Self-consistent approximation for dimerization of ferrimagnets on chains and square lattices

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(Received 10 August 1998)

We have used nonlinear spin-wave theory to study dimerization of a Heisenberg system with alternating spins s_1 and s_2 on a linear chain and a square lattice for several possible dimerized configurations. It shows that the ground-state energy in both dimensions gets lowered against the dimerization for the three alternating spin systems. In two dimensions the plaquette configuration is found to be the most favorite one among the rest. An ansatz on variable nearest-neighbor exchange coupling gives rise to uniform power law $\delta^{\nu}/|\ln\delta|$ for the dependence of magnetic energy gain, energy gap, and magnetization for both the alternating chains as well as square lattices over the entire range of the dimerization parameter δ for the three spin systems. Our calculations using the unexpanded exchange coupling also allows the energy of the gapped excitation spectrum to be δ dependent. [S0163-1829(99)07709-7]

I. INTRODUCTION

Extensive theoretical as well as experimental work has been done over the years to study the behavior of pure regular magnetic chains. Yet another challenging topic has been opened up by the experimental discovery of ferrimagnetism in bimetallic chains, which are made up of two unequal sublattices.[1] This behavior was seen in several Mn(II)-nitronyl nitoxide derivatives and $Mn^{II}Cu^{II}$ bimetallic chains. The general formula for these chains is $ACu(pbaOH)(H_2O)_3 \cdot 2H_2O$, where *pba* OH is 2-hydroxy-1,3-propylenebis (oxamato) and A=Mn, Fe, Co, and Ni.[2] These chains consist of two sublattices with unequal spin magnitudes s_1 and s_2 with a net nonzero spin per unit cell. These are also referred to as alternating or mixed spin chains and are regarded as a Heisenberg system.[3–6]

A classical approach of solving an alternating quantum spin system was followed by Blöte by taking one of the two sublattices made of classical spins[7] in which he confirms the existence of two excitation modes, optical (gapped) and acoustic (gapless) modes. A computational paper to study the magnetic behavior of alternating spin-1/spin$\frac{1}{2}$ system on a finite closed chains was done, in which the influence of ratio between Landé factors was emphasized.[3]

Recently, Brehmer *et al.*[5] carried out an analytical study to calculate the ground-state properties and the low-lying excited state of an alternating spin-1/spin$\frac{1}{2}$ chain. They found by using spin-wave theory that there are two low-lying excitations, a ferromagnetic spin wave has a gapless excitation, and the antiferromagnetic spin wave is gapped. These results are in good agreement with the quantum Monte Carlo (QMC) method. Kolezhuk *et al.*[4] constructed matrix product states to study a Heisenberg alternating spin-1/spin$\frac{1}{2}$ chain.

A linear spin wave (LSW) analysis followed by a density-matrix renormalization group (DMRG) study[6] corresponding to the Cu-Ni bimetallic chain with the simple Heisenberg model with and without dimerization was carried out. Both methods predict the gapped and gapless excitations. The LSW theory showed that the energy gap at $k=0$ in the gapped mode does not depend on the dimer parameter δ, while the DMRG predicted an almost linear dependence.

The above studies confirm that these ferrimagnetic systems can be accurately described by a pure Heisenberg spin model. This confirmation has motivated us to investigate a dimerized alternating spin Heisenberg model by using a nonlinear spin-wave theory in which we expect to get better results. We would like also to extend our paper for a square lattice.

It is known that dimerization lowers the ground-state energy of a spin-half isotropic Heisenberg antiferromagnet.[8] In other words, the system stands to gain energy by such lattice deformations that render it dimerized with alternate weaker and stronger bonds between up and down spins on neighboring sites. Recently Pati *et al.*[6,9] studied the dimerization of chains with spins s_1 and s_2 ($s_1 > s_2$) on alternating sites using the Hamiltonian defined as

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

where the total number of sites (or bonds) is $2N$ and the sum is over the total number of unit cells N. δ is the dimerization parameter and is taken to vary between 0 and 1. They used the LSW theory and the DMRG study 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.

The laws proposed for the dependence of magnetic energy gain on δ in one-dimensional antiferromagnets have been of interest to many. In particular, the logarithmic δ dependence of various quantities like the gain in the magnetic ground-state energy, etc., $\varepsilon_g \sim \delta^{\nu}/|\ln\delta|$, was reported to be a result of corrections due to including umklapp processes.[10] It has, however, been shown recently[8] that the logarithmic dependence can also be a result of using for the nearest-neighbor spin-spin exchange coupling in the Hamiltonian (1) the ansatz[11]

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

instead of the often-used approximated form $J(1\pm\delta)$, which can be regarded as the approximated form of $J/(1\pm\delta)$. The form in Eq. (2) gives a logarithmic dependence over not just the $\delta\rightarrow 0$ regime, but over the entire range of δ from 0 to 1. In what follows, we shall use for exchange interaction the form in Eq. (2).

The same argument should apply to an alternating chain. 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 nonlinear spin wave (NLSW) theory in the Hartree-Fock approximation, which is known to give surprisingly good results for spin-half Heisenberg antiferromagnet. We can construct 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 $(\frac{3}{2},1)$. In Sec. II we will study these three alternating spin systems for a Heisenberg linear chain. The energy and magnetization of such systems will then be computed using the NLSW theory, and a comparison with earlier calculations by other methods will be made to gauge the validity of the NLSW method. Critical exponents of the dimer alternating chains will also be calculated. We shall also study alternating spin systems on a square lattice with several dimer configurations within the NLSW theory in Sec. III.

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 unexpanded form of the exchange coupling defined in Eq. (2) as

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

A nonlinear spin-wave analysis is usually performed with the help of either Holstein-Primakoff (HP) transformations or the Dyson-Maleev (DM) transformations of the spin operators to boson spin-deviation operators. We choose to use HP transformations since the DM transformations make the above Hamiltonian non-Hermitian. For the two sublattices the HP transformations are: for spin-s_1

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

$$S^-_{1,n}=a^\dagger_n(2s_1-a^\dagger_n a_n)^{1/2}, \quad (4b)$$

$$S^z_{1,n}=s_1-a^\dagger_n a_n, \quad (4c)$$

and for the second sublattice with spin-s_2

$$S^+_{2,n}=b^\dagger_n(2s_2-b^\dagger_n b_n)^{1/2}, \quad (5a)$$

$$S^-_{2,n}=(2s_2-b^\dagger_n b_n)^{1/2}b_n, \quad (5b)$$

$$S^z_{2,n}=b^\dagger_n b_n-s_2, \quad (5c)$$

where s_i is the magnitude of the spin on sublattice i.

