what we have targeted) lying below the quintet. Fig. 7 shows an even larger number of low-lying states for the case  $J_2 = 1.0$ . The lowest three states with  $S^z = 0$  (including the ground state), the lowest six states with  $S^z = 0$  and the lowest state with  $S^z = 2$  are shown there. To summarize, we have found an unexpectedly large number of low-lying (and possibly gapless) singlet and triplet excitations in this system. We will provide an explanation for some of these states below. We note that the low-energy spectrum has a resemblance to that found in the Kagome lattice in two dimensions<sup>5</sup>. However there is an important difference between the two cases; the gapless band of excitations in the Kagome problem consists only of singlets, and the first gap is to a triplet state.

In Fig. 8 (a), we show the DMRG values of the bond order for the middle two bonds, namely, the ground state expectation values  $\langle \hat{s}_{p,2} \cdot \hat{s}_{p+1,3} \rangle$  for  $p = N/2 - 1$  and  $N$ , as a function of  $N$  at  $J_2 = 1.0$  for the spin-1/2 system. In Fig. 8 (b), we plot the bond order alternation (or spontaneous dimerization), defined to be the magnitude of the difference of the two neighbouring bond orders in Fig. 8 (a) extrapolated to the limit  $N \rightarrow \infty$ , as a function of  $J_2$ . We observe that the alternation is quite large for small values of  $J_2$  and that it remains non-zero even at a large value of  $J_2 = J_1$ . Following Mila<sup>8</sup>, we can qualitatively understand the dimerization occurring at small values of  $J_2$  as follows. Since the spin and pseudospin degrees of freedom appear very asymmetrically in the LEHs, we perform a mean-field decoupling of these two. We assume that the pseudospin variables take some fixed values and find the quantum ground state of the spin variables in that fixed background. For two triangles coupled together by the LEH1 in Eq. (11), we see that the lowest energy is attained if the pseudospin variables satisfy  $\tau_{l,a} = \tau_{m,b} = -1$  (for the case  $S = 1/2$ ), and the effective spin-1/2 of the two triangles then form a singlet. In the same way, the mean-field ground state of the chain is given by the dimerized configuration in which  $\tau_{2n,2} = \tau_{2n+1,3} = -1$  and the effective spin-1/2 of triangles  $2n$  and  $2n + 1$  form a singlet. Such a dimerized ground state can also be obtained by translating the above state by one triangle; it is therefore two-fold degenerate as in a spin-Peierls system.

It is instructive to contrast this system with the antiferromagnetic spin-1/2 chain with a nearest-neighbor coupling  $J_1$  and a next-nearest-neighbor coupling  $J_2$ <sup>11</sup>. For  $J_2/J_1 > 0.241\dots$ , this is known to spontaneously dimerize, and there is also a finite gap to excitations in the bulk and a finite correlation length  $\xi$  in the thermodynamic limit. In the dimerized phase, an open chain with an even number of sites has five ground states corresponding to two singlets and one triplet. One singlet and one triplet arise from the free spin-1/2 degrees of freedom which reside at the two ends of the chain. The finite correlation length  $\xi$  means that the splitting between these states vanishes exponentially with the size of the system.

Similarly, for our system with a chain of an even number of triangles, we may expect at least some of the low-energy states to arise from the effective spin-1/2 degrees of freedom residing on the two end triangles. Due to the presence of the pseudospin-1/2 degrees of freedom in each triangle, we expect that there will be a total of  $4^2 = 16$  low-energy states forming four triplets and four singlets. The number of low-energy singlets and triplets that we actually find is more than this; this implies that there are some additional low-energy degrees of freedom (probably associated with the bulk) which we do not yet

understand.

In Table 2, we show the ground state energy per site versus  $J_2$  for the case in which each site has spin-3/2. The numerical results obtained by exact diagonalization of the LEH1 and LEH2 are also shown. A comparison with Table 1 for the spin-1/2 case shows that the LEH2 starts deviating from the exact results for smaller values of  $J_2$  as the site spin is increased.

Finally, in Fig. 9, we show the ground state energy per site versus  $J_2$  for a group of five triangles forming a sub-system of a two-dimensional Kagome lattice. The reason for studying this system is to test how well the LEHs do as the coordination number of the triangles is increased from two, thereby making the system more two-dimensional. We find that the LEH2 fails even faster with increasing  $J_2$  than it does for the chains studied above.

#### IV. SUMMARY AND OUTLOOK

We have studied a chain of coupled triangles of half-odd-integer spins using both the DMRG method and the LEH approaches. We find that the LEH approach is accurate for the ground state energy only for small values of the ratio of the coupling between triangles and the coupling within each triangle; the accuracy for the low-energy gaps is less than that for the ground state energy. The range of accuracy of the LEHs is also smaller for larger values of the site spin as well as for a larger number of neighbors coupled to each triangle. We therefore conclude that the LEH approaches may not be very reliable for the low-energy properties of the currently existing experimental systems in which all couplings are of the same order and the geometry is two-dimensional<sup>8</sup>.

