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The combined effect of frustration and dimerization in ferrimagnetic chains and square lattices

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Abstract. Within the zero-temperature linear spin-wave theory we have investigated the effect of frustration and dimerization of a Heisenberg system with alternating spins s_1 and s_2 on one- and two-dimensional lattices. The combined effect appears most visibly in the elementary excitation spectra. In contrast to the ground-state energy that decreases with dimerization and increases with frustration, the excitation energies are shown to be suppressed in energy by both dimerization and frustration. The excitation modes also exhibit softening beyond a threshold value of frustration, signalling a transition from a classical ferrimagnetic state to a spiral state. The threshold value of frustration in a chain decreases with dimerization, showing that dimerization further assists in the phase transition. That the long-range classical ferrimagnetic order is destroyed is shown by the correlation length as well as sublattice magnetization decreasing with both dimerization and frustration. These effects have also been studied for a square lattice taking the dimerization interaction as $J/(1 \mp \delta)$ rather than $J(1 \pm \delta)$ where the linear spin-wave theory shows that dimerization initially opposes the frustration-led transition to a spiral magnetic state, but then higher magnitudes of lattice deformation facilitate the transition.

(Some figures in this article are in colour only in the electronic version; see www.iop.org)

1. Introduction

Ferrimagnetic spin systems consisting of two sublattices with spins s_1 and s_2 of unequal magnitudes, with a net non-zero spin per unit cell, have recently been a focus of considerable attention. Referred to also as alternating- or mixed-spin systems, these are regarded as Heisenberg systems. Several theoretical studies have been carried out to calculate the ground-state properties and the low-lying excited states of an alternating s_1 – s_2 chain [1–12]. There are two low-lying elementary excitations: the gapless ferromagnetic spin-wave excitation and the antiferromagnetic spin wave with a gap. The ground-state energy, sublattice magnetization, and excitation energies of a Heisenberg ferrimagnetic system are lowered in dimerization[†].

Frustration due to competing antiferromagnetic second-neighbour interactions can in principle destroy any LRO of the Néel type. Not much is, however, known about the effect of frustration on ferrimagnetic systems. Recently Ivanov *et al* [6] used spin-wave expansion, the density matrix renormalization group, and an exact-diagonalization technique to investigate the effect of weak frustration on the ground-state energy of a Heisenberg ferrimagnetic chain. They have identified several critical frustration parameters. The first, called J_c , heralds a transition from the classical commensurate ferrimagnetic state to a spiral state. The second special point,

[†] For more details see reference [5] and the references therein.

called J_D and termed the disorder point, marks the onset of incommensurate finite-range spin-spin correlations. The third special point, called J_T , is a first-order transition point from the long-range-ordered ferrimagnetic state with total spin $S_g = N(s_1 - s_2)$ to a singlet state with $S_g = 0$. They found that frustration causes an increase in the ground-state energy.

In this paper we will study alternating-spin systems with (s_1, s_2) equal to $(1, \frac{1}{2})$, $(\frac{3}{2}, 1)$, and $(\frac{3}{2}, \frac{1}{2})$ using a zero-temperature linear spin-wave (LSW) theory[†]. The choice is guided by the recent assertion that the three systems have different predominant characters: the first has a mixed ferromagnetic and antiferromagnetic character, the second is more antiferromagnetic, and the third is more ferromagnetic in character [10–12]. It has already been argued by Ivanov *et al* that the LSW theory yields satisfactory results, at least for small values of frustration [6]. Section 2 below sets up the LSW formalism for a dimerized ferrimagnetic chain in the presence of frustration. Effects of frustration on a dimerized square-lattice system are then studied in section 3.

