Dimerization of Ferrimagnets on Chains and Square Lattices

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(1998)

Abstract

A linear spin wave analysis of dimerization of alternating Heisenberg system with spins $s_1$ and $s_2$ on linear chain as well as square lattice is presented. Among the several possible dimerized configurations considered in two dimensions the plaquette configuration is found to be energetically the most favored one. Inclusion of a variable nearest neighbor exchange coupling $J(a) = \frac{\delta}{a}$ leads to a uniform power law behavior: that is to say, the same $\delta$-dependence is found (i) in chains as well as in square lattices; (ii) in systems consisting of different pairs of spins $s_1$ and $s_2$; (iii) for the magnetic energy gain, the energy gap, the energy of the gapped magnetic excitation mode as well as for the sublattice magnetization; (iv) for all the configurations of the square lattice; and (v) in the entire range of $\delta : (0 \leq \delta < 1)$. The variable exchange coupling also allows the energy of the gapped excitation spectrum to be $\delta$-dependent even in the linear spin wave theory.

PACS numbers: 75.10.Jm, 74.65.+n, 75.50.Ee

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I. INTRODUCTION:

Extensive interest is being currently shown in alternating spin systems consisting of two sublattices with unequal spin magnitudes $s_1$ and $s_2$ with a net non-zero spin per unit cell, as shown in Fig.1. Such systems are realized in bi-metallic chains with the general formula of $ACu(pbaOH)(H_2O)_22H_2O$ where $pba$ OH is 2-hydroxy-1,3-propylenebis (oxamato) and $A = Mn, Fe, C, Ni$. These ferrimagnetic chains are also referred to as alternating or mixed spin chains and are regarded as Heisenberg systems [2,3].

Alternating spin systems have been studied extensively by various techniques: by spin wave theory (SWT) [4–10], spin wave expansion (SWE) [1,12], Monte Carlo (MC) method [4,8–10], density matrix renormalization group (DMRG) technique [5,10,12], the method of matrix product (MP) states [3] and by exact diagonalization (ED) method [5,12].

For an alternating-spin chain the zero temperature ground state energy and sublattice magnetization were evaluated using SWT [3,10,11], SWE [1,12], DMRG [5,11,12] and QMC [8]. The results are summarized in Table 1. The linear spin wave theory gives higher values for the ground state energy and lower values for the sub-lattice magnetization compared to the more exact methods. Recently Ivanov et al. [1,12] used a second-order spin wave expansion to calculate ground state energy as well as sublattice magnetization. Their results differ by 0.03% for the ground state energy and 0.2% for the sublattice magnetization from the DMRG results, as shown in Table 1.

The thermal behavior was also investigated for ferrimagnetic chains [3,3]. Besides verifying the evidence of two (gapped and gapless) excitation modes, the specific heat and magnetic susceptibility of ferrimagnetic chains were also shown to depend upon temperature as $T^{1/2}$ and $T^{-2}$ respectively at low temperatures [10]. It was also shown that this model behaved as a ferromagnet at low temperature, but as a gapped antiferromagnet at moderate temperatures.

Modified spin wave theory, which includes Takahashi constraint, was also shown to give results in surprisingly good agreement with those from quantum Monte Carlo method in the thermodynamic limit of this system [3,10].

Dimerization of chains with spins $s_1$ and $s_2$ ($s_1 > s_2$) on alternating sites was recently [3] studied using the Hamiltonian

$$H = J \sum_n \left((1 + \delta) S_{1,n} \cdot S_{2,n} + (1 - \delta) S_{2,n} \cdot S_{1,n+1}\right),$$

(1)

where the total number of sites (or bonds) is $2N$ and the sum is over the total number of unit cells $N$. $\delta$ is the dimerization parameter and is taken to vary between 0 and 1. Linear spin wave theory and DMRG were used [3,10] to investigate the ground and low-lying excited states for both uniform and dimerized chains. In both the methods the ground state was found to be ferrimagnetic. One point of focus for us in the study of chains is that the LSW theory with the Hamiltonian in Eq.(1) showed that the energy gap at $k = 0$ in the gapped mode did not depend on the dimer parameter $\delta$, while the DMRG predicted an almost linear dependence [3,10]. The DMRG results on chains also show that the transition to a spin-Peierls state is conditional in that the ground state energy depends upon the dimerization parameter as $\delta^\nu$ with $\nu=2 \pm 0.01$.