#### Acknowledgments

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Ground State					First Excited State		
$J_2$	N	LEH1	LEH2	Exact	LEH1	LEH2	Exact
0.1	4	-3.15843	-3.16139	-3.16135	-3.14912	-3.15131	-3.15130
	6	-4.73740	-4.73772	-4.73768	-4.72959	-4.73334	-4.73331
	8	-6.30976	-6.31541	-6.31536	-6.30795	-6.31319	-6.31315
0.2	4	-3.31686	-3.32892	-3.32864	-3.29825	-3.30728	-3.30713
	6	-4.96681	-4.98455	-4.98417	-4.95918	-4.97455	-4.97427
	8	-6.61952	-6.64278	-6.64229	-6.61590	-6.63740	-6.63699
0.5	4	-3.79215	-3.87213	-3.86669	-3.74562	-3.80782	-3.80463
	6	-5.66702	-5.78669	-5.77885	-5.64794	-5.75145	-5.74604
	8	-7.54881	-7.70679	-7.69665	-7.53975	-7.68454	-7.67639
1.0	4	-4.58430	-4.93506	-4.88000	-4.49125	-4.78367	-4.74638
	6	-6.83404	-7.02622	-7.28614	-6.79589	-7.26277	-7.20489
	8	-9.09761	-9.81245	-9.70079	-9.07950	-9.73152	-9.64368

Table 1. Ground state and first excited state energies in units of  $J_1$  obtained for the spin-1/2 system using exact diagonalization of the full Hamiltonian and the LEH1 and LEH2.

$J_2$	N=3			N=4		
	LEH1	LEH2	Exact	LEH1	LEH2	Exact
0.1	-16.0462	-16.1044	-16.1071	-21.4577	-21.5211	-21.5224
0.2	-16.3424	-16.5793	-16.5692	-21.9154	-22.1775	-22.1611
0.3	-16.6386	-17.1799	-17.1132	-22.3731	-22.9829	-22.8961
0.4	-16.9348	-17.9107	-17.7212	-22.8308	-23.9508	-23.7107
0.5	-17.2309	-18.7747	-18.3807	-23.2884	-25.0933	-24.5916
0.6	-17.5271	-19.7746	-19.0834	-23.7461	-26.4200	-25.5294
0.7	-17.8233	-20.9118	-19.8233	-24.2038	-27.9539	-26.5170
0.8	-18.1195	-22.1878	-20.5966	-24.6615	-29.7650	-27.5488
0.9	-18.4157	-23.6032	-21.4006	-25.1192	-31.8070	-28.6206
1.0	-18.7119	-25.1586	-22.2325	-25.5769	-34.0823	-29.7285

Table 2. Ground state energies obtained for the spin-3/2 system using exact diagonalization of the full Hamiltonian and the LEH1 and LEH2 for 3 and 4 triangles.

## Figure Captions

1. The chain of triangles showing the labeling of the vertices of each triangle and the antiferromagnetic couplings  $J_1 = 1$  and  $J_2$ .
2. The triangle and site labels used in deriving the LEHs.
3. The ground state energy/site in units of  $J_1$  vs  $1/N$ , for a few values of  $J_2$  with spin-1/2 at each site.
4. The ground state energy/site in units of  $J_1$ , for values of  $J_2 = 0.1$  to  $1.0$ . The three sets of points show the results using DMRG for the full Hamiltonian and the results from the LEH1 and LEH2.
5. The energy gaps from the ground state in units of  $J_1$ , for values of  $J_2 = 0.1$  to  $1.0$  for the spin-1/2 system. (a) shows the triplet gap and (b) shows the singlet gap. The two sets of points show the results obtained from the LEH2 and DMRG.
6. Some low-lying energy gaps for  $J_2 = 0.0$  to  $1.0$  for the spin-1/2 system obtained by DMRG. The two lowest  $S^z = 0$  states (including the ground state at the bottom of the figure), the two lowest  $S^z = 1$  states and the lowest  $S^z = 2$  state are shown.
7. A larger number of low-lying energy gaps for  $J_2 = 1.0$  for the spin-1/2 system obtained by DMRG. The two lowest  $S^z = 0$  states (including the ground state at the bottom), the six lowest  $S^z = 1$  states and the lowest  $S^z = 2$  state are shown.
8. The spin bond orders for the spin-1/2 system obtained by DMRG. (a) shows the bond orders for two neighbouring bonds in the middle of the chain as a function of  $N$  for  $J_2 = 1.0$ . (b) shows the difference of the two bond orders (called the bond alternation or dimerization) for various values of  $J_2$ .
9. The ground state energy in units of  $J_1 = 1$ , for values of  $J_2 = 0.1$  to  $1.0$  for spin-1/2 at each site of a system of five triangles as shown in the inset. The two sets of points show the results from the exact diagonalization of the LEH2 and of the full Hamiltonian.