A nonlinear spin-wave method was first applied by Oguchi[12] using $1/2s$ expansion of the HP transformation to study the two-dimensional (2D) and 3D antiferromagnetic Heisenberg models and obtained more accurate results for the ground-state energy than the linear spin-wave theory presented by Anderson.[13] Later it was shown that the $1/(2s)^2$ works well and gives a small correction to the problem.[14]

An NLSW analysis can be performed by expanding spin operators defined in Eqs. (4) and (5) up to quartic terms in the spin-deviation operators a and b and substituting them in the Hamiltonian of Eq. (3). Following Takahashi[15] by taking the average value of boson operators in the ground state of the Hamiltonian giving

$$\langle a_i a_i\rangle=\langle b^\dagger_j b^\dagger_j\rangle=0,\langle a^\dagger_i b_j\rangle=\langle a_i b^\dagger_j\rangle=0,$$

due to Bogoliubov transformation defied in Eqs. (10), the quartic terms can then be linearized using Hartree-Fock approximation as

$$a^\dagger_i a_i a_i b_j \simeq a^\dagger_i a_i\langle a_i b_j\rangle+\langle a^\dagger_i a_i\rangle a_i b_j-\langle a^\dagger_i a_i\rangle\langle a_i b_j\rangle, \quad (6a)$$

$$a^\dagger_i b^\dagger_j b^\dagger_j b_j \simeq a^\dagger_i b^\dagger_j\langle b^\dagger_j b_j\rangle+\langle a^\dagger_i b^\dagger_j\rangle b^\dagger_j b_j-\langle a^\dagger_i b^\dagger_j\rangle\langle b^\dagger_j b_j\rangle, \quad (6b)$$

$$a^\dagger_i a_i a^\dagger_i b^\dagger_j \simeq a^\dagger_i a_i\langle a^\dagger_i b^\dagger_j\rangle+\langle a^\dagger_i a_i\rangle a^\dagger_i b^\dagger_j-\langle a^\dagger_i a_i\rangle\langle a^\dagger_i b^\dagger_j\rangle, \quad (6c)$$

$$a_i b_j b^\dagger_j b_j \simeq a_i b_j\langle b^\dagger_j b_j\rangle+\langle a_i b_j\rangle b^\dagger_j b_j-\langle a_i b_j\rangle\langle b^\dagger_j b_j\rangle, \quad (6d)$$

$$2a^\dagger_i a_i b^\dagger_j b_j \simeq a^\dagger_i a_i\langle b^\dagger_j b_j\rangle+\langle a^\dagger_i a_i\rangle b^\dagger_j b_j-\langle a^\dagger_i a_i\rangle\langle b^\dagger_j b_j\rangle \quad (6e)$$

$$+a^\dagger_i b^\dagger_j\langle a_i b_j\rangle+\langle a^\dagger_i b^\dagger_j\rangle a_i b_j-\langle a^\dagger_i b^\dagger_j\rangle\langle a_i b_j\rangle. \quad (6f)$$

The linearized Hamiltonian in Fourier transformed variables is

$$H=\sum_k \left[A_1 a^\dagger_k a_k+A_2 b^\dagger_k b_k+B(k)(a^\dagger_k b^\dagger_k+b_k a_k)+C\right], \quad (7)$$

with

$$A_1=J_p\left[s_2-\frac{\langle O\rangle}{2}\sqrt{\frac{s_2}{s_1}}-\frac{\langle D\rangle}{2}\right], \quad (8a)$$

$$A_2=J_p\left[s_1-\frac{\langle O\rangle}{2}\sqrt{\frac{s_1}{s_2}}-\frac{\langle D\rangle}{2}\right], \quad (8b)$$

$$B(k)=\Lambda_k\left[\sqrt{s_1 s_2}-\langle D\rangle\frac{s_1+s_2}{4\sqrt{s_1 s_2}}-\frac{\langle O\rangle}{2}\right], \quad (8c)$$

$$C=J_p\left[-s_1 s_2+\langle D\rangle\langle O\rangle\frac{s_1+s_2}{2\sqrt{s_1 s_2}}+\frac{\langle D\rangle^2}{2}+\frac{\langle O\rangle^2}{2}\right]. \quad (8d)$$

Here,

$$\langle D\rangle\equiv\langle a^\dagger_i a_i\rangle=\langle b^\dagger_j b_j\rangle, \quad (9a)$$

$$\langle O\rangle\equiv\langle a^\dagger_i b^\dagger_j\rangle=\langle a_i b_j\rangle, \quad (9b)$$

and $\Lambda_k=\sqrt{[J_p\cos(k)]^2+[J_m\sin(k)]^2}$, $J_p=[J/(1-\delta)+J/(1+\delta)]/2$ and $J_m=[J/(1-\delta)-J/(1+\delta)]/2$. All the averages are taken in the ground state, which is the Neel state, at zero temperature.

The linearized Hamiltonian in Eq. (7), can be diagonalized using Bogoliubov transformations

$$a_k=u_k\alpha_k+v_k\beta_k^\dagger, \tag{10a}$$

$$b_k=u_k\beta_k+v_k\alpha_k^\dagger, \tag{10b}$$

to

$$\tilde{H}=\sum_k\ [\varepsilon_g+E_1(k)\alpha_k^\dagger\alpha_k+E_2(k)\beta_k^\dagger\beta_k], \tag{11}$$

where the coefficients u_k and v_k are constrained by the condition $u_k^2-v_k^2=1$, α_k and β_k are the normal mode boson operators, $E_1(k)$ and $E_2(k)$ are the energies of the two excitation modes, and ε_g is the ground-state energy per site.