2. Ferrimagnetic chains with dimerization and frustration

Mixed-spin-chain systems have recently been studied extensively within the spin-wave approximation in both undimerized [1, 4, 6, 9, 10] and dimerized [3, 5, 8, 11, 12] regimes. We consider a chain consisting of two sublattices occupied by spins s_1 and s_2 ($s_1 > s_2$) allowing for both intersublattice and intrasublattice nearest-neighbour interactions J_1 and J_2 respectively. We choose to describe this system by the Hamiltonian

$$H = \sum_n [J^+ \mathbf{S}_{1,n} \cdot \mathbf{S}_{2,n} + J^- \mathbf{S}_{2,n} \cdot \mathbf{S}_{1,n+1} + J_2 (\mathbf{S}_{1,n} \cdot \mathbf{S}_{1,n+1} + \mathbf{S}_{2,n} \cdot \mathbf{S}_{2,n+1})] \quad (1)$$

where $J^\pm = J_1(1 \pm \delta)$, and δ is the dimerization parameter that varies from 0 to 1. The total number of sites (or bonds) is $2N$ and the sum is over the N unit cells.

The usual boson representation of spin operators in the two sublattices is

$$\begin{aligned} S_{1,n}^+ &= (2s_1 - a_n^\dagger a_n)^{1/2} a_n & S_{2,n}^+ &= b_n^\dagger (2s_2 - b_n^\dagger b_n)^{1/2} \\ S_{1,n}^z &= s_1 - a_n^\dagger a_n & S_{2,n}^z &= b_n^\dagger b_n - s_2. \end{aligned} \quad (2)$$

In terms of the normal-mode operators:

$$\alpha_k = u_k a_k - v_k b_k^\dagger \quad (3a)$$

$$\beta_k = u_k b_k - v_k a_k^\dagger \quad (3b)$$

the linearized Hamiltonian in equation (1) becomes

$$\tilde{H} = \varepsilon_g + \sum_k [E_1(k) \alpha_k^\dagger \alpha_k + E_2(k) \beta_k^\dagger \beta_k]. \quad (4)$$

The ground-state energy per unit cell ε_g and the energies of the two excitation modes $E_1(k)$ and $E_2(k)$ are given by

$$\varepsilon_g = C - \sum_k [A_1(k) + A_2(k) - \xi(k)] \quad (5)$$

$$E_1(k) = \frac{1}{2} (A_1(k) - A_2(k) + \xi(k)) \quad (6)$$

$$E_2(k) = \frac{1}{2} (A_2(k) - A_1(k) + \xi(k)). \quad (7)$$

[†] It was assumed [3, 10] as well as shown explicitly [5] earlier that the behaviour of an alternating-spin chain remains similar regardless of the values of s_1 and s_2 .

In these equations,

$$\xi_k = \sqrt{(A_1(k) + A_2(k))^2 - 4B^2(k)} \quad (8a)$$

$$A_1(k) = J_p s_2 - \alpha s_1 [1 - \cos(2k)] \quad (8b)$$

$$A_2(k) = J_p s_1 - \alpha s_2 [1 - \cos(2k)] \quad (8c)$$

$$B(k) = \sqrt{s_1 s_2} \Lambda_k \quad (8d)$$

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

$$C = -J_p s_1 s_2 + \frac{\alpha}{2}(s_1^2 + s_2^2) \quad (8f)$$

where $J_p = \frac{1}{2}(J^+ + J^-)$ and $\alpha = J_2/J_1$ is the frustration parameter.

The coefficients $u(k)$ and $v(k)$, constrained by the condition $u^2(k) - v^2(k) = 1$, are given by

$$u(k) = \sqrt{\frac{A_1(k) + A_2(k) + \xi(k)}{2\xi(k)}} \quad (9a)$$

$$v(k) = \sqrt{\frac{A_1(k) + A_2(k) - \xi(k)}{2\xi(k)}}. \quad (9i)$$

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

$$M_1 = S_1 - \langle D \rangle \quad (10a)$$

$$M_2 = \langle D \rangle - S_2 \quad (10b)$$

where $\langle D \rangle = \langle a_k^\dagger a_k \rangle = \langle b_k^\dagger b_k \rangle$ is the average taken in the ground state, which is the Néel-like state at zero temperature:

$$\langle D \rangle = \frac{1}{N} \sum_k v^2(k) \quad (11)$$

with k running from $-\pi/2$ to $\pi/2$ which is the first reduced Brillouin zone.

