

## Dimerization of ferrimagnets on chains and square lattices

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

1999 J. Phys.: Condens. Matter 11 465

(<http://iopscience.iop.org/0953-8984/11/2/012>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 171.66.16.210

The article was downloaded on 14/05/2010 at 18:28

Please note that [terms and conditions apply](#).

## Dimerization of ferrimagnets on chains and square lattices

Aiman Al-Omari<sup>†</sup> and A H Nayyar<sup>‡</sup>

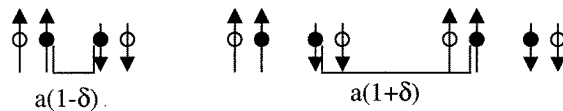
Department of Physics, Quaid-i-Azam University, Islamabad 45320, Pakistan

Received 26 June 1998, in final form 22 September 1998

**Abstract.** A linear spin-wave analysis of the dimerization of an alternating Heisenberg system with spins  $s_1$  and  $s_2$  on a linear chain as well as on a square lattice is presented. Among the several possible dimerized configurations considered in two dimensions, the plaquette configuration is found to be energetically the most favoured one. Inclusion of a variable nearest-neighbour exchange coupling  $J(a) = J/a$  leads to a uniform power-law behaviour—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 of the configurations of the square lattice; and (v) over 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.

### 1. Introduction

Extensive interest is currently being 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 figure 1. Such systems are realized in bi-metallic chains with the general formula  $ACu(pbaOH)(H_2O)_3 \cdot 2H_2O$  where pbaOH is 2-hydroxy-1, 3-propylenebis (oxamato) and  $A = Mn, Fe, C, Ni$  [1]. These ferrimagnetic chains are also referred to as alternating or mixed-spin chains and are regarded as Heisenberg systems [2–6].



**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.

Alternating-spin systems have been studied extensively by various techniques: by spin-wave theory (SWT) [4–10], spin-wave expansion (SWE) [11, 12], Monte Carlo (MC) methods [4, 8–10], the density matrix renormalization group (DMRG) technique [5, 10, 12], the method of matrix product (MP) states [3], and by the exact-diagonalization (ED) method [9, 12].

For an alternating-spin chain, the zero-temperature ground-state energy and sublattice magnetization were evaluated using SWT [5, 6, 10], SWE [11], DMRG [5, 6, 10], and QMC

<sup>†</sup> E-mail address: aiman@physics.sdnpk.undp.org.

<sup>‡</sup> E-mail address: nayyar@physics.sdnpk.undp.org.

**Table 1.** A 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	Method	$\varepsilon_g$	$M_1$	$M_2$
$(1, \frac{1}{2})$	MP [3]	-0.7245	0.779	-0.279
	QMC [3,4]	-0.7275	0.793	-0.293
	LSWT [4, 6]	-0.718	0.695	-0.195
	DMRG [6]	-0.727 09	0.794 28	-0.292 48
	SWE [11, 12]	-0.727 15	0.793 88	
	MSW [10]	-0.7295		
$(\frac{3}{2}, \frac{1}{2})$	LSWT [6]	-0.979	1.315	-0.314
	DMRG [6]	-0.983 62	1.357 42	-0.357 42
	SWE [11, 12]	-0.9834	1.3666	
$(\frac{3}{2}, 1)$	LSWT [6]	-1.914	1.040	-0.540
	DMRG [6]	-1.930 96	1.144 27	-0.644
	SWE [11, 12]	-1.9316	1.1461	

methods [3]. 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 sublattice magnetization compared to the more exact methods. Recently, Ivanov *et al* [11, 12] used a second-order spin-wave expansion to calculate the ground-state energy as well as the 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 behaviour was also investigated for ferrimagnetic chains [5, 6, 8–10]. Besides verifying the existence 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 [8, 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 the Takahashi constraint, was also shown to give results in surprisingly good agreement with those from the quantum Monte Carlo method in the thermodynamic limit of this system [8, 10].

Dimerization of chains with spins  $s_1$  and  $s_2$  ( $s_1 > s_2$ ) on alternating sites was recently [5, 6] studied using the Hamiltonian

$$H = J \sum_n [(1 + \delta)S_{1,n}S_{2,n} + (1 - \delta)S_{2,n}S_{1,n+1}] \quad (1)$$

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 the DMRG were used [5, 6] to investigate the ground and low-lying excited states for both uniform and dimerized chains. In both 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 equation (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 [5, 6]. 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-neighbour 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-neighbour exchange interaction as dimerization sets in for 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-neighbour distances be reflected in the nearest-neighbour spin–spin exchange couplings. Since an exchange integral for a nearest-neighbour distance  $a$  is roughly [14]

$$J(a) = \frac{J}{a} \quad (2)$$

we assume that when the nearest-neighbour distances change from  $a$  to  $a(1 \pm \delta)$ , the exchange couplings change from  $J$  to  $J/(1 \pm \delta)$ . Thus, to linear order in  $\delta$ , the interaction  $J/(1 \pm \delta)$  has the familiar form,  $J(1 \pm \delta)$ . The form in equation (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 such as the gain in the magnetic ground-state energy,  $\varepsilon_g \sim \delta^v/|\ln \delta|$ , in both one- and two-dimensional lattices, can also be a result of using this variable nearest-neighbour spin–spin exchange coupling. The form in equation (2) gives a logarithmic dependence not just over the  $\delta \rightarrow 0$  regime, but also over the entire range of  $\delta$  from 0 to 1. In what follows, we shall use for the exchange interaction the form in equation (2).