The diagonal and off-diagonal averages of spin-deviation operators are determined from the following self-consistent equations

$$\langle D\rangle=\frac{1}{N}\sum_k\ v^2(k), \tag{12a}$$

$$\langle O\rangle=\frac{1}{N}\sum_k\ \gamma_k v(k)u(k), \tag{12b}$$

with γ_k, $u(k)$ and $v(k)$ defined as

$$\gamma_k=\frac{1}{z}\sum_d\ e^{ikd}, \tag{13a}$$

$$u(k)=\sqrt{\frac{A_1+A_2+\xi_k}{2\xi_k}}, \tag{13b}$$

$$v(k)=\sqrt{\frac{A_1+A_2-\xi_k}{2\xi_k}}, \tag{13c}$$

$$\xi_k=\sqrt{(A_1+A_2)^2-4B(k)^2}, \tag{13d}$$

where d is the nearest-neighbor distance. The two excitation modes are

$$E_1(k)=(A_1-A_2+\xi_k)/2, \tag{14a}$$

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

The self-consistent evaluation later shows that $A_2>A_1$, giving E_1 as the gapless mode and E_2 with a gap.

The ground-state energy per site ε_g is given by

$$\varepsilon_g=C-A_1-A_2+\sum_k\ \xi_k, \tag{15}$$

and the staggered magnetization in the two sublattices corresponding to the spins s_1 and s_2, respectively, is

$$M_1=S_1-\langle D\rangle, \tag{16a}$$

$$M_2=\langle D\rangle-S_2. \tag{16b}$$

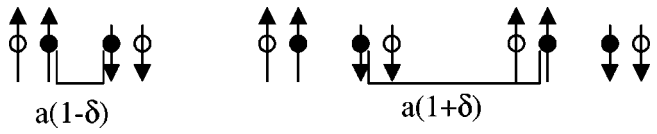

FIG. 1. A schematic sketch for 1D alternating spin chain. The large arrow indicates the large spin magnitude s_1 value and the small one indicates the small one s_2. While the hollow circles represent the undisturbed chain the filled circles represent the dimerized chain.

Excitation energies of both the modes increase upon dimerization, producing thereby a gap in the gapless mode also. $\Delta_i(\delta)=E_i(\delta)-\varepsilon_g(\delta)$ gives the δ dependence of the gap for the ith mode.

Setting all the quartic terms in Eq. (3) equal to zero, which amounts to putting $\langle D\rangle$ and $\langle O\rangle$ to zero, we recover the results of the linear spin-wave theory.[5,6]

Now we will investigate the three kinds of alternating spin chains referred to as $(1,\frac{1}{2})$,$(\frac{2}{3},\frac{1}{2})$, and $(\frac{2}{3},1)$. (See Fig. 1.) The ground-state energy, the energy gap, and magnetization can now be calculated as functions of the dimerization parameter δ. Previous calculations have invariably taken 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. (3) to order δ, implying that the results are valid only in the critical regime $\delta\to 0$. The advantage of taking the unexpanded form is that the results will then be valid also in the limit $\delta\to 1$. Our calculations confirm that the ground-state energy of all the three systems described above decreases with δ. This is shown in Fig. 2, where energy gain $\varepsilon(\delta)-\varepsilon(0)$ is plotted against δ.

The ground-state energies ε_g per site for undimerized chains $\delta=0$ were found to be $-0.725J$ for $(1,\frac{1}{2})$,

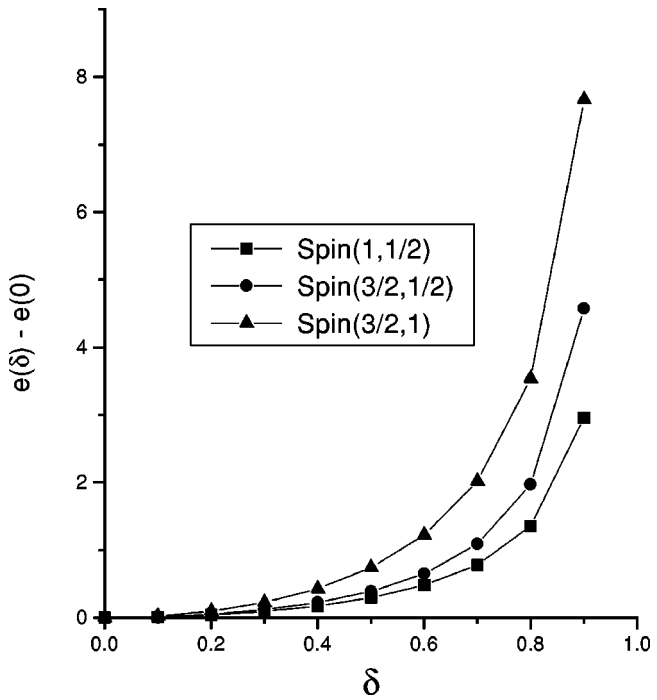

FIG. 2. The magnetic energy gain $\varepsilon(\delta)-\varepsilon(0)$ vs the dimer parameter δ for 1D alternating spin chain in the full dimer range $0\leqslant\delta<1$.

TABLE I. The physical quantities calculated by the different method 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	ε_o	M_1	M_2
$(1,\frac{1}{2})$	MPS (Ref. 4)	-0.7245	0.779	-0.279
	QMC (Refs. 4 and 5)	-0.7275	0.793	-0.293
	LSWT (Refs. 5 and 9)	-0.718	0.695	-0.195
	DMRG (Ref. 9)	-0.72709	0.79428	-0.29248
	NLSWT	-0.725	0.735	-0.235
$(\frac{3}{2},\frac{1}{2})$	LSWT (Ref. 9)	-0.979	1.315	-0.314
	DMRG (Ref. 9)	-0.98362	1.35742	-0.35742
	NLSWT	-0.9826	1.333	-0.333
$(\frac{3}{2},1)$	LSWT (Ref. 9)	-1.914	1.040	-0.540
	DMRG (Ref. 9)	-1.93096	1.14427	-0.644
	NLSWT	-1.924	1.081	-0.581

$-0.9826J$ for $(\frac{3}{2},\frac{1}{2})$, and $-1.924J$ for $(\frac{3}{2},1)$. While the linear spin-wave theory used by Pati *et al.*[9] gives a little higher ground-state energy comparing with DMRG and QMC, the nonlinear spin-wave theory used here gives very close values for all the three systems. Similarly, the sublattice magnetization of the undimerized chains is also in agreement with earlier calculations. The NLSW calculations show a small improvement over the LSW results. These values are listed in Table I. In the dimerization limit, the ground-state energy has a logarithmic behavior $\delta^{\nu}/|\ln\delta|$ of δ, with $\nu=1.4$–1.8 for the three spin systems. Figure 2 shows that while the energy gain increases with δ for the three systems, the chain $(\frac{3}{2},1)$ has a higher gain than the other two.