This has motivated us to investigate a dimerized alternating spin Heisenberg model by using a linear spin wave theory using an ansatz of a variable nearest neighbor exchange coupling that was recently used to study dimerization in a uniform (single spin) Heisenberg system [13]. We would also like to extend our work to square lattices.

A need for describing nearest neighbor exchange interaction as dimerization sets in two dimensional lattices was recently discussed [13]. Among the various lattice deformation modes which allow for dimerization, some require that the effect of the ensuing elongation or contraction of nearest neighbor distances be reflected in the nearest neighbor spin-spin exchange couplings. Since an exchange integral for a nearest neighbor distance $a$ is roughly [14]

$$J(a) = \frac{J}{a},$$

(2)

we assume that when the nearest neighbor distances change from $a$ to $a(1 + \delta)$, the exchange couplings change from $J$ to $J/a$. Thus, to linear order in $\delta$, the interaction $\frac{J}{a}$ has the familiar form $J(1 + \delta)$. The form in Eq.(2) allows for incorporating changes in the nn exchange couplings in various situations of lattice deformations. It was shown [13] that the logarithmic $\delta$-dependence of various quantities like the gain in the magnetic ground state energy, etc., $\delta g$ $\frac{\delta}{\mu a}$, both in one and two dimensional lattices, can also be a result of using this variable nearest neighbor spin-spin
exchange coupling. The form in Eq. (3) gives a logarithmic dependence over not just the $\delta \to 0$ regime, but over the entire range of $\delta$ from 0 to 1. In what follows, we shall use for exchange interaction the form in Eq. (3).

In this paper we will study alternating spin systems formed with different pairs of spin values; $\frac{1}{2}$, 1, and $\frac{3}{2}$ using a zero temperature linear spin wave theory. We have considered three alternating spin systems from these spin values: $(1, \frac{1}{2})$ (denoting $s_1 = 1$ and $s_2 = \frac{1}{2}$); $(\frac{3}{2}, \frac{1}{2})$ and $(\frac{3}{2}, 1)$. We would like to see the effect of including the variable nearest neighbor exchange coupling on the $\delta$-dependence of the physical quantities such as the gain in magnetic energy, the sub-lattice magnetization and energies of the excitation modes in both one and two space-dimensions and for different spin systems. We would also like to see if it gives a $\delta$-dependence of the gapped excitation energy mode. In section two we will study these three alternating spin systems for a Heisenberg linear chain using the variable nearest neighbor exchange coupling. The energy and magnetization of such systems will be computed using LSW theory. Critical exponents of the dimer alternating chains will also be calculated. We shall then study alternating spin systems on a square lattice for several proposed dimer configurations in section III.

The question of frustration in a ferrimagnetic chain or a ladder due to an antiferromagnetic second neighbor interaction has also received some attention recently. It was shown that a strong frustration leads to disappearance of the long range ferrimagnetic order through a discontinuous transition to a singlet state [12,15,16]. It was also shown that the spin wave theory can predict realistic results for a frustrated system at least for the case of weak frustration [12]. We shall attempt to see the effect of including the variable nearest neighbor exchange interaction on weakly frustrated chains and square lattices in a future publication.

II. ONE DIMENSIONAL ALTERNATING SYSTEM

The alternating dimer Hamiltonian on a chain with two spins $s_1$ and $s_2$ can be rewritten using the variable nearest neighbor exchange coupling defined in Eq. (3) as

$$H = \sum_i \left[ \frac{J}{1+\delta} S_{1,2i} \cdot S_{2,2i+1} + \frac{J}{1-\delta} S_{2,2i+1} \cdot S_{1,2i+2} \right]$$ (3)