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-neighbour exchange coupling on the  $\delta$ -dependence of the physical quantities such as the gain in magnetic energy, the sublattice magnetization, and the energies of the excitation modes in both one and two space dimensions and for different spin systems. We would also like to see whether it gives rise to a  $\delta$ -dependence of the gapped excitation energy mode. In section 2 we will study these three alternating-spin systems for a Heisenberg linear chain using the variable nearest-neighbour 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 3.

The question of frustration in a ferrimagnetic chain or a ladder due to an antiferromagnetic second-neighbour interaction has also received some attention recently. It was shown that a strong frustration leads to the 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 the variable nearest-neighbour exchange interaction on weakly frustrated chains and square lattices in a future publication.

## 2. The one-dimensional alternating system

The dimer alternating Hamiltonian on a chain with two spins  $s_1$  and  $s_2$  can be rewritten, using the variable nearest-neighbour exchange coupling defined in equation (2), as

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

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  $s_1$ ,

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

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

$$S_{1,n}^z = s_1 - a_n^\dagger a_n \quad (4c)$$

and for the second sublattice, with spin  $s_2$ ,

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

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

$$S_{2,n}^z = b_n^\dagger b_n - s_2 \quad (5c)$$

where  $s_i$  is the magnitude of the spin on sublattice  $i$ . A linearized Hamiltonian is obtained by substituting HP transformations into equation (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

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

with

$$A_1 = J_p s_2 \quad (7a)$$

$$A_2 = J_p s_1 \quad (7b)$$

$$B(k) = \Lambda_k \sqrt{s_1 s_2} \quad (7c)$$

$$C = -J_p s_1 s_2. \quad (7d)$$

Here

$$\Lambda_k = \sqrt{(J_p \cos(k))^2 + (J_m \sin(k))^2} \quad (8)$$

$$J_p = \frac{J}{1 - \delta^2} \quad (9)$$

and

$$J_m = J_p \delta. \quad (10)$$

The linearized Hamiltonian in equation (6) can be diagonalized using Bogoliubov transformations:

$$a_k = u_k \alpha_k + v_k \beta_k^\dagger \quad (11a)$$

$$b_k = u_k \beta_k + v_k \alpha_k^\dagger \quad (11b)$$

to

$$\tilde{H} = \sum_k \left[ \varepsilon_g + E_1(k) \alpha_k^\dagger \alpha_k + E_2(k) \beta_k^\dagger \beta_k \right] \quad (12)$$

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

$$u(k) = \sqrt{\frac{A_1 + A_2 + \xi_k}{2\xi_k}} \quad (13a)$$

$$v(k) = \sqrt{\frac{A_1 + A_2 - \xi_k}{2\xi_k}} \quad (13b)$$

$$\xi_k = \sqrt{(A_1 + A_2)^2 - 4B(k)^2}. \quad (13c)$$

The two excitation modes are

$$E_1(k) = (A_1 - A_2 + \xi_k)/2 \quad (14a)$$

$$E_2(k) = (A_2 - A_1 + \xi_k)/2. \quad (14b)$$

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

$$\varepsilon_g = C - A_1 - A_2 + \sum_k \xi_k \quad (15)$$

and the staggered magnetizations in the two sublattices corresponding to the spins  $s_1$  and  $s_2$  respectively are

$$M_1 = S_1 - \langle D \rangle \quad (16a)$$

$$M_2 = \langle D \rangle - S_2 \quad (16b)$$

where  $\langle D \rangle = \langle a_i^\dagger a_i \rangle = \langle b_j^\dagger b_j \rangle$  is the average taken in the ground state, which is the Néel state, at zero temperature. This average of spin-deviation operators can be determined from

$$\langle D \rangle = \frac{1}{N} \sum_k v^2(k) \quad (17)$$

with  $k$  running over half of the Brillouin zone.

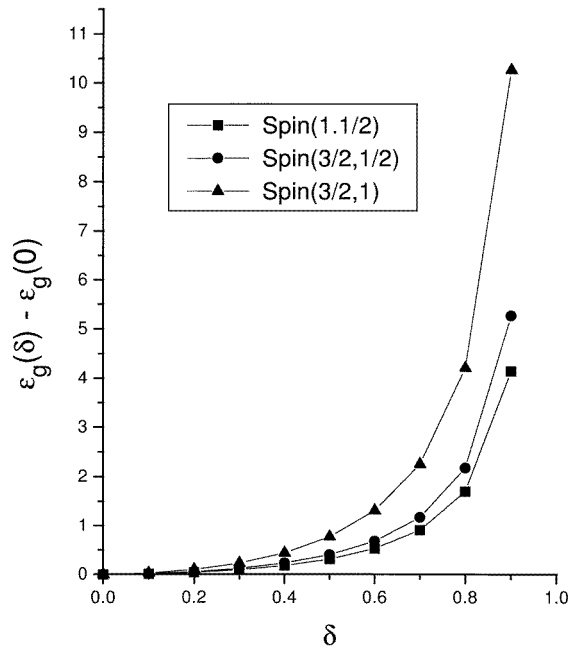
For the three kinds of alternating-spin chain, referred to as  $(1, \frac{1}{2})$ ,  $(\frac{3}{2}, \frac{1}{2})$ , and  $(\frac{3}{2}, 1)$ , the ground-state energy, the excitation energies, and the magnetization can now be calculated as functions of the dimerization parameter  $\delta$ . Previous calculations 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 equation (2) to the first order in  $\delta$ , implying that the results are valid only in the critical regime where  $\delta \rightarrow 0$ . The advantage of taking the variable nearest-neighbour exchange coupling is that the results will then be valid also in the limit where  $\delta \rightarrow 1$ .