As expected, our calculations also find two branches of the excitation spectrum, one gapless and the other with a gap at $k=0$. We have found as in agreement with previous calculations that the energy excitation mode $E_1(k)$ defined in Eq. (14a) is gapless while there is a gap in the second mode of Eq. (14b) in the three systems. The LSW theory found no δ dependence of the energy gap in the second mode, while the DMRG found a linear dependence for $(1,\frac{1}{2})$.[6,9] If we include the NLSW calculation we found that the energy gap has a delta dependance. This dependance follows a power low if we take the coupling constant in the expanded form up to the first order of δ, but if we take the unexpanded form of the coupling constant we found that this dependence follows logarithmic law behavior $\delta^{\nu}/|\ln\delta|$, with ν varies between 1.4–1.8. This is true for the three spin systems defined here. Figure 3 shows the delta dependance of the gap energy $D(\delta)$. As we have seen in the energy gain, the chain with spin $(\frac{3}{2},1)$ gets the largest dependence on the dimer parameter.

For the dimerized lattices, the staggered magnetization $M(0)-M(\delta)$ gets lowered with the effect of dimerization. This is illustrated in Fig. 4. We found that the staggered magnetization has the same delta dependence as the energy ground state for $\delta\leqslant 0.5$ after it follows a different behavior.

It is worth it to mention here that critical exponents ν were evaluated for small values of dimerization; $\delta<0.1$ for

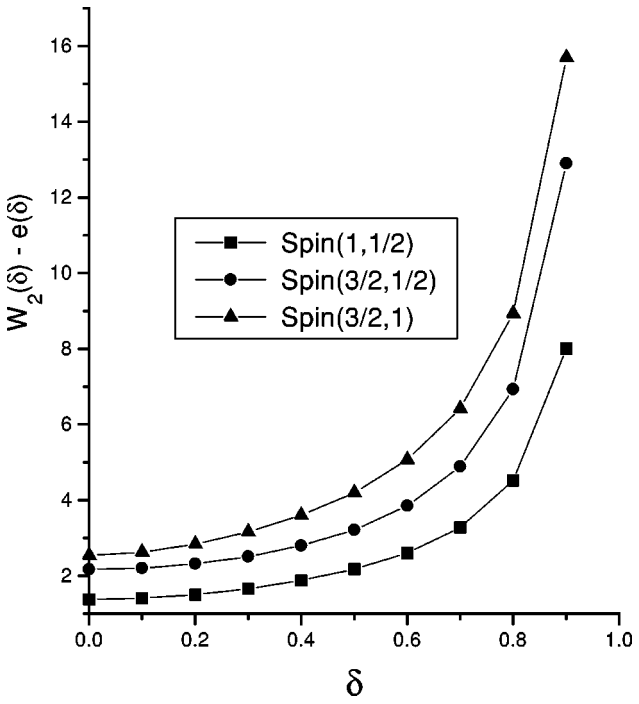

FIG. 3. The behavior of the energy-gap parameter D as dimerization sets on for a chain with different spins magnitudes.

the physical quantities described above using the same logarithmic law $\delta^{\nu}/|\ln\delta|$. We found that $\nu=1.5$ for the magnetic energy gain, the energy gap, and the sublattice magnetization for the three spin systems. The universality behavior of these physical quantities in the full dimerized range is of more interest to us.

III. TWO-DIMENSIONAL ALTERNATING HAMILTONIAN

A simple dimerization of a square lattice is interesting in its own right because the lattice distortions can take place in

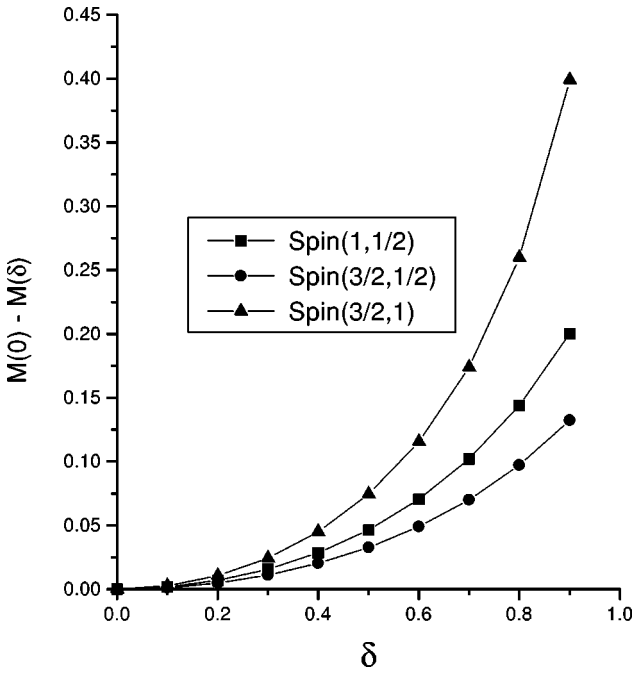

FIG. 4. Staggered imagination for one of the two sublattices; S_1, against δ in the full range, for the three chains.