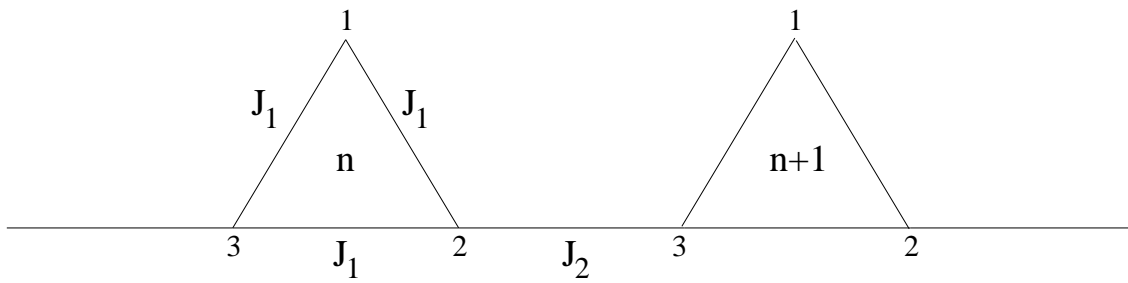


Fig. 1

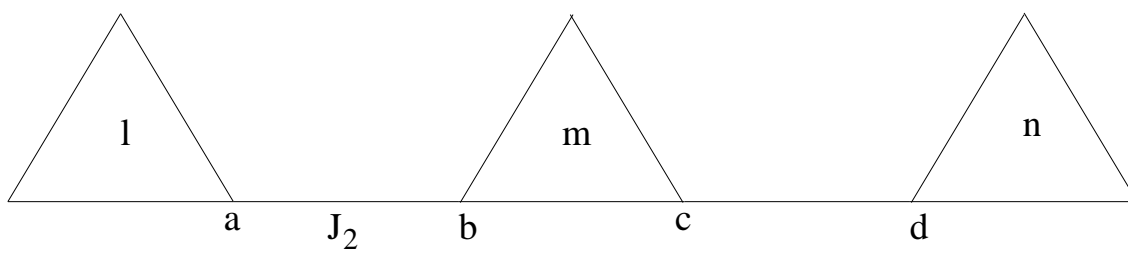


Fig. 2

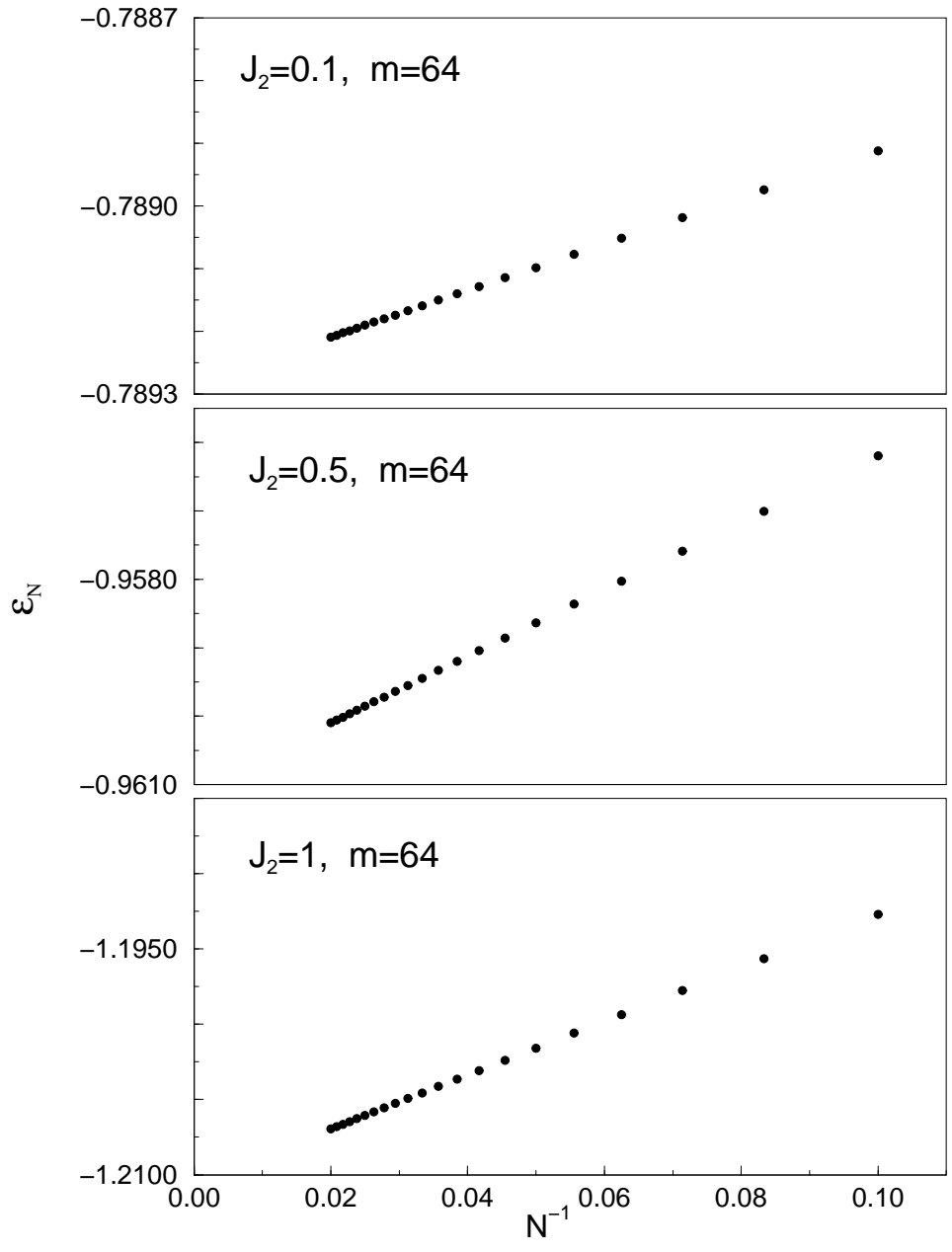