The ground-state energies  $\varepsilon_g$  per site for undimerized chains,  $\delta = 0$ , were found, as expected, to be the same as those found earlier [6] for the three systems. After including dimerization, our calculations also confirm that the ground-state energy of all three systems described above decreases with  $\delta$ . This is shown in figure 2, where the 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  $\delta$ ,  $\delta^\nu / |\ln \delta|$ , for the three systems discussed here, with values of  $\nu$  between 1.4 and 1.6 over the entire range  $0 \leq \delta < 1$ . Figure 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  $k = 0$ , for 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 method found an almost linear  $\delta$ -dependence for these spin systems [5, 6]. We find that on including a variable nearest-neighbour coupling constant defined in equation (2), the spin-wave theory also allows for a  $\delta$ -dependent energy gap

$$\Delta(\delta) = E_2(\delta) - \varepsilon_g(\delta)$$



**Figure 2.** The magnetic energy gain  $\varepsilon_g(\delta) - \varepsilon_g(0)$  versus the dimerization parameter  $\delta$  for a 1D alternating-spin chain over the full range of the dimerization parameter,  $0 \leq \delta < 1$ .

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 behaviour,  $\delta^\nu / |\ln \delta|$ , as the ground-state energy, with  $\nu$  varying between 1.4 and 1.6 over the entire range  $0 \leq \delta < 1$ . This is true for all three spin systems defined here, and is larger for the  $(\frac{3}{2}, 1)$  system than the other two.

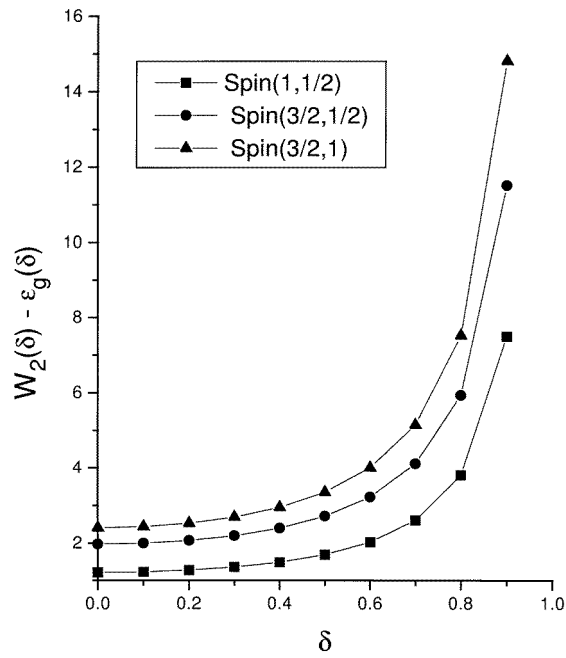
The staggered magnetization  $M(\delta)$  was also found to follow the logarithmic  $\delta$ -dependence up to  $\delta \leq 0.5$ , but follows a different behaviour for  $\delta > 0.5$ .  $M_1(\delta)$  is shown against dimerization in figure 4 for the three systems. Again we see, from figure 4, that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

It is worth mentioning here that by using the coupled-cluster method [13], we had found that the ground-state energy and the staggered magnetization of a spin-half Heisenberg chain follow the same logarithmic behaviour using the variable exchange coupling defined in equation (2) for both small and large values of the dimerization  $\delta$ . This gives us more confidence in the results that we have obtained from LSW theory.

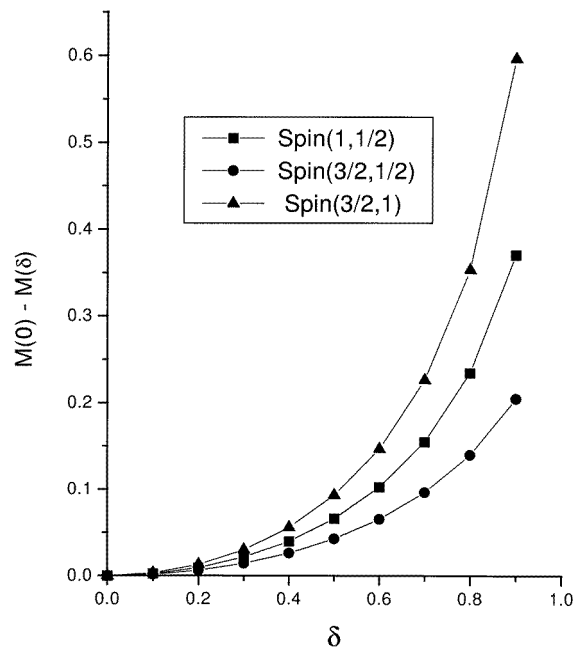
### 3. The two-dimensional alternating Hamiltonian

The dimerization on two-dimensional 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 [13].