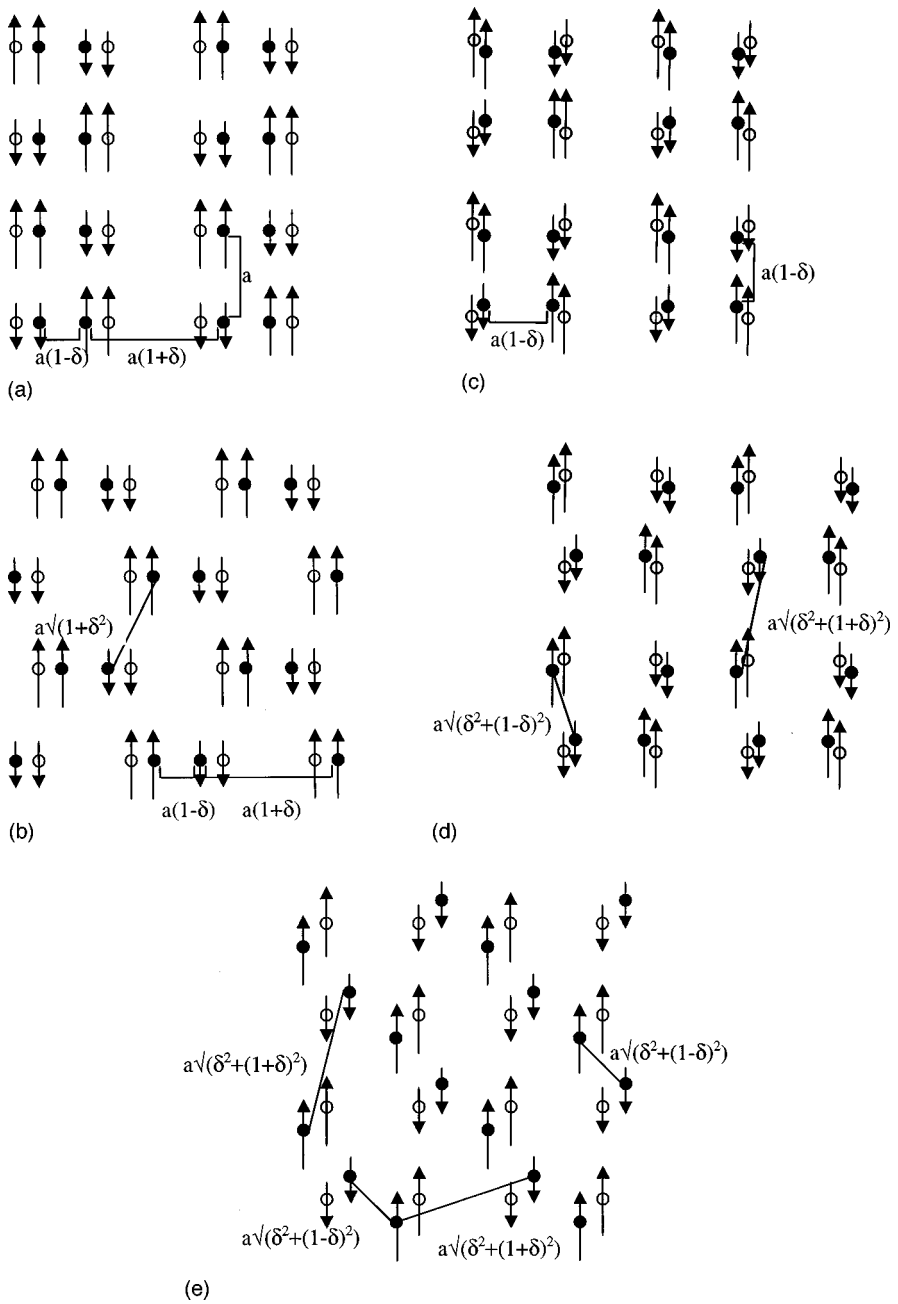

FIG. 5. Five configurations for the dimerization of a square lattice. (a) a columnar configuration caused by a longitudinal $(\pi,0)$ static phonon. (b) a staggered configuration caused by a (π,π) static phonon with polarization along the x direction. Like (a), the dimerization occurs along one direction 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-neighbor 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 (π,π) phonon. Large arrow belongs to the first sublattice while the short arrow belongs to second sublattice. And the open circles indicate the square lattice sites and the solid ones show the dimerized lattice.

more than one way, each one of the possible configurations giving a different dependence of the ground-state energy on the dimerization parameter. We have studied the dimerization of a spin-half Heisenberg antiferromagnet on a square lattice for configurations proposed earlier by us.[8] We will use these configurations, illustrated in Fig. 5, to study the alternating spin-square lattice. The alternating dimerized Hamiltonian for a two-dimensional system can be written in general as

$$H=\sum_{i,j}^{\sqrt{N}}\sum_{\sigma=\pm}\left[J_{x,\sigma}\mathbf{S}_{1,i,j}\cdot\mathbf{S}_{2,i+\sigma,j}+J_{y,\sigma}\mathbf{S}_{1,i,j}\cdot\mathbf{S}_{2,i,j+\sigma}\right], \tag{17}$$

with different coupling constants for each configuration defined as:

Configuration (a)

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

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

Configuration (b)

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

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

Configuration (c)

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

Configuration (d)

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

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

Configuration (e)

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

in which 1 denotes that this spin operators belong to the sublattice of spin s_1, and 2 belongs to sublattice of spin s_2 for the system defined in general as (s_1,s_2) and $\sigma=\pm 1$.

We would like to investigate the five configurations 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 untruncated exchange coupling leads to a single scaling law valid for the entire range of δ, and (iii) the behavior of staggered magnetization.

Now the same procedure that was used in solving linear chains will be used in treating square lattices. The HP transformation defined in Eqs. (4) and (5) can be substituted in the Hamiltonians of the five configurations. Using the Hartree-Fock approximation for the nonlinear terms of the Hamiltonian as defined in Eqs. (6). Putting these equations in the Hamiltonian defined in Eq. (17) and after collecting terms, we can use Bogoliubov transformations defined in Eqs. (10) to diagonalize the Hamiltonian. Having done that, the self consistent diagonal and off-diagonal averages of spin deviation operators defined in Eqs. (14) can be computed with

$$A_1=J_p\left[s_2-\frac{\langle O\rangle}{2}\sqrt{\frac{s_2}{s_1}}-\frac{\langle D\rangle}{2}\right], \tag{18a}$$

$$A_2=J_p\left[s_1-\frac{\langle O\rangle}{2}\sqrt{\frac{s_1}{s_2}}-\frac{\langle D\rangle}{2}\right], \tag{18b}$$

$$B(k)=\Gamma(k)\left[\sqrt{s_1s_2}-\langle D\rangle\frac{s_1+s_2}{4\sqrt{s_1s_2}}-\frac{\langle O\rangle}{2}\right], \tag{18c}$$