Fig. 3

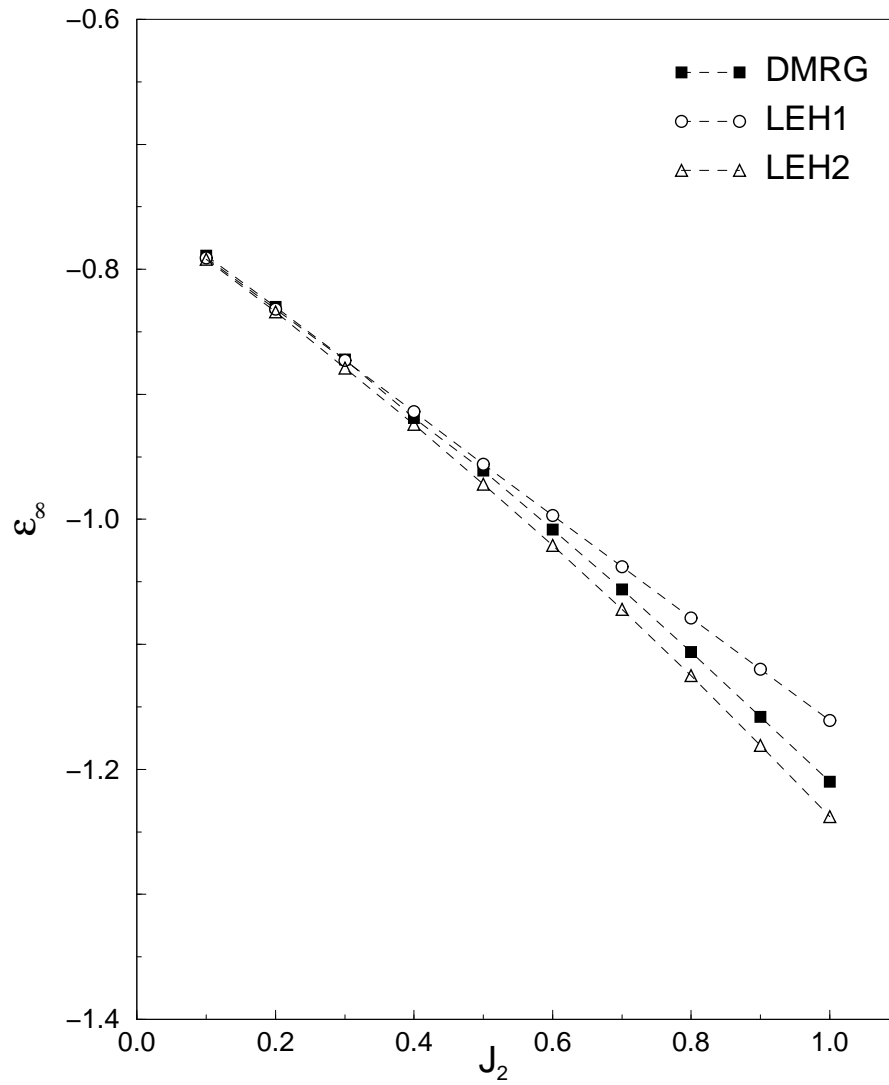


Fig. 4

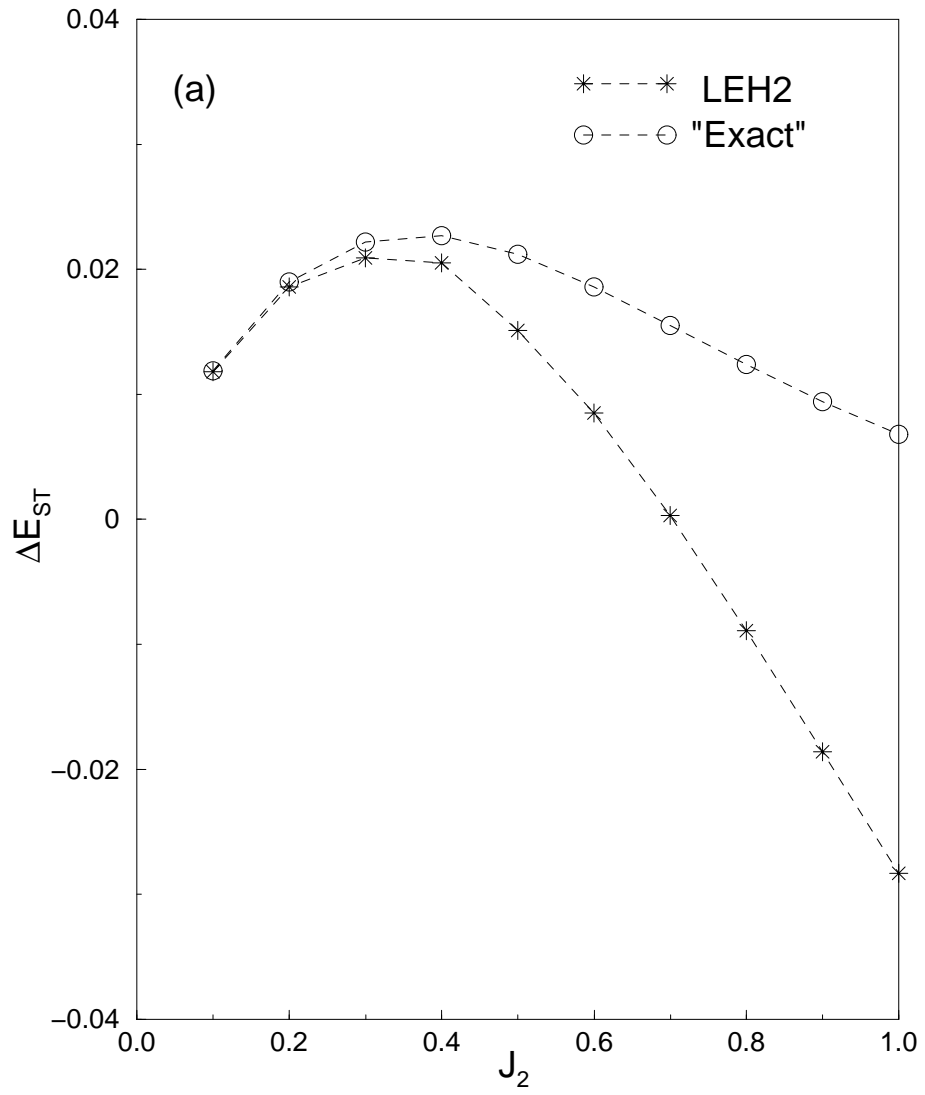


Fig. 5 (a)