A linear spin-wave analysis is usually performed with the help of Holstein-Primakoff (HP) transformations to bosonic spin-deviation operators. For the two sublattices the HP transformations are: for spin-$s_1$

$$S_{1,n}^+ = (2s_1 - a_n^\dagger a_n)^{1/2} a_n$$ (4a)
$$S_{1,n}^- = a_n^\dagger (2s_1 - a_n^\dagger a_n)^{1/2}$$ (4b)
$$S_{1,n}^z = s_1 - a_n^\dagger a_n$$ (4c)

and for the second sublattice with spin-$s_2$

$$S_{2,n}^+ = b_n^\dagger (2s_2 - b_n^\dagger b_n)^{1/2}$$ (5a)
$$S_{2,n}^- = (2s_2 - b_n^\dagger b_n)^{1/2} b_n$$ (5b)
$$S_{2,n}^z = b_n^\dagger b_n - s_2$$ (5c)

where $s_i$ is the magnitude of the spin on sublattice $i$. A linearized Hamiltonian is obtained by substituting HP transformations into Eq. (3) and keeping terms up to the quadratic order in the spin-deviation operators $a$ and $b$. The linearized Hamiltonian in Fourier transformed variables is

$$H = \sum_k \left[ A_1 a_k^\dagger a_k + A_2 b_k^\dagger b_k + B(k) \left( a_k^\dagger b_k^\dagger + b_k a_k \right) + C \right]$$ (6)

with

$$A_1 = J_p s_2$$ (7a)
$$A_2 = J_p s_1$$ (7b)
$$B(k) = \Lambda_k \sqrt{s_1 s_2}$$ (7c)
$$C = -J_p s_1 s_2.$$ (7d)
Here

\[ \Lambda_k = \sqrt{(J_p \cos(k))^2 + (J_m \sin(k))^2} \]  
\[ J_p = \frac{J}{1 - \delta^2} \]

and

\[ J_m = J_p \delta \]

The linearized Hamiltonian in Eq.(6) can be diagonalized using Bogoliubov transformations

\[ a_k = u_k \alpha_k + v_k \beta_k \]  
\[ b_k = u_k \beta_k + v_k \alpha_k \]

to

\[ \tilde{H} = \sum_k \left[ \varepsilon_g + E_1(k) \alpha_k \alpha_k^\dagger + E_2(k) \beta_k \beta_k^\dagger \right] \]

where the coefficients \( u_k, v_k \) are constrained by the condition \( u_k^2 - v_k^2 = 1 \), \( \alpha_k \) and \( \beta_k \) are the normal mode boson operators, \( E_1(k) \) and \( E_2(k) \) are the energies of the two excitation modes and \( \varepsilon_g \) is the ground state energy per site, with \( u(k) \) and \( v(k) \) defined as

\[ u(k) = \sqrt{\frac{A_1 + A_2 + \xi_k}{2\xi_k}} \]  
\[ v(k) = \sqrt{\frac{A_1 + A_2 - \xi_k}{2\xi_k}} \]

\[ \xi_k = \sqrt{(A_1 + A_2)^2 - 4B(k)^2} \]

The two excitation modes are

\[ E_1(k) = (A_1 - A_2 + \xi_k) / 2 \]  
\[ E_2(k) = (A_2 - A_1 + \xi_k) / 2 \]

It is easy to see that \( E_1 \) is the gapless mode and \( E_2 \) has a gap.

The ground state energy per site \( \varepsilon_g \) is given by

\[ \varepsilon_g = C - A_1 - A_2 + \sum_k \xi_k \]

and the staggered magnetization in the two sub-lattices corresponding to the spins \( s_1 \) and \( s_2 \) respectively is

\[ M_1 = S_1 - < D > \]  
\[ M_2 = < D > - S_2 \]

with \( < D > = < a_i^\dagger a_i > = < b_j^\dagger b_j > \) is the average taken in the ground state, which is the Neel state, at zero temperature. This average of spin deviation operators can be determine from

\[ < D > = \frac{1}{N} \sum_k v^2(k) \]

with \( k \) running over half the Brillouin zone.
For the three kinds of alternating spin chains, referred to as \((1, \frac{1}{2})\), \((\frac{3}{2}, \frac{1}{2})\) and \((\frac{3}{2}, 1)\), the ground state energy, the excitation energies and magnetization can now be calculated as functions of the dimerization parameter \(\delta\). Previous calculations invariably took spin-spin exchange couplings alternately as \(J(1 \pm \delta)\), which, as mentioned above, can be taken as an expansion of the interaction in Eq. (2) to the first order in \(\delta\), implying that the results are valid only in the critical regime \(\delta \to 0\). The advantage of taking the variable nearest neighbor exchange coupling is that the results will then be valid also in the limit \(\delta \to 1\).