For a two-spin system, we can think of three types of spin-spin correlation function: $\langle S_{1,0}^z S_{1,n}^z \rangle$, $\langle S_{2,0}^z S_{2,n}^z \rangle$, and $\langle S_{1,0}^z S_{2,n}^z \rangle$. We are interested in the antiferromagnetic correlations which we define as

$$C_n \equiv \langle S_{1,0}^z S_{2,n}^z \rangle - \langle S_{1,0}^z \rangle \langle S_{2,n}^z \rangle \quad (12)$$

$$= -\langle O \rangle^2 \quad (13)$$

where

$$\langle O \rangle = \frac{1}{N} \sum_k \cos(kn) u(k) v(k) \quad (14)$$

and u and v are defined in equations (9). Pati *et al* [3] in their linear spin-wave analysis when fitting this correlation function to $e^{-r/\xi}$ found the inverse correlation length $\xi^{-1} = \ln(s_1/s_2)$. For $(s_1, s_2) = (1, \frac{1}{2})$, this gives $\xi = 1.44$, whereas their variational calculation gives $\xi = 0.75$. Others [6] fitted it to the Ornstein-Zernike form

$$C(r) \sim \frac{e^{-r/\xi}}{\sqrt{r}} \quad (15)$$

and found it to be 1.01.

Results of the spin-wave theory of ferrimagnets have been discussed earlier [1,3–6,8–12], but none with dimerization and frustration together.

There is a critical value of the frustration parameter α in the linear spin-wave theory at which the energies do not remain real, signalling destruction of the long-range order. This critical value, that we call α_c , is strongly δ -dependent, as shown in figure 1. At $\delta = 0$, $\alpha_c = s_1/[2(s_1 + s_2)]$. For a $(1, \frac{1}{2})$ chain this is $1/3$, whereas earlier DMRG results [6] gave $\alpha_c = 0.28$.

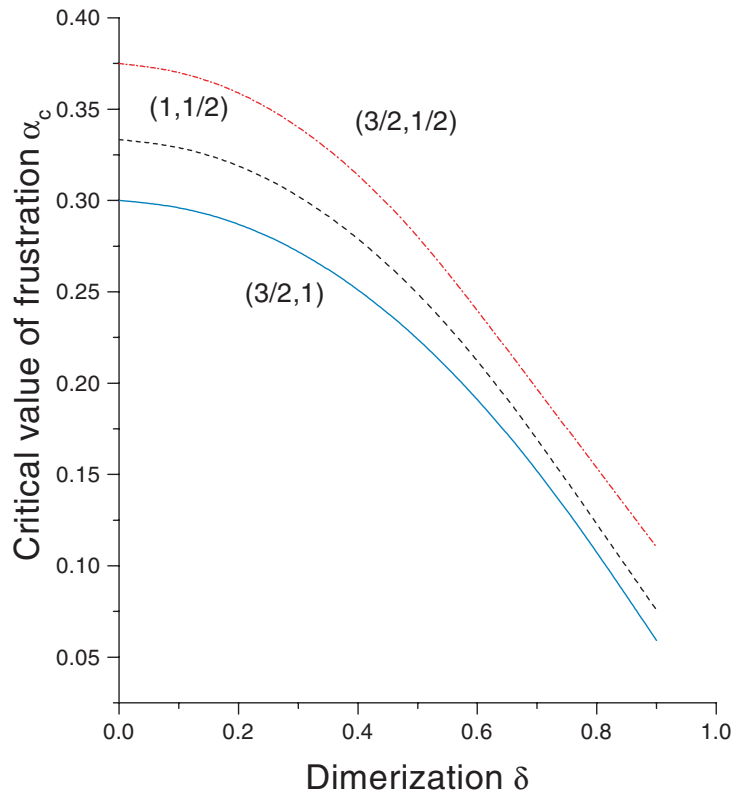


Figure 1. The dependence of the critical frustration parameter α_c on the dimerization parameter δ for the one-dimensional spin systems $(1, \frac{1}{2})$, $(\frac{3}{2}, 1)$, and $(\frac{3}{2}, \frac{1}{2})$. This is for the case where the dimerization dependence of the nearest-neighbour interaction is taken as $J^\pm = J_1(1 \pm \delta)$.