$$C=J_p\left[-s_1s_2+\langle D\rangle\langle O\rangle\frac{s_1+s_2}{2\sqrt{s_1s_2}}+\frac{\langle D\rangle^2}{2}+\frac{\langle O\rangle^2}{2}\right], \tag{18d}$$

$$\Gamma(k)=\sqrt{[J_{px}\cos(k_x)+J_{py}\cos(k_y)]^2+[J_{mx}\sin(k_x)+J_{my}\sin(k_y)]^2}, \tag{18e}$$

where $J_p=(J_{x,+1}+J_{x,-1}+J_{y,+1}+J_{y,-1})/4$, $J_{px}=(J_{x,+1}+J_{x,-1})/4$, $J_{py}=(J_{y,+1}+J_{y,-1})/4$, $J_{mx}=(J_{x,+1}-J_{x,-1})/4$, and $J_{my}=(J_{y,+1}-J_{y,-1})/4$. After this has been done, the gain in magnetic energy $\varepsilon(\delta)-\varepsilon(0)$, the gap parameter $D(\delta)$, and staggered magnetization $M(0)-M(\delta)$ defined in Eqs. (18) can now be calculated as functions of the dimerization parameter δ.

The ground-state energy $\varepsilon(\delta=0)$ is found to be $-1.204J$, $-1.7179J$, and $-3.3758J$ for the three spin systems $(1,\frac{1}{2})$, $(\frac{3}{2},\frac{1}{2})$, and $(\frac{3}{2},1)$, respectively. These values are listed in Table II.

Our calculations confirm that, like the chain, the ground-state energy of the five configurations decreases with δ. This is shown in Fig. 6, where the energy gain $\varepsilon(\delta)-\varepsilon(0)$ is plotted against δ for the proposed configurations. What is significant is that the ground-state energy goes down with δ more rapidly for some configurations than others. In fact, Fig. 6 shows that the δ dependence is markedly different for the types of dimerized configurations. It also shows that the plaquette configuration of Fig. 5(c) is energetically the most favorable state. While the plaquette configuration stands out as the most preferred one, there is hardly a discernible difference among the configurations (a), (b), and (d).

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

TABLE II. The ground-state energy and the staggered magnetization of the undimerized square alternating lattice for the three spin systems as calculated by nonlinear spin-wave theory.

Spin system	ε_o	M_1	M_2
$(1,\frac{1}{2})$	-1.204	0.895	-0.395
$(\frac{3}{2},\frac{1}{2})$	-1.7179	1.427	-0.427
$(\frac{3}{2},1)$	-3.3758	1.362	-0.862

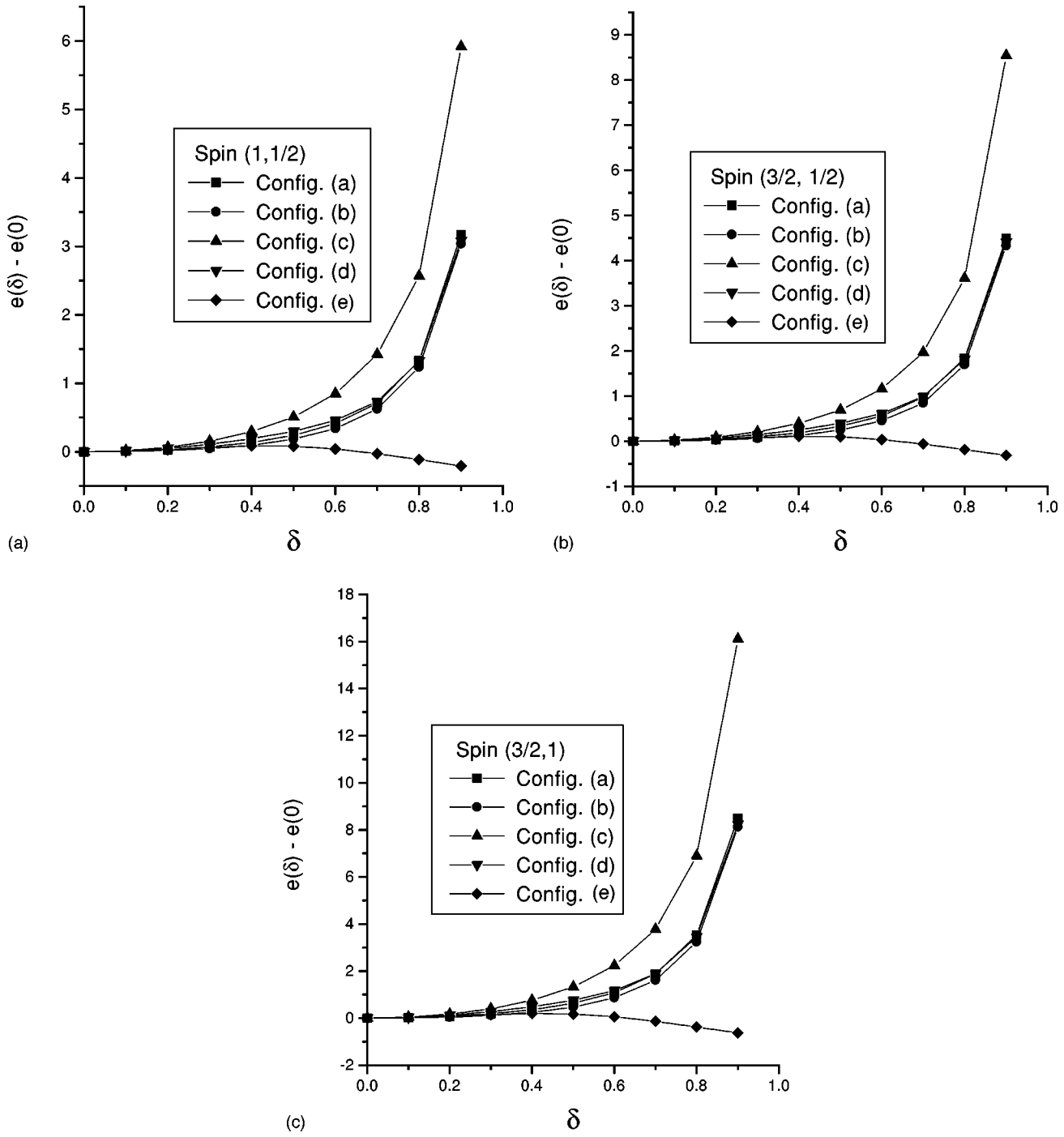

FIG. 6. The gain in magnetic energy $\varepsilon(\delta)-\varepsilon(0)$ as dimerization sets in with increasing δ for the five configurations of the square lattices in the range $0 \leqslant \delta < 1$ for (a) spin $(1,\frac{1}{2})$, (b) spin $(\frac{3}{2},\frac{1}{2})$, and (c) spin $(\frac{3}{2},1)$.

point the distortions give rise to a rectangular lattice with sides $\sqrt{2}$ and $1/\sqrt{2}$. The energy gain increases with δ up to $\delta=\frac{1}{2}$, and then goes down.