The ground state energies \(\varepsilon_g\) per site for undimerized chains, \(\delta = 0\), were found, expectedly, to be the same as those found earlier \[\text{(1)}\] for the three systems. After including dimerization, our calculations also confirm that the ground state energy of all the three systems described above decreases with \(\delta\). This is shown in Figure 2, where energy gain \(\varepsilon_g(\delta) - \varepsilon_g(0)\) is plotted against \(\delta\). Numerical fitting shows that, as against earlier results, the magnetic energy gain \(\varepsilon_g(\delta) - \varepsilon_g(0)\) has a logarithmic dependence on \(\delta\), \(\varepsilon_g(\delta) \sim \ln(\delta(0))\), for the three systems discussed here, with values of \(\nu\) between 1.4 – 1.6 in the entire range \(0 \leq \delta < 1\). Fig. 2 shows that the chain \((\frac{3}{2}, 1)\) has higher gain than the other two systems.

As expected, our calculations also find two branches of the excitation spectrum, one gapless and the other with a gap at \(k = 0\), in the three systems.

As stated above, the LSW theory with the spin-spin exchange coupling \(J(1 \pm \delta)\) in the presence of dimerization allowed no \(\delta\)-dependence of the energy gap in the second mode, while the DMRG found almost a linear \(\delta\)-dependence for these spin systems \[\text{(2)}\]. We find that by including a variable nearest neighbor coupling constant defined in Eq. (2) the spin wave theory also allows for a \(\delta\)-dependent energy gap \(\Delta(\delta) = E_2(\delta) - \varepsilon_g(\delta)\) in the second mode. This is because the \(\delta\)-dependent terms do not now cancel out for the gapped excitation mode at \(k = 0\) as they did with the coupling \(J(1 \pm \delta)\). The dependence is found to follow the same logarithmic behavior, \(\varepsilon_g(\delta) \sim \ln(\delta(0))\), as the ground state energy, with \(\nu\) varying between 1.4 – 1.6 for the entire range \(0 \leq \delta < 1\). This is true for all the three spin systems defined here, and is larger for the \((\frac{3}{2}, 1)\) system than the other two (see figure 3).

The staggered magnetization \(M(\delta)\) was also found to follow the logarithmic \(\delta\)-dependence up to \(\delta \leq 0.5\), but follows a different behavior for \(\delta > 0.5\). \(M_1(\delta)\) against dimerization is shown in Fig. (4) for the three systems. Again we see from Fig. (4) that the chain with \((\frac{3}{2}, 1)\) has higher value of magnetization than the other two systems.

It is worth mentioning here that by using coupled cluster method \[\text{(3)}\], we had found that the ground state energy and the staggered magnetization of a spin-half Heisenberg chain follow the same logarithmic behavior using the variable exchange coupling defined in Eq. (2), in both small and large values of dimerization \(\delta\). This gives us more confidence about the results we have obtained by LSW theory.

### III. TWO DIMENSIONAL ALTERNATING HAMILTONIAN :-

The dimerization on two dimensions lattices differs from that on chains. There are several ways in which distortions of a square lattice can occur, each one of the possible configurations giving a different dependence of the ground state energy on the dimerization parameter \[\text{(4)}\].

We will use some of these configurations, illustrated in Fig. (5), to study the alternating spin square lattices. The alternating dimerized Hamiltonian for a two dimensional system can be written in general as

\[
H = \sum_{i,j} \sum_{\mu=\pm 1} \left[ J_{x,\mu} S_{1,i,j} \cdot S_{2,i+\mu,j} + J_{y,\mu} S_{1,i,j} \cdot S_{2,i,j+\mu} \right]
\]

where the indices 1 and 2 on the spin vectors refer to the two sublattices with spins of magnitude \(s_1\) and \(s_2\). For reasons described earlier, we use variable nearest neighbor exchange couplings. These are defined for different configurations as follows:

- **Configuration (a)**
  \[
  J_{x,\mu} = \frac{J}{(1+\mu \delta)} \approx J(1-\mu \delta)
  \]
  \[
  J_{y,\mu} = J.
  \]
Configuration (b)
\[
J_{x,\mu} = \frac{J}{1 + \mu \delta} \simeq J(1 - \mu \delta)
\]
\[
J_{y,\mu} = \frac{J}{\sqrt{\delta^2 + (1 + \mu \delta)^2}} \simeq J(1 - \delta^2 / 2)
\]

Configuration (c)
\[
J_{x,\mu} = J_{y,\mu} = \frac{J}{1 + \mu \delta} \simeq J(1 - \mu \delta)
\]

Configuration (d)
\[
J_{x,\mu} = \frac{J}{1 + \mu \delta} \simeq J(1 - \mu \delta)
\]
\[
J_{y,\mu} = \frac{J}{\sqrt{\delta^2 + (1 + \mu \delta)^2}} \simeq J \left(1 - \mu \delta - (1 - \frac{\delta^2}{2}) \delta^2\right)
\]

Configuration (e)
\[
J_{x,\mu} = J_{y,\mu} = \frac{J}{\sqrt{\delta^2 + (1 + \mu \delta)^2}} \simeq J \left(1 - \mu \delta - (1 - \frac{\delta^2}{2}) \delta^2\right)
\]

We would like to investigate the five configurations resulting from dimerization of a square lattice in order to see (i) which one of these leads to the largest gain in magnetic energy as the dimerization sets in, (ii) if the use of variable exchange coupling leads to a single power law behavior valid for the entire range of \(\delta\), and how the law differs from that in the case of chains, (iii) the \(\delta\)-dependence of the second mode of excitation \(E_2\), (iv) the behavior of staggered magnetization, and (v) the generality of these investigations regarding the three spin systems discussed here.

The linear spin wave analysis follows the same procedure as for the chain above. The same equations are applicable in this case, but the various coefficients entering the theory have now the following values:

\[
A_1 = J_p s_2
\]
\[
A_2 = J_p s_1
\]
\[
B(k) = \Gamma(k) \sqrt{s_1 s_2}
\]
\[
C = -J_p s_1 s_2
\]

where
\[
\Gamma(k) = \sqrt{(J_{px} \cos(k_x) + J_{py} \cos(k_y))^2 + (J_{mx} \sin(k_x) + J_{my} \sin(k_y))^2}
\]

and
\[
J_p = (J_{x,+1} + J_{x,-1} + J_{y,+1} + J_{y,-1})/4,
\]
\[
J_{px} = (J_{x,+1} + J_{x,-1})/4,
\]
\[
J_{py} = (J_{y,+1} + J_{y,-1})/4,
\]
\[
J_{mx} = (J_{x,+1} - J_{x,-1})/4,
\]
\[
J_{my} = (J_{y,+1} - J_{y,-1})/4.
\]

The ground state energy \(\varepsilon_g(\delta)\) defined in Eq.(15), energies of the two excitation modes \(E_i(k)\) in Eq.(14) and staggered magnetization \(M(\delta)\) defined in Eqs.(19) can now be calculated as functions of the dimerization parameter \(\delta\).

The ground state energy \(\varepsilon_g(\delta = 0)\) is found to be \(-1.2, -1.7158\) and \(-3.3709\) for the three spin systems \((1, \frac{1}{2}), (\frac{3}{2}, \frac{1}{2})\) and \((\frac{3}{2}, 1)\) respectively. Staggered magnetization \(M_1\{M_2\}\) on the first \{second\} sublattice is \(0.8907 \{-0.3907\}, 1.4241 \{-0.4241\}\) and \(1.3597 \{-0.8597\}\) for the three systems . These values are listed in Table 2.