It is already known that with $J^\pm = J_1(1 \pm \delta)$ the ground-state energy decreases with δ and scales as δ^2 [3, 13]. It however has a more interesting behaviour with respect to α . As shown in figure 2, the ground-state energy per site initially increases with α and then decreases before the long-range order is destroyed by frustration. This is true even when there is no dimerization, where the results agree with those of Ivanov *et al* [6] who give values only up to where the maximum occurs. The maximum shifts to lower values of α with δ as δ^2 . The curves in figure 2 terminate at α_c for the corresponding δ . This behaviour is correct for all three spin systems considered.

With two atoms per unit cell, a ferrimagnet has to have two modes of elementary excitations. The acoustic mode is gapless ($E_1(k=0) = 0$) and has ferromagnetic character while the optic mode $E_2(k)$ is antiferromagnetic and has a gap at $k = 0$.

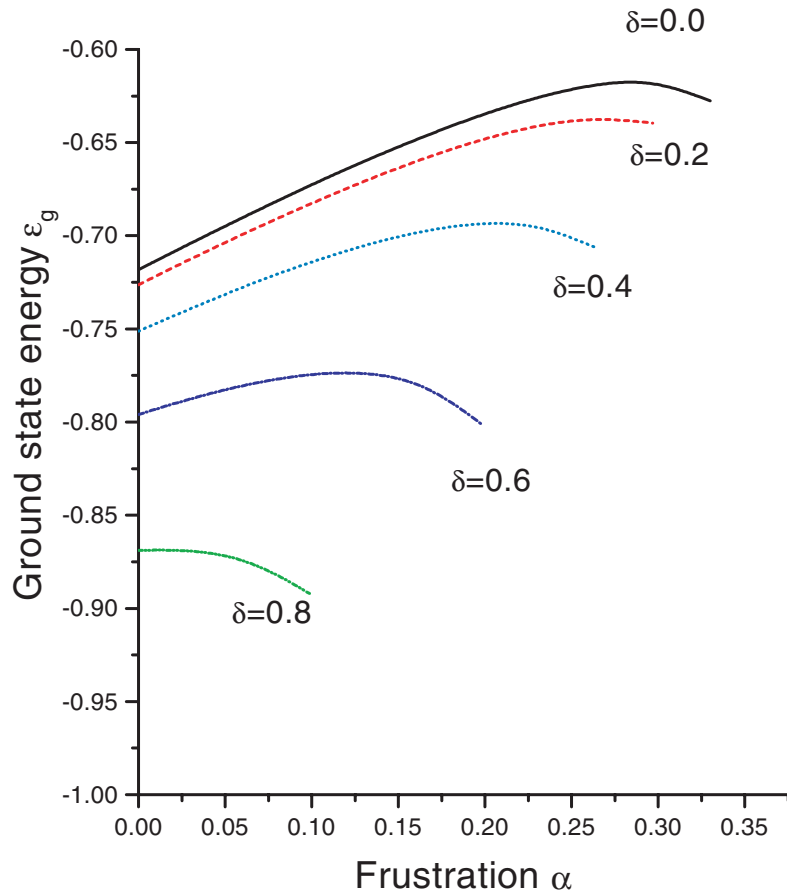


Figure 2. The ground-state energy ε_g of the alternating-spin chain $(1, \frac{1}{2})$ versus the frustration parameter α for different values of δ for $J^\pm = J_1(1 \pm \delta)$. The curve for each δ terminates at the respective α_c . The maximum in the ground-state energy occurs at an α that shifts to lower values with higher δ . In the dimer limit ($\delta \rightarrow 1$), the energy monotonically decreases with α , a feature peculiar to the combined effect of dimerization and frustration.