We will use some of these configurations, illustrated in figure 5, to study the alternating-spin square lattices. The alternating dimerized Hamiltonian for a two-dimensional system can

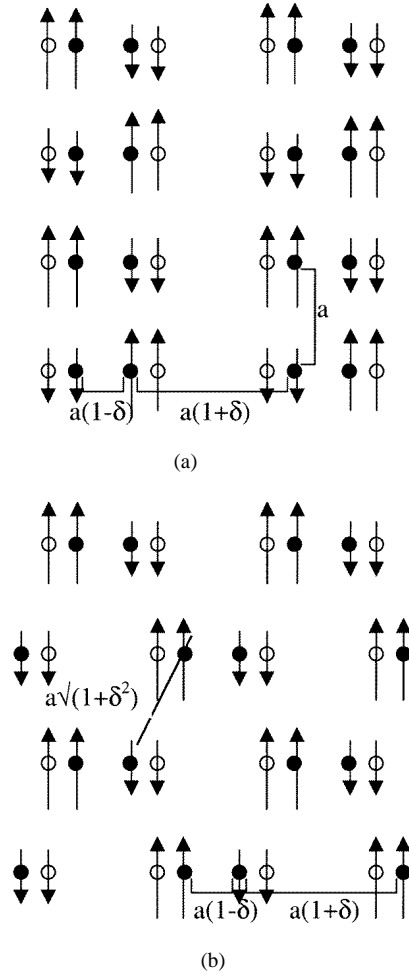


**Figure 3.** The  $\delta$ -dependence of the energy gap as dimerization sets in for different alternating-spin chains.



**Figure 4.** The  $\delta$ -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 the  $x$ -direction; like in (a), the dimerization occurs along one direction only, but the sequence of alternate couplings itself alternates along the other direction; (c) dimerization along both of the directions, caused by  $(\pi, 0)$  and  $(0, \pi)$  phonons, making a plaquette of four nearest-neighbour spins; (d) again, dimerization along both of the directions, but taken staggered along the vertical direction; (e) another staggered dimerization that is caused by a longitudinal  $(\pi, \pi)$  phonon. The large arrow belongs to the first sublattice and the short one belongs to second sublattice. Also, the open circles indicate the square-lattice sites and the solid ones show the dimerized lattice.

be written in general as

$$H = \sum_{i,j} \sum_{\mu=\pm 1}^{\sqrt{N}} [J_{x,\mu} \mathbf{S}_{1,i,j} \cdot \mathbf{S}_{2,i+\mu,j} + J_{y,\mu} \mathbf{S}_{1,i,j} \cdot \mathbf{S}_{2,i,j+\mu}] \quad (18)$$

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-neighbour exchange couplings. These are defined for different configurations as follows.

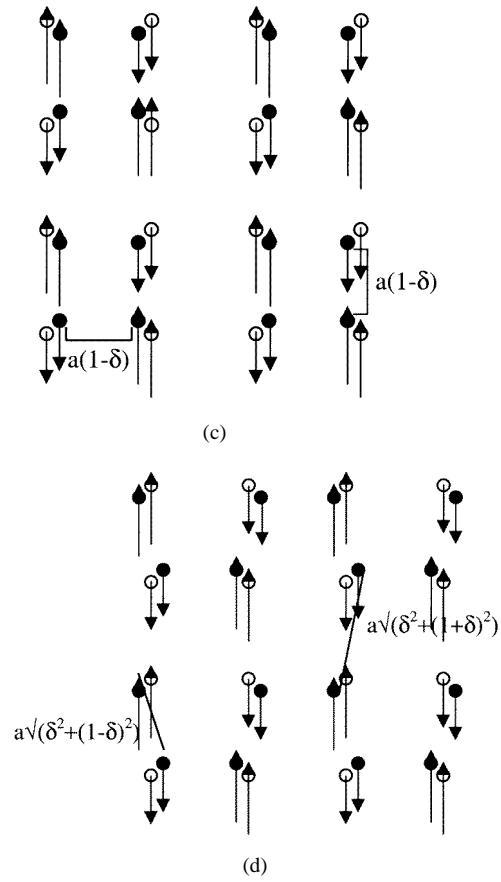


Figure 5. (Continued)

**Configuration (a):**

$$J_{x,\mu} = \frac{J}{(1 + \mu\delta)} \simeq J(1 - \mu\delta)$$

$$J_{y,\mu} = J.$$

**Configuration (b):**

$$J_{x,\mu} = \frac{J}{(1 + \mu\delta)} \simeq J(1 - \mu\delta)$$

$$J_{y,\mu} = \frac{J}{\sqrt{1 + \delta^2}} \simeq J\left(1 - \frac{\delta^2}{2}\right).$$