It is interesting to note that as we found for the chains, the dimerization of a square alternating spin lattice also varies as $\delta^{\nu}/|\ln\delta|$, as summarized with $\nu=1.4$–1.8. There, the factor of $1|\ln\delta|$ is again due to the use of unexpanded exchange coupling defined in the Hamiltonian for the five configurations.

The δ dependence of the energy gap $D(\delta)$ defined above for the five configurations is shown in Fig. 7, showing greater stabilization of the dimerized state with increasing δ. We also find that, like the magnetic energy gain, the gap energy D increases with δ as $\delta^{\nu}/|\ln\delta|$ in the small δ regime for all the five configurations with $\nu=1.4$–1.8. The configurations (a)–(d) also have the same dependence on δ in the entire range of δ with $\nu=1.4$–1.8.

The difference between the dimerization of a square lattice for these configurations is again markedly brought out in Fig. 7. Also, the plaquette configurations (e) again appear as preferred modes of dimerization over the rest of configurations for having higher values of the energy gap.

Our calculations using the NLSW theory give staggered magnetization for the undimerized square alternating spin

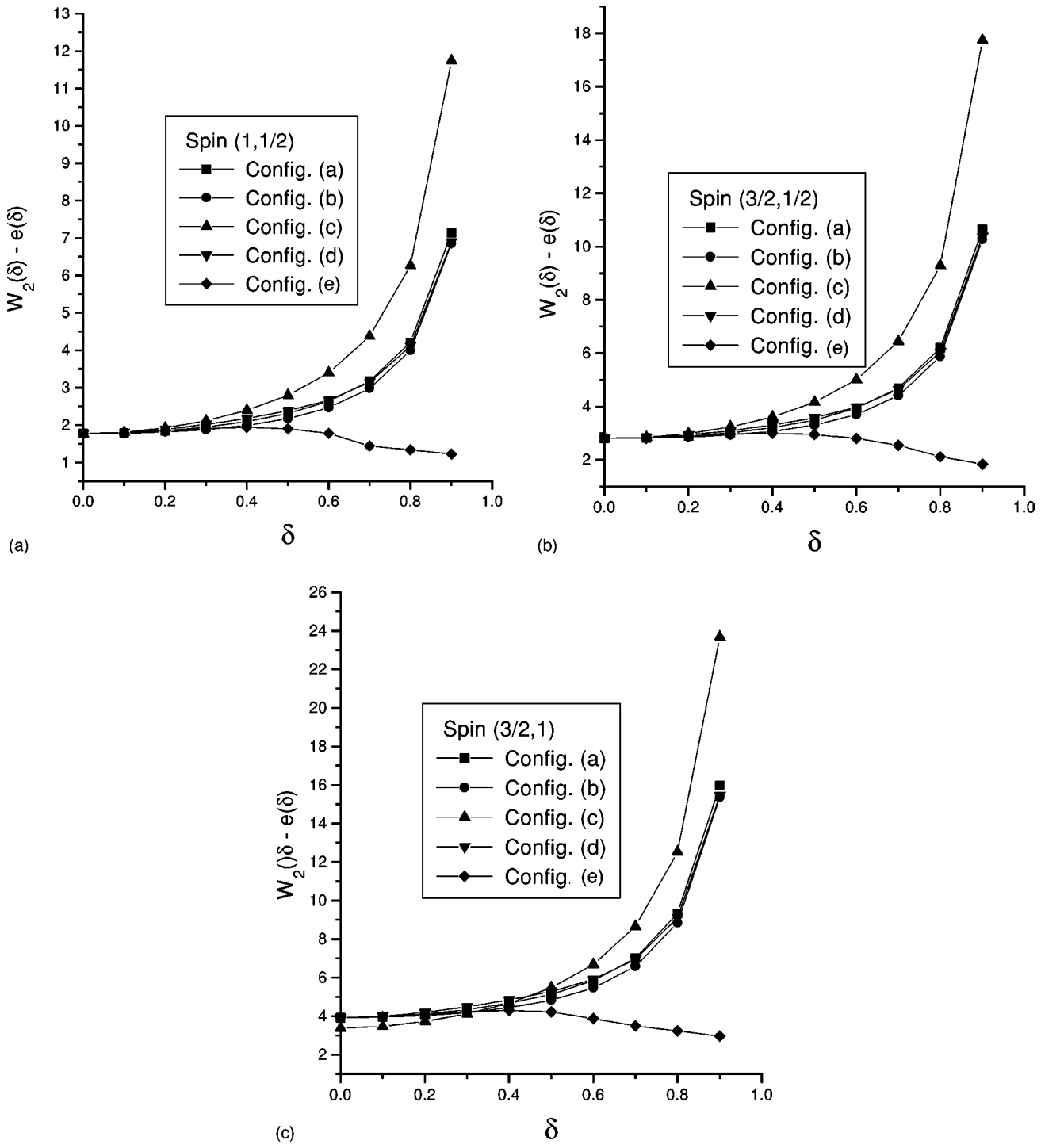

FIG. 7. Dependence of the energy-gap parameter D on δ for the five dimerization configurations of the alternating square lattices in the full range of dimer parameter δ for (a) spin $(1,\frac{1}{2})$, (b) spin $(\frac{3}{2},\frac{1}{2})$, and (c) spin $(\frac{3}{2},1)$.

lattice $M_1(\delta=0)=0.895$, 1.427, and 1.362 and $M_2(\delta=0)=-0.395$, -0.4427, and -0.862 for $(1,\frac{1}{2})$, $(\frac{3}{2},\frac{1}{2})$, and $(\frac{3}{2},1)$, respectively, as shown in Table II.