Both acoustic and optic excitation mode energies decrease as δ increases, as they do when α increases. This behaviour is shown in figure 3. The two excitation modes in all three spin systems scale with δ as δ^2 and linearly with α . There is a critical value of α at which the elementary excitation modes start to soften, signalling a transition from a Néel-like spin structure to a spiral structure [6]. This critical value, that we call α^* and evaluate from the changing signs of the slopes of the dispersion curves, is different for the acoustic and optical modes and in the presence of dimerization is δ -dependent:

$$\alpha_{acoustic}^* = \frac{s_1 s_2}{2(s_1^2 + s_2^2)} J_p (1 - \delta^2) \quad (16a)$$

$$\alpha_{optical}^* = \frac{1}{4} J_p (1 - \delta^2). \quad (16b)$$

For $\delta = 0$, the first of these reproduces the critical value reported by Ivanov *et al* [6] (denoted therein as J_c). A uniform decrease of α^* with δ leads one to conclude that the transition to a spiral spin state caused by frustration is facilitated by dimerization. The spin-

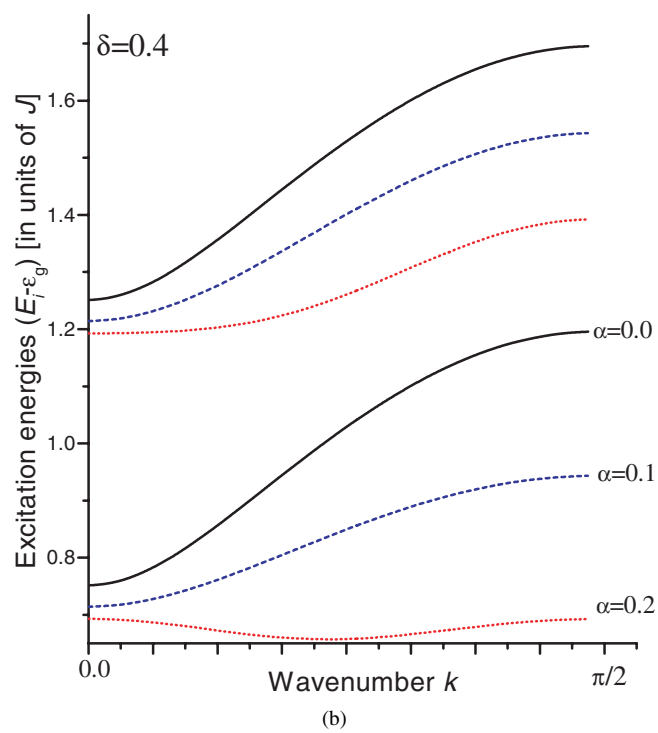
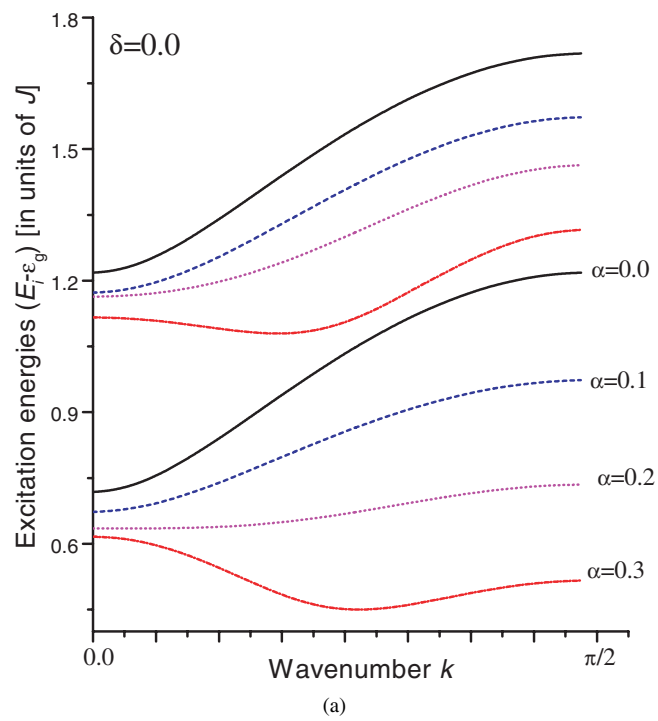