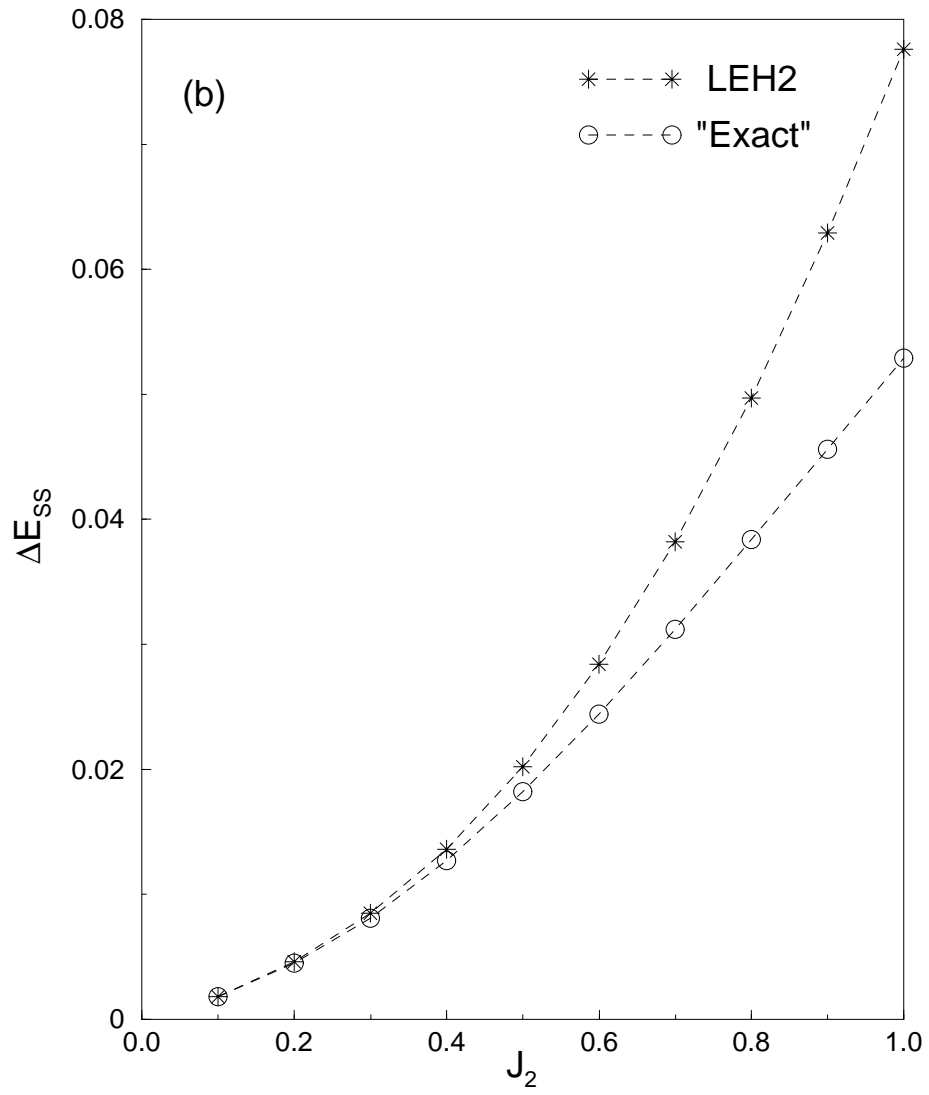


Fig. 5 (b)