**Configuration (c):**

$$J_{x,\mu} = J_{y,\mu} = \frac{J}{(1 + \mu\delta)} \simeq J(1 - \mu\delta).$$

**Configuration (d):**

$$J_{x,\mu} = \frac{J}{(1 + \mu\delta)} \simeq J(1 - \mu\delta)$$

$$J_{y,\mu} = \frac{J}{\sqrt{\delta^2 + (1 + \mu\delta)^2}} \simeq J\left(1 - \mu\delta - \left(1 - \frac{\mu^2}{2}\right)\delta^2\right).$$

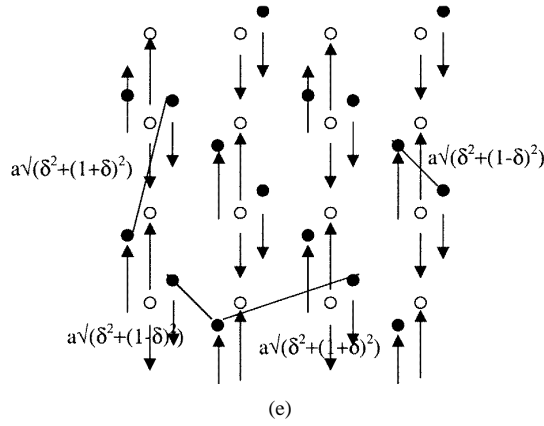


Figure 5. (Continued)

**Configuration (e):**

$$J_{x,\mu} = J_{y,\mu} = \frac{J}{\sqrt{\delta^2 + (1 + \mu\delta)^2}} \simeq J \left( 1 - \mu\delta - \left( 1 - \frac{\mu^2}{2} \right) \delta^2 \right).$$

We would like to investigate the five configurations resulting from dimerization of a square lattice in order to see

- which one of these leads to the largest gain in magnetic energy as the dimerization sets in,
- whether the use of variable exchange coupling leads to a single power-law behaviour valid for the entire range of  $\delta$ , and how the law differs from that in the case of chains,
- the  $\delta$ -dependence of the second mode of excitation  $E_2$ ,
- the behaviour of the staggered magnetization, and
- the generality of these investigations as regards 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 now have the following values:

$$A_1 = J_p s_2 \quad (19a)$$

$$A_2 = J_p s_1 \quad (19b)$$

$$B(k) = \Gamma(k) \sqrt{s_1 s_2} \quad (19c)$$

$$C = -J_p s_1 s_2 \quad (19d)$$

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

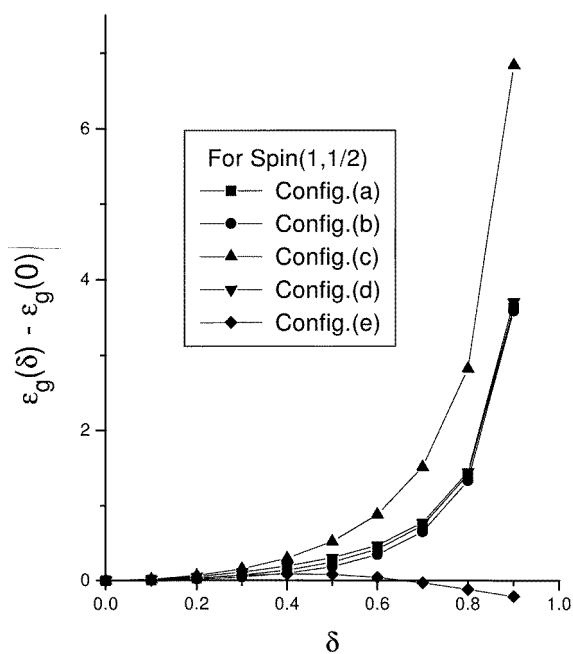
$$J_p = (J_{x,+1} + J_{x,-1} + J_{y,+1} + J_{y,-1})/4$$

$$J_{px} = (J_{x,+1} + J_{x,-1})/4$$

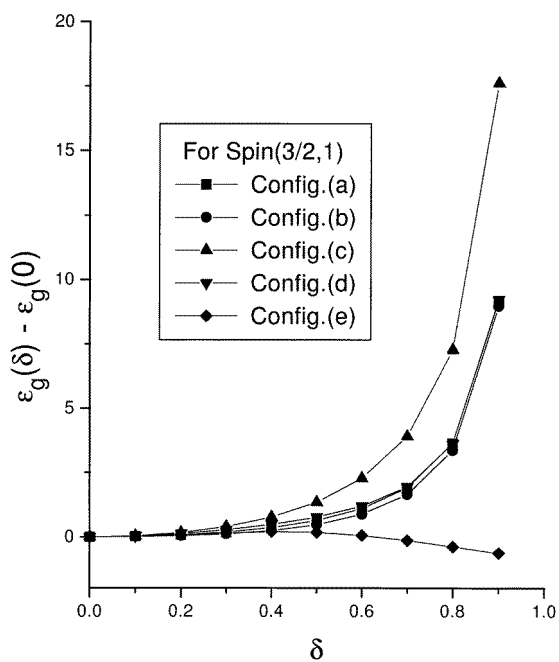
$$J_{py} = (J_{y,+1} + J_{y,-1})/4$$

$$J_{mx} = (J_{x,+1} - J_{x,-1})/4$$

$$J_{my} = (J_{y,+1} - J_{y,-1})/4.$$



(a)



(c)

**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 over the range  $0 \leq \delta < 1$  for (a) spin  $(1, \frac{1}{2})$ , (b) spin  $(\frac{3}{2}, \frac{1}{2})$ , and (c) spin  $(\frac{3}{2}, 1)$ .