Our calculations confirm that, like in chain, the gain in magnetic energy increases with \(\delta\) in all the proposed configurations. This is shown in Figure 6, where the energy gain \(\varepsilon_g(\delta) - \varepsilon_g(0)\) is plotted against \(\delta\) for the five configurations. It also shows that the plaquette configuration of Fig. 5(c) is energetically the most favorable state, while there is hardly a discernible difference among the configurations (a), (b) and (d). It is also interesting to note that the magnetic energy gain under dimerization of an alternating spin square lattice also varies as \(\frac{\delta^\nu}{\ln \delta}\) with \(\nu = 1.4 - 1.6\) in the entire range \(0 \leq \delta < 1\), exactly as in the case of a chain. This is singularly an effect of taking the variable exchange
coupling defined above.

The $\delta$ dependence of the energy gap, $\Delta(\delta) = E_2(\delta) - \varepsilon_0(\delta)$, for the five configurations is shown in Fig. 7, showing greater stabilization of the dimerized state with increasing $\delta$. We also find that, like the magnetic energy gain, the energy gap increases with $\delta$ as $\delta^{\nu|\ln \delta|}$ in the small $\delta$ regime for all the five configurations with $\nu = 1.4 - 1.6$. The configurations (a) - (d) also have the same dependence on $\delta$ in the entire range of $\delta$. The difference between the dimerization of a square lattice for these configurations is again markedly brought out in Fig. 7.

Our calculations give staggered magnetization for the un-dimerized alternating spin square lattice $M_1(\delta = 0) = 0.8907, 1.4241$ and $1.3597$ and $M_2(\delta = 0) = -0.3907, -0.4241$ and $-0.8597$ for $(1, \frac{1}{2})$, $(\frac{3}{2}, \frac{1}{2})$ and $(\frac{3}{2}, 1)$ respectively. As dimerization sets in, magnetization decreases in all the configurations we have chosen, as shown in Fig. 8. This is also the case for the entire range $0 \leq \delta < 1$, except in the case of configuration (e) for which the magnetization rises again after $\delta > \frac{1}{2}$.

Configuration (e) is peculiar in the sense that $\delta = \frac{1}{2}$ is a special point for it; the shorter bond length is symmetric about this point, having a minimum value of $\frac{1}{\sqrt{2}}$. At this point the distortions give rise to a rectangular lattice with sides $\sqrt{2}$ and $\frac{1}{\sqrt{2}}$. The energy gain increases with $\delta$ up to $\delta = \frac{1}{2}$, and then decreases.

For all the five configurations, we found that the magnetization also varies as $\delta^{\nu|\ln \delta|}$ in the small $\delta$ regime with the exponent $\nu = 1.4 - 1.6$, exactly as the energy gain and the energy gap. However, while for configurations (a-d) in the full range $0 \leq \delta < 1$ the magnetization follows the same power law with the exponents as $\nu = 1.4 - 1.6$, configuration (e) has a distinctly different behavior in this regime.

In summary, we have studied the spin-Peierls dimerization of an alternating spin Heisenberg system on a chain and a square lattice taking variable exchange couplings based on the ansatz $J(\alpha) = J_{a}$, for three kind of alternating spin systems, namely $(1, \frac{1}{2})$, $(\frac{3}{2}, \frac{1}{2})$ and $(\frac{3}{2}, 1)$. We have included different possibilities of dimerization in the case of a square lattice. The ground state energy as well as staggered magnetization decrease continuously with increasing dimerization in both 1D and 2D. In 2D, the plaquette configuration with dimerization taking place simultaneously along both the principal square axes has markedly lower ground state energy and magnetization than the other configurations; (a), (b), (d) and (e). The plaquette configuration stands out as the most favored mode of dimerization. The energy gap also corroborates the above conclusions. It has also been shown that the magnetic energy gain, energy gap and staggered magnetization follow a uniform dependence upon the dimerization parameter $\delta$ as $\delta^{\nu|\ln \delta|}$ (i) in chains as well as in square lattices; (ii) in systems consisting of different pairs of spins $s_1$ and $s_2$; (iii) for the magnetic energy gain, the energy gap, the energy of the gapped magnetic excitation mode as well as for the sublattice magnetization; (iv) for all the configurations of the square lattice; and (v) in the entire range $0 \leq \delta < 1$. The variable exchange coupling also allows the energy of the gapped excitation spectrum to be $\delta$-dependent even in the linear spin wave theory.