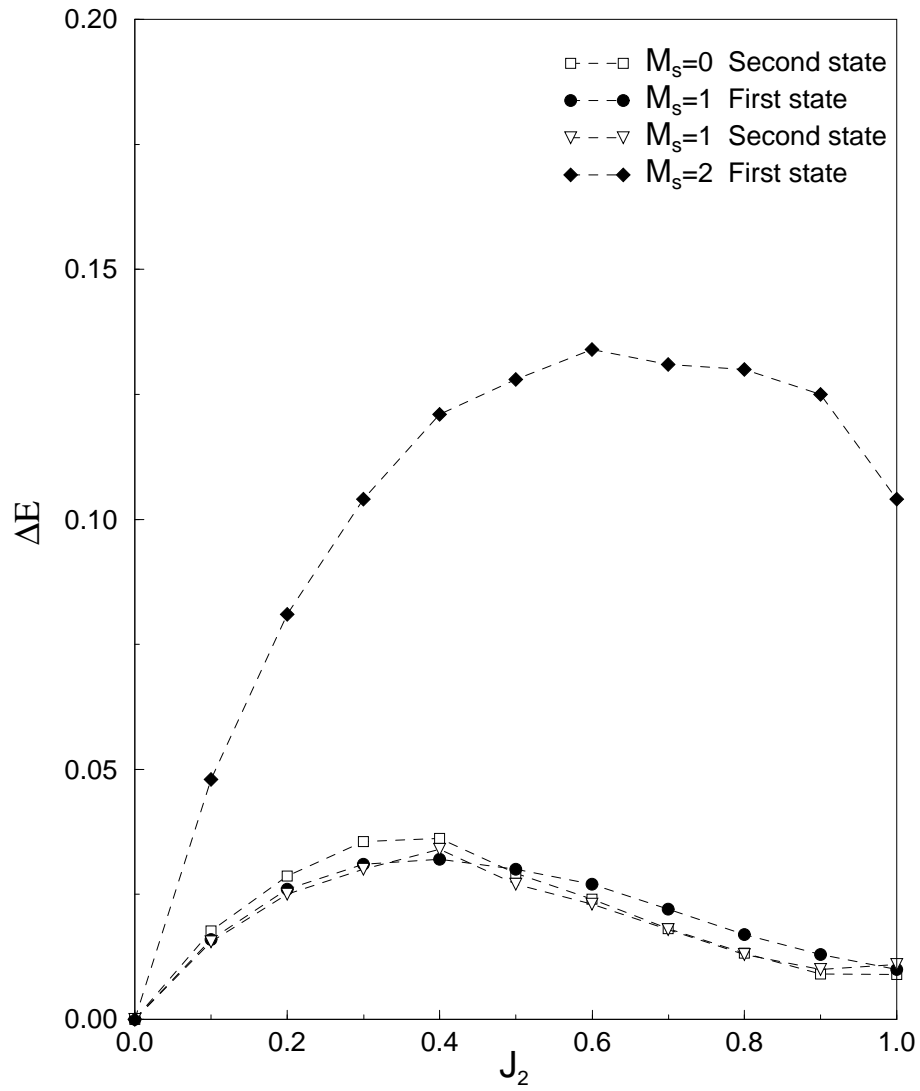


Fig. 6

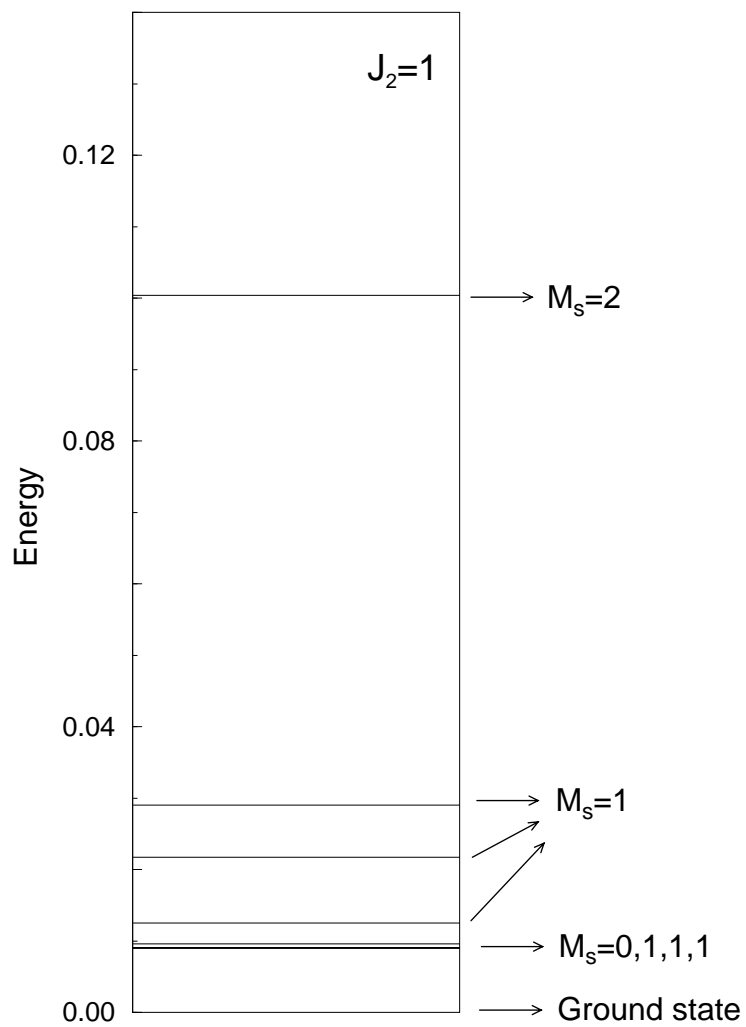