Figure 3. The elementary excitation spectra for the chain $(1, \frac{1}{2})$ for various values of the frustration parameter: (a) $\delta = 0.0$ and (b) $\delta = 0.4$. The behaviour is schematically the same for the other two spin systems $(\frac{3}{2}, 1)$ and $(\frac{3}{2}, \frac{1}{2})$.

wave theory gives different behaviours of the mode softening in the two elementary excitation modes; in the case of the ferromagnetic mode, the mode softening starts at an α that depends upon the magnitudes of the two component spins, while in the case of the antiferromagnetic mode it is uniform for all of the pairs of the ferrimagnet-forming spins.

The magnetization of the two sublattices, as given by equation (10), decreases in magnitude with both δ and α as shown in figure 4. The decrease with α indicates the destruction of magnetic order.

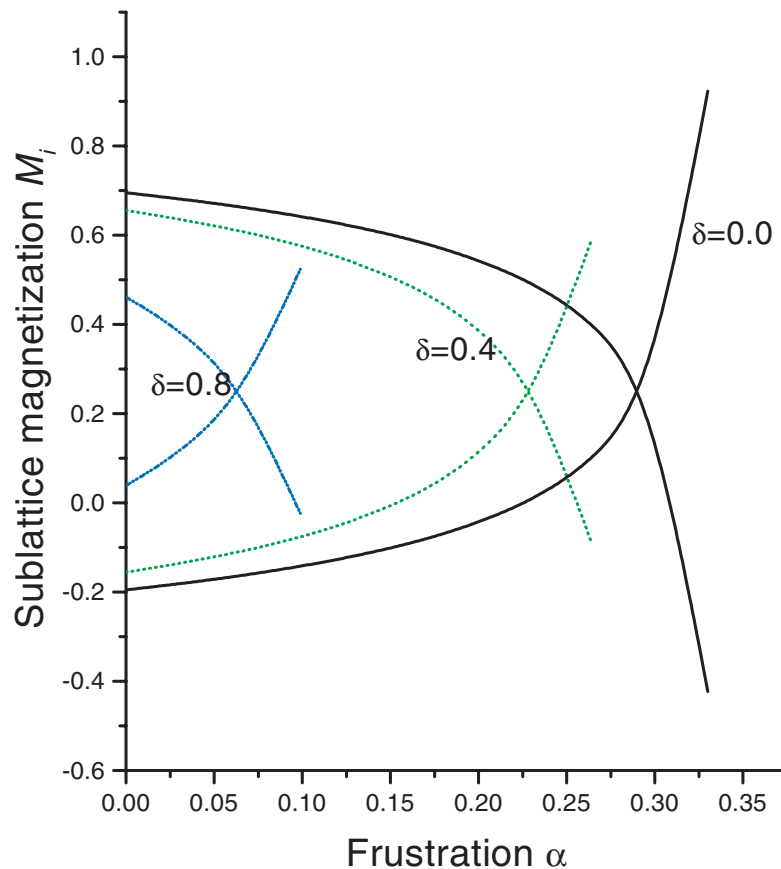


Figure 4. Sublattice magnetizations for the chain $(1, \frac{1}{2})$ as functions of α and δ . The curves terminate at the respective values of α_c . The behaviour is schematically the same for the other two spin systems.

The spin–spin correlations decay rapidly with the spin–spin separation, as noted earlier also [6]. When fitted to the Ornstein–Zernike form, equation (15), the correlation length is also found to decrease with both δ and α , as shown in figure 5, indicating again the destruction of order.