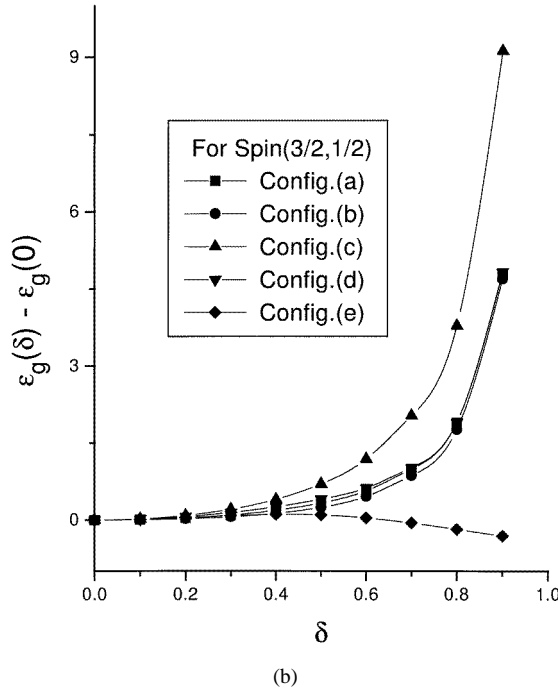


Figure 6. (Continued)

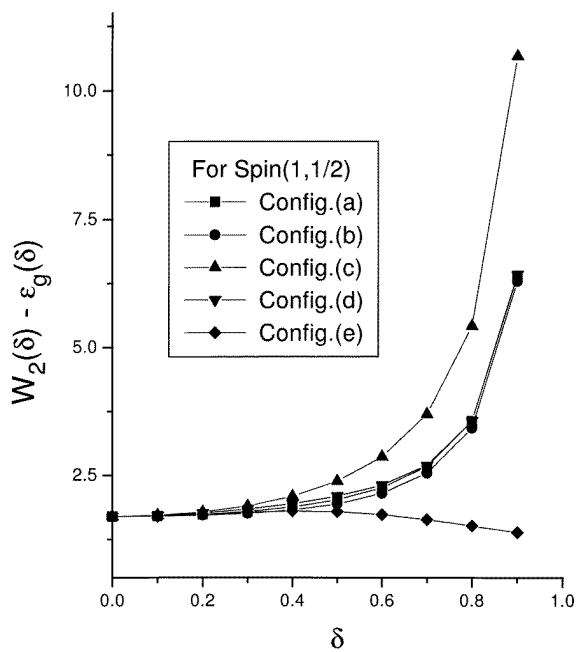
The ground-state energy  $\varepsilon_g(\delta)$  defined in equation (15), the energies of the two excitation modes  $E_i(k)$  in equation (14), and the staggered magnetization  $M(\delta)$  defined in equations (19) can now be calculated as functions of the dimerization parameter  $\delta$ .

**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.

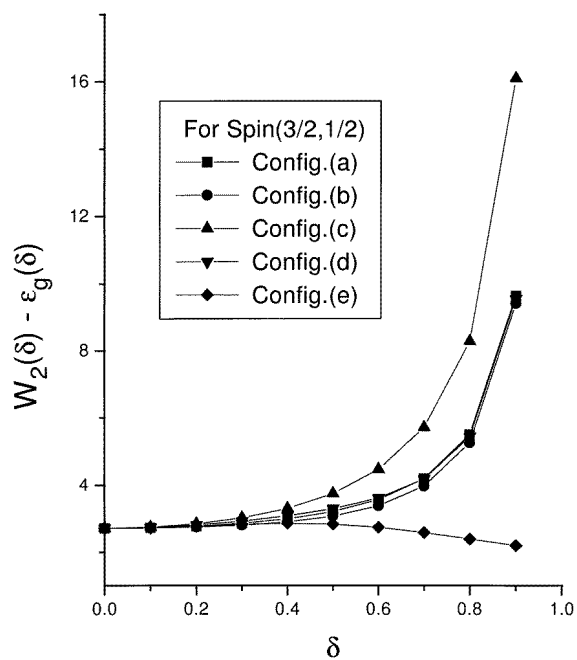
Spin system	$\varepsilon_g$	$M_1$	$M_2$
$(1, \frac{1}{2})$	-1.2	0.8907	-0.3907
$(\frac{3}{2}, \frac{1}{2})$	-1.7158	1.4241	-0.4241
$(\frac{3}{2}, 1)$	-3.3709	1.3597	-0.83597

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. The 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 for a chain, the gain in magnetic energy increases with  $\delta$  in all of 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 figure 5(c) is energetically the most favourable 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  $\delta^\nu / |\ln \delta|$  with  $\nu = 1.4$ – $1.6$  over the entire range  $0 \leq \delta < 1$ , exactly as in the case of



(a)



(b)