Fig. 7

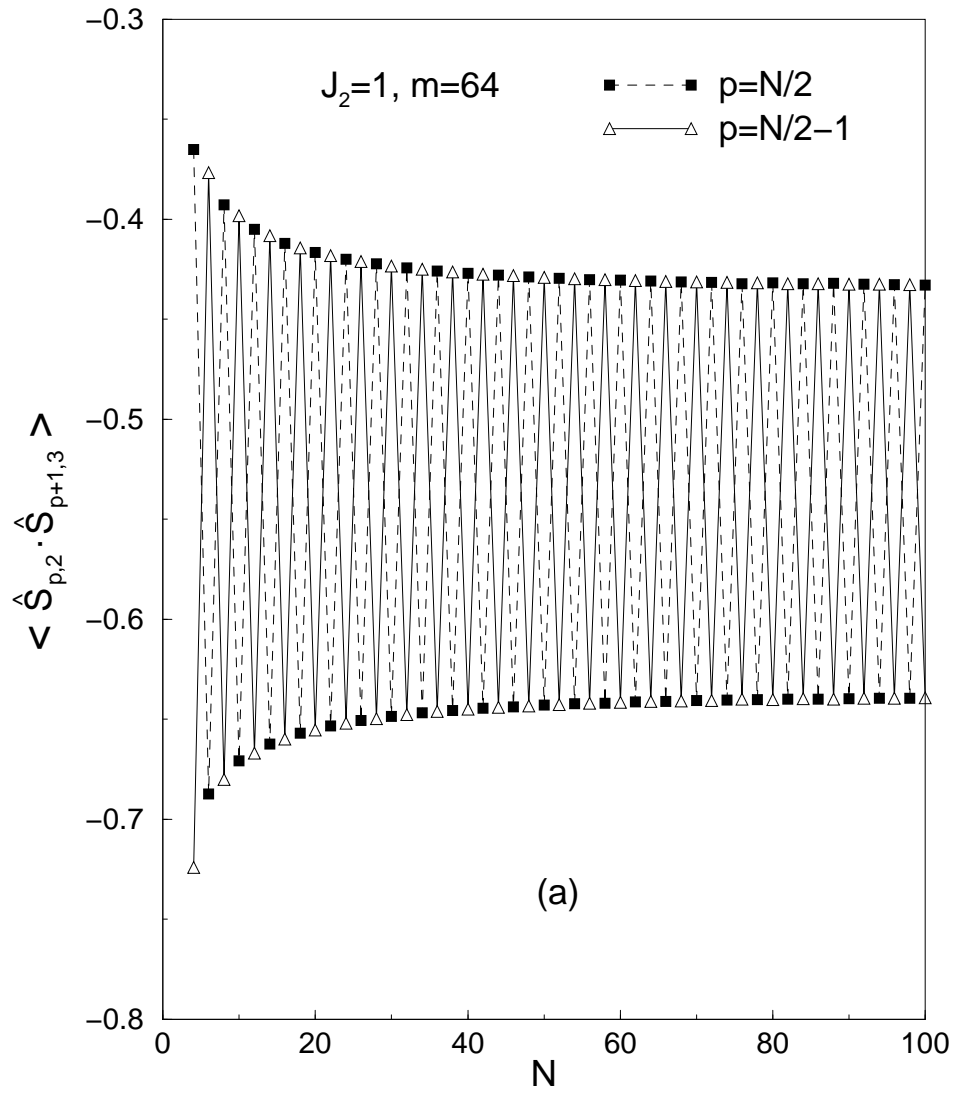


Fig. 8 (a)

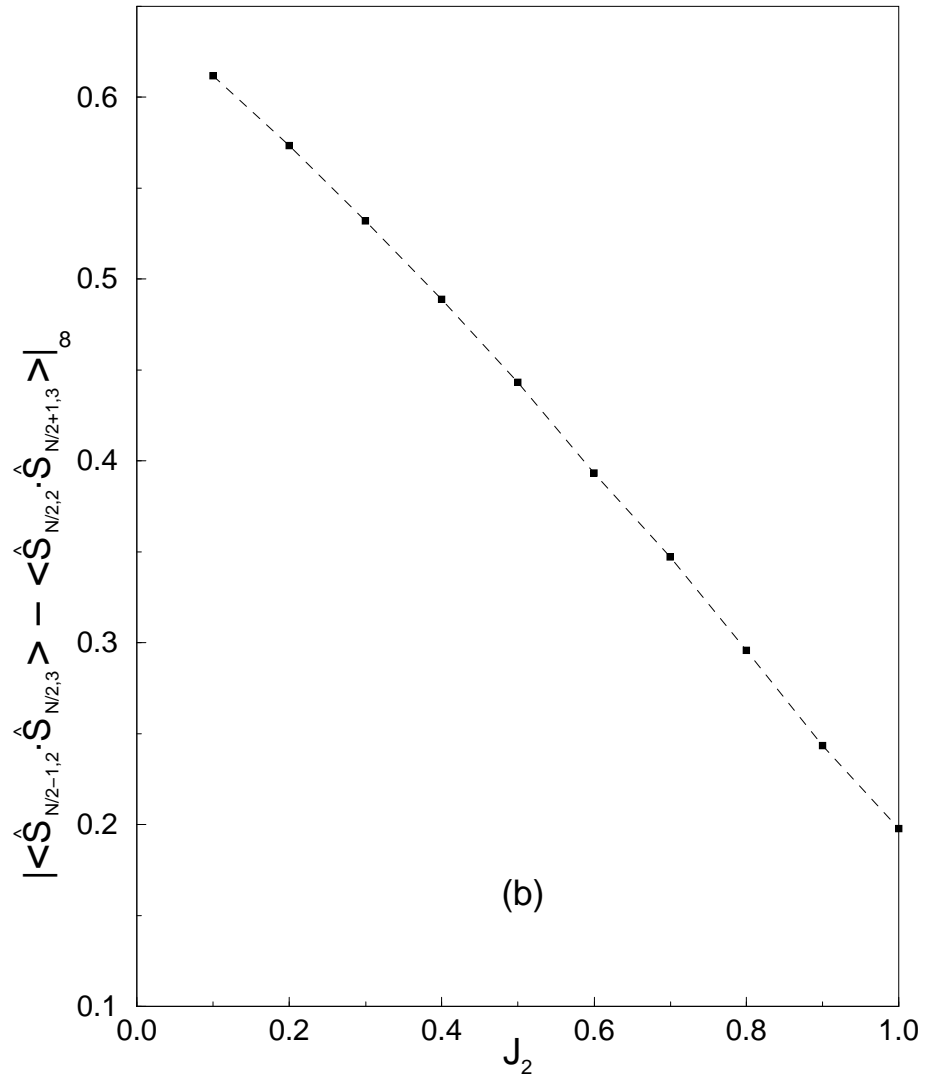


Fig. 8 (b)

Fig. 9

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