3. Frustration on a square lattice

It has been argued earlier [5] that the choice of the nearest-neighbour interaction as $J(1 \pm \delta)$ is restrictive in the sense that it does not allow taking into account the change in spin–spin

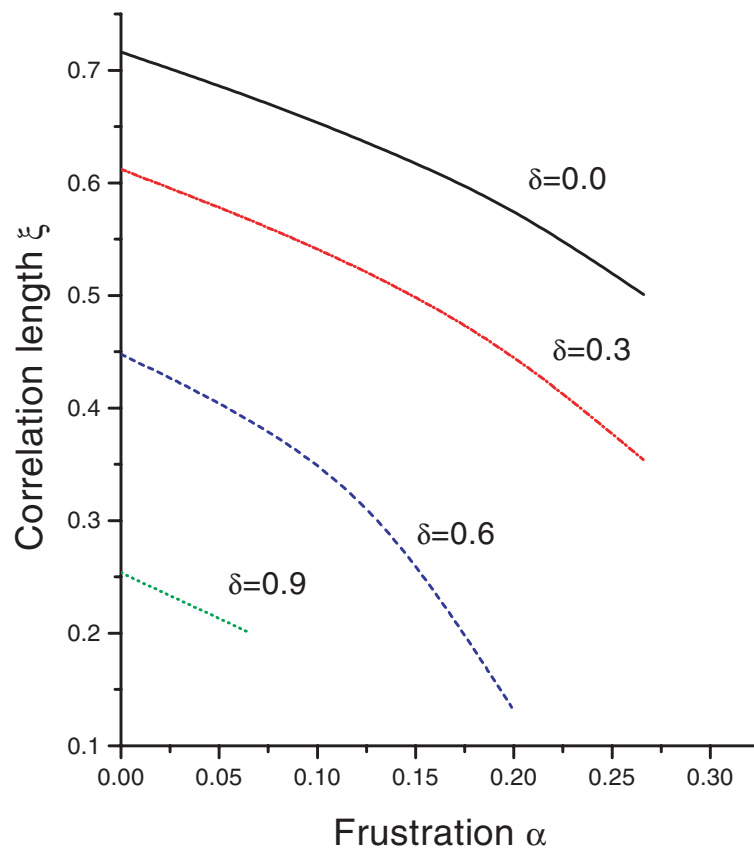


Figure 5. The variation of the correlation length ξ as a function of δ and α for the alternating-spin chain $(1, \frac{1}{2})$ versus the frustration parameter α , for different values of δ when $J^\pm = J_1(1 \pm \delta)$. The curves stop short of the respective critical values α_c because of the strong fluctuations that ξ experiences near these points. The results are schematically the same for the other two spin systems.

distances when looking into the several possible ways of distorting a square lattice during dimerization. A more general choice that we proposed was $J(r) \sim J/r$. In the case of nearest-neighbour coupling, this means that the amplitudes J^\pm in equation (1) are $J^\pm = J_1/(1 \mp \delta)$ which approximate to the more familiar $J_1(1 \pm \delta)$ in the limit of small δ . It was possible to show with this choice that among the various possibilities, the plaquette configuration is the lowest-energy deformation. It will be seen below that the choice also appears inevitable in studying the effects of frustration in a dimerizing two-dimensional lattice. In studying the combined effect of dimerization and the competing second-neighbour interactions on a square lattice, it becomes imperative to work with this form of interaction.

Since it has already been established that among the possible deformations of a square lattice, one that involves two phonons—one with wavevector $(\pi, 0)$ and the other with wavevector $(0, \pi)$ —forming a plaquette lattice, is energetically the most favourable one [5, 14–16], we will restrict our investigation to this kind of deformation alone.