**Figure 7.** The 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  $(\frac{3}{2}, 1)$ .

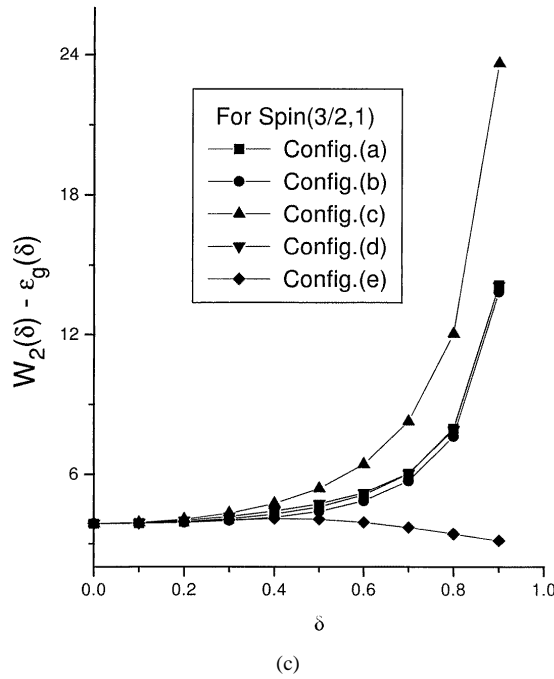


Figure 7. (Continued)

a chain. This is singularly an effect of taking the variable exchange coupling defined above.

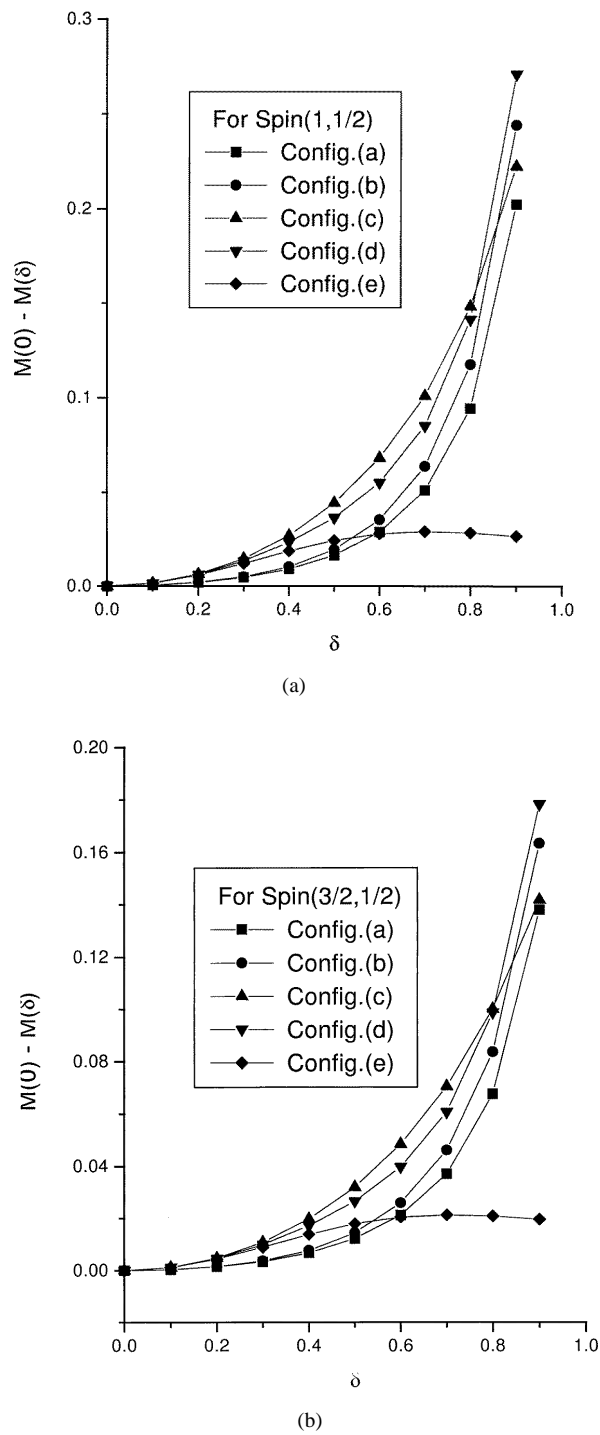
The  $\delta$ -dependence of the energy gap,  $\Delta(\delta) = E_2(\delta) - \epsilon_g(\delta)$ , for the five configurations is shown in figure 7, showing greater stabilization of the dimerized state with increasing  $\delta$ . We also find that, like the magnetic energy gain, the energy gap increases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

Our calculations give staggered magnetization for the undimerized alternating-spin square lattice  $M_1(\delta = 0) = 0.8907, 1.4241, \text{ and } 1.3597$  and  $M_2(\delta = 0) = -0.3907, -0.4241, \text{ and } -0.8597$  for  $(1, \frac{1}{2}), (\frac{3}{2}, \frac{1}{2}), \text{ and } (\frac{3}{2}, 1)$  respectively. As dimerization sets in, magnetization decreases in all of the configurations that we have chosen, as shown in figure 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  $1/\sqrt{2}$ . At this point the distortions give rise to a rectangular lattice with sides  $\sqrt{2}$  and  $1/\sqrt{2}$ . The energy gain increases with  $\delta$  up to  $\delta = \frac{1}{2}$ , and then decreases.