We would like to thank to Dr. D. Sen for providing Refs. 5 and 6.
In order to take a reasonably correct account of the energetics, there is a need to know how the exchange integral $J$ depends upon the varying distances in the process of dimerization. There is, however, no way to find an exact $r$-dependence of $J(r)$. What is known is that it should fall off rapidly as distance increases. In the nearest neighbour model that we are considering, the exchange integral is taken to be appreciable only over the nearest neighbour distance $a$, in which case $J$ is proportional to $1/a$ (see, for example, Spin Waves by A. I. Akhiezer, V. G. Bar'yakhtar and S. V. Peletminskii, translation edited by S. Doniach, North Holland, Amsterdam, 1968, p 9). It thus seems reasonable to assume that if the nearest neighbour distance $a$ changes to $a'$ due to dimerization, then the changed $J$ is proportional to $1/a'$. This is the ansatz we are using.
Figure captions

Figure 1: A schematic sketch of an alternating spin chain. The larger and smaller arrows indicate the larger ($s_1$) and the smaller ($s_2$) spins. The hollow (filled) circles represent the positions of spins in the undisturbed (dimerized) chain.

Figure 2: The magnetic energy gain $\varepsilon_g(\delta) - \varepsilon_g(0)$ vs the dimerization parameter $\delta$ for 1D alternating spin chain in the full range of the dimerization parameter $0 \leq \delta < 1$.

Figure 3: $\delta$-dependence of the energy gap as dimerization sets in for different alternating spin chains.

Figure 4: $\delta$-dependence of the staggered magnetization for one of the two sublattices, $S_1$, for the three alternating spin chains.

Figure 5: Five configurations for the dimerization of a square lattice. (a) a columnar configuration caused by a longitudinal ($\pi, 0$) static phonon; (b) a staggered configuration caused by a ($\pi, \pi$) static phonon with polarization along $x$-direction. Like (a), the dimerization occurs along one direction only, but the sequence of alternate couplings itself alternates along the other direction; (c) dimerization along both the directions, caused by ($\pi, 0$) and ($0, \pi$) phonons, making a plaquette of four nearest neighbour spins; (d) again, dimerization along both the directions, but taken staggered along the vertical direction; (e) another staggered dimerization that is caused by a longitudinal ($\pi, \pi$) phonon. Large arrow belongs to the first sublattice and short one belongs to second sublattice. And the open circles indicate the square lattice sites and the solid ones show the dimerized lattice.

Figure 6: The gain in magnetic energy $\varepsilon_g(\delta) - \varepsilon_g(0)$ as dimerization sets in with increasing $\delta$ for the five configurations of a square lattice in the range $0 \leq \delta < 1$ for (a) spin $(1, \frac{1}{2})$, (b) spin $(\frac{3}{2}, \frac{1}{2})$ and (c) spin $(\frac{3}{2}, 1)$.

Figure 7: Dependence of the energy gap $\Delta$ on $\delta$ for the five dimerization configurations of the alternating square lattices for (a) spin $(1, \frac{1}{2})$, (b) spin $(\frac{3}{2}, \frac{1}{2})$ and (c) spin $(\frac{3}{2}, 1)$.

Figure 8: $\delta$-dependence of the staggered magnetization of an alternating spin square lattice calculated for the five dimerization configurations. (a) spin $(1, \frac{1}{2})$, (b) spin $(\frac{3}{2}, \frac{1}{2})$ and (c) spin $(\frac{3}{2}, 1)$.
Table 1: Summary of the ground state energy per site and sublattice magnetization values calculated by different methods for alternating spin chains made up of the three spin systems namely $(1, \frac{1}{2})$, $(\frac{3}{2}, \frac{1}{2})$ and $(\frac{3}{2}, 1)$.