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 of δ $(0\leqslant\delta<1)$, except in the case of configuration (e), for which the magnetization rises again after $\delta=\frac{1}{2}$.

For all the five configurations, we found that the magnetization also varies as $\delta^{\upsilon}/|\ln\delta|$ in the small δ regime with the exponent $\nu=1.4$–1.8, exactly as the energy gain and the energy gap. However, in the far critical regime $(0\leqslant\delta<1)$ the magnetization goes with the exponents as $\nu=1.4$–1.8. Configuration (e) has a distinctly different behavior in this regime.

In summary, we have studied the spin-Peierls dimeriza-

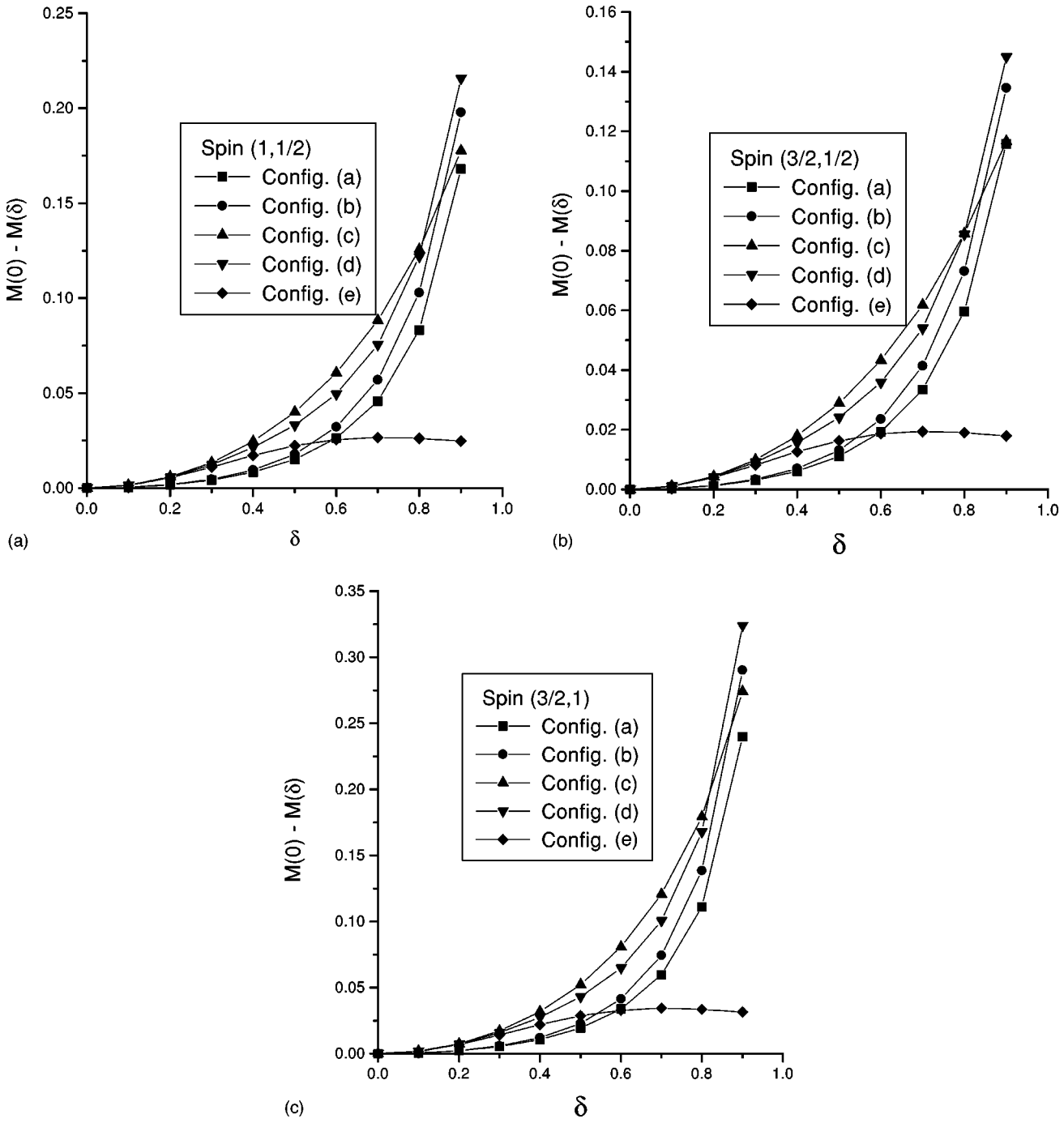

FIG. 8. Staggered magnetization for a 2D alternating lattice varying with δ calculated for the five dimerization configurations. The dimerization is taken in the full range for (a) spin $(1,\frac{1}{2})$, (b) spin $(\frac{3}{2},\frac{1}{2})$, and (c) spin $(\frac{3}{2},1)$.

tion of an alternating spin Heisenberg system on a chain and a square lattice taking unapproximated exchange couplings based on the ansatz $J(a)=J/a$, for three kinds of alternating spins, 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 2D. The ground-state energy as well as staggered magnetization, decrease continuously with increasing dimerization for one and two dimensions. In two dimensions, 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 parameter also corroborates the above conclusions. It has also been shown that the magnetic energy gain as well as the gap parameter and staggered magnetization depend upon the dimerization parameter δ as $\delta^{\nu}/|\ln\delta|$ in both regimes of critical δ, i.e., $\delta\rightarrow 0$, and full length of dimerization, i.e., $\delta\rightarrow 1$. The $|\ln\delta|$factor coming in without any considerations of umklapp processes being included.

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[11] 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-neighbor model that we are considering, the exchange integral is taken to be appreciable only over the nearest-neighbor distance a, in which case J is proportional to $1/a$ [see, for example, A. I. Akhiezer, V. G. Bar'yakhtar, and S. V. Peletminskii, in *Spin Waves*, translation edited by S. Doniach (North Holland, Amsterdam, 1968), p. 9]. It thus seems reasonable to assume that if the nearest-neighbor distance a changes to a' due to dimerization, then J is proportional to $1/a'$. This is the ansatz we are using.

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