For all 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 like the energy gain and the energy gap. However, while for configurations (a)–(d) over the full range  $0 \leq \delta < 1$  the magnetization follows the same power law with the exponents  $\nu = 1.4$ – $1.6$ , configuration (e) shows a distinctly different behaviour in this regime.

In summary, we have studied the spin–Peierls dimerization of an alternating-spin



**Figure 8.** The  $\delta$ -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  $(\frac{3}{2}, 1)$ .



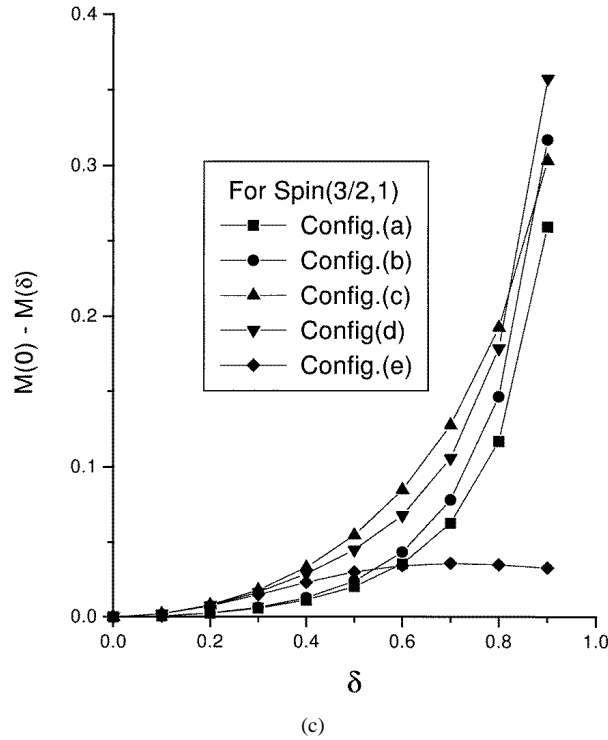


Figure 8. (Continued)

Heisenberg system on a chain and a square lattice taking variable exchange couplings based on the *ansatz*  $J(a) = J/a$ , for three kinds of alternating-spin system, namely  $(1, \frac{1}{2})$ ,  $(\frac{3}{2}, \frac{1}{2})$ , and  $(\frac{3}{2}, 1)$ . We have included different possibilities for dimerization in the case of a square lattice. The ground-state energy and the 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 favoured 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 of the configurations of the square lattice; and (v) over 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.

### Acknowledgment

We would like to thank Dr D Sen for providing references [5] and [6].

## References

- [1] van Koningsbruggen P J, Kahn O, Nakatani K, Pei Yu, Renard J P, Drillon M and Legoll P 1990 *Inorg. Chem* **29** 3325
- [2] Drillon M, Gianduzzo J C and Georges R 1983 *Phys. Lett.* **96A** 413  
Georges R, Curely J and Drillon M 1985 *J. Appl. Phys.* **58** 914
- [3] Kolezhuk A K, Mikeska H-J and Yamamoto S 1997 *Phys. Rev. B* **55** R3336
- [4] Brehmer S, Mikeska H-J and Yamamoto S 1997 *J. Phys.: Condens. Matter* **9** 3921
- [5] Pati S K, Ramasesha S and Sen D 1997 *Phys. Rev. B* **55** 8894
- [6] Pati S K, Ramasesha S and Sen D 1997 *J. Phys.: Condens. Matter* **9** 8707  
(Pati S K, Ramasesha S and Sen D 1997 *Preprint cond-mat/9704057*)
- [7] Blote H W J 1979 *J. Appl. Phys.* **50** 7401
- [8] Yamamoto S and Fukui T 1998 *Phys. Rev. B* **57** 14 008  
(Yamamoto S and Fukui T 1997 *Preprint cond-mat/9710291*)
- [9] Yamamoto S, Brehmer S and Mikeska H-J 1998 *Phys. Rev. B* **57** 13 610  
(Yamamoto S, Brehmer S and Mikeska H-J 1997 *Preprint cond-mat/9710332*)
- [10] Yamamoto S, Fukui T, Maisinger K and Schollwöck U 1998 *Preprint cond-mat/9806344*
- [11] Ivanov N B 1998 *Phys. Rev. B* **57** R14 024  
(Ivanov N B 1997 *Preprint cond-mat/9712014*)
- [12] Ivanov N B, Richter J and Schollwöck U 1998 *Phys. Rev. B* **58**  
(Ivanov N B, Richter J and Schollwöck U 1998 *Preprint cond-mat/9803150*)
- [13] Al-Omari A and Nayyar A H 1998 *J. Phys.: Condens. Matter* submitted
- [14] 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, Akhiezer A I, Bar'yakhtar V G and Peletminskii S V 1968 *Spin Waves* ed S Doniach (Amsterdam: North-Holland) 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* that we are using.
- [15] Kolezhuk A K and Mikeska H-J 1997 *Preprint cond-mat/9712022*
- [16] Koga A, Kumada S and Kawakami N 1998 *J. Phys. Soc. Japan* **67** 622  
(Koga A, Kumada S and Kawakami N 1997 *Preprint cond-mat/9711061*)