| Spin system | Method | $\varepsilon_g$ | $M_1$ | $M_2$ |
|-------------|--------|-----------------|-------|-------|
| $(1, \frac{1}{2})$ | MP     | $-0.7245$       | 0.779 | $-0.279$ |
|           | QMC    | $-0.7275$       | 0.793 | $-0.293$ |
|           | LSWT   | $-0.718$        | 0.695 | $-0.195$ |
|           | DMRG   | $-0.72709$      | 0.79428 | $-0.29248$ |
|           | SWE    | $-0.72715$      | 0.79388 |       |
|           | MSW    |                |       | $-0.7295$ |
| $(\frac{3}{2}, \frac{1}{2})$ | LSWT   | $-0.979$        | 1.315 | $-0.314$ |
|           | DMRG   | $-0.98362$      | 1.35742 | $-0.35742$ |
|           | SWE    | $-0.9834$       | 1.3666 |       |
| $(\frac{3}{2}, 1)$ | LSWT   | $-1.914$        | 1.040 | $-0.540$ |
|           | DMRG   | $-1.93096$      | 1.14427 | $-0.644$ |
|           | SWE    | $-1.9316$       | 1.1461 |       |

Table 2: The ground state energy per site and the staggered magnetization of the undimerized alternating spin square lattice for the three spin systems as calculated in the linear spin wave theory.

| Spin system | $\varepsilon_g$ | $M_1$ | $M_2$ |
|-------------|-----------------|-------|-------|
| $(1, \frac{1}{2})$ | $-1.2$       | 0.8907 | $-0.3907$ |
| $(\frac{3}{2}, \frac{1}{2})$ | $-1.7158$     | 1.4244 | $-0.4241$ |
| $(\frac{3}{2}, 1)$ | $-3.3709$     | 1.3597 | $-0.83597$ |
Figure 1
Fig. 2

$\varepsilon_g(\delta) - \varepsilon_g(0)$ vs $\delta$

- Spin(1.1/2)
- Spin(3/2,1/2)
- Spin(3/2,1)
Fig 3

$W_2(\delta) - \varepsilon_g(\delta)$ vs $\delta$

- □ - Spin(1, 1/2)
- • - Spin(3/2, 1/2)
- △ - Spin(3/2, 1)
Fig. 4

- Spin(1,1/2)
- Spin(3/2,1/2)
- Spin(3/2,1)

$M(0) - M(\delta)$

$\delta$
Figure 5 (a)

Figure 5 (b)
\[ a(1-\delta) \]

Figure 5 (c)

\[ a\sqrt{\delta^2 + (1+\delta)^2} \]

\[ a\sqrt{\delta^2 + (1-\delta)^2} \]

Figure 5 (d)
\[ a\sqrt{\delta^2+(1+\delta)^2} \]

\[ a\sqrt{\delta^2+(1-\delta)^2} \]

\[ a\sqrt{\delta^2+(1-\delta)^2} \]

\[ a\sqrt{\delta^2+(1+\delta)^2} \]
For Spin(1,1/2)
- Config.(a)
- Config.(b)
- Config.(c)
- Config.(d)
- Config.(e)
Fig. 6(b)
Fig. 6(c)

For Spin(3/2,1)
- ■ - Config.(a)
- ● - Config.(b)
- ▲ - Config.(c)
- ▼ - Config.(d)
- ♦ - Config.(e)

$\varepsilon_g(\delta) - \varepsilon_g(0)$ vs $\delta$
Fig. 7(a)

For Spin(1, 1/2)
- Config.(a)
- Config.(b)
- Config.(c)
- Config.(d)
- Config.(e)
Fig. 7(b)

For Spin(3/2,1/2)

- ■ Config.(a)
- ● Config.(b)
- ▲ Config.(c)
- ▼ Config.(d)
- ♦ Config.(e)

\[ W_2(\delta) - \varepsilon_g(\delta) \]
For Spin(3/2,1)

$W_2(\delta) - \varepsilon_g(\delta)$

Fig. 7(c)
Fig. 8(a)

For Spin(1,1/2)

- ■ Config.(a)
- ● Config.(b)
- ▲ Config.(c)
- ▼ Config.(d)
- ♦ Config.(e)

\[ M(0) - M(\delta) \]
For Spin(3/2,1/2)

- Config.(a)
- Config.(b)
- Config.(c)
- Config.(d)
- Config.(e)

Fig. 8(b)
Fig. 8(c)

For Spin(3/2,1)

- ■ Config.(a)
- ● Config.(b)
- ▲ Config.(c)
- ▼ Config.(d)
- ● Config.(e)

$M(0) - M(\delta)$

$\delta$