We will write the Hamiltonian of a ferrimagnetic square lattice as a sum of the nearest-neighbour and the next-nearest-neighbour (or intersublattice and intrasublattice nearest-

neighbour) parts:

$$H = H_1 + H_2 \quad (17)$$

where

$$H_1 = \sum_{i,j}^{\sqrt{N}} \sum_{\lambda=\pm 1} J_\lambda [\mathbf{S}_{1,i,j} \cdot \mathbf{S}_{2,i+\lambda,j} + \mathbf{S}_{1,i,j} \cdot \mathbf{S}_{2,i,j+\lambda}] \quad (18)$$

$$H_2 = \sum_{i,j}^{\sqrt{N}} \sum_{\lambda,\lambda'=\pm 1} J_{\lambda,\lambda'} [\mathbf{S}_{1,2i,2j} \cdot \mathbf{S}_{1,2i+\lambda,2j+\lambda'} + \mathbf{S}_{2,2i,2j} \cdot \mathbf{S}_{2,2i+\lambda,2j+\lambda'}] \quad (19)$$

with $J_\lambda = 1/((1 - \lambda\delta))$, and

$$J_{1,1} = J_{-1,-1} = \frac{1}{\sqrt{2(1 + \delta^2)}} \quad (20a)$$

$$J_{-1,1} = \frac{1}{\sqrt{2}(1 + \delta)} \quad (20b)$$

$$J_{1,-1} = \frac{1}{\sqrt{2}(1 - \delta)}. \quad (20c)$$

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

$$A_1(k) = 2J_p s_2 - \frac{\alpha}{8} \left\{ \zeta_1^{(1)}(J_{1,1} + J_{-1,1}) + \zeta_{-1}^{(1)}(J_{1,1} + J_{1,-1}) \right\} \quad (21a)$$

$$A_2(k) = 2J_p s_1 - \frac{\alpha}{8} \left\{ \zeta_1^{(2)}(J_{1,1} + J_{-1,1}) + \zeta_{-1}^{(2)}(J_{1,1} + J_{1,-1}) \right\} \quad (21b)$$

$$B(k) = \Gamma(k) \sqrt{s_1 s_2} \quad (21c)$$

$$C = -2J_p s_1 s_2 + \frac{1}{2} \alpha (s_1^2 + s_2^2) \quad (21d)$$

$$\Gamma(k) = \sqrt{J_p^2 (\cos(k_x) + \cos(k_y))^2 + J_m^2 (\sin(k_x) + \sin(k_y))^2} \quad (22)$$

where

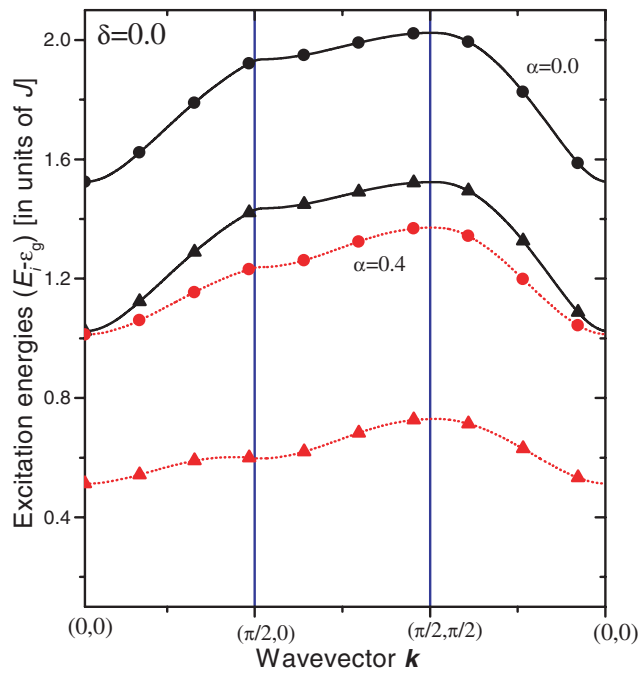
$$J_p = (J_{+1} + J_{-1})/4 = \frac{1}{2(1 - \delta^2)}$$

$$J_m = (J_{+1} - J_{-1})/4 = \delta J_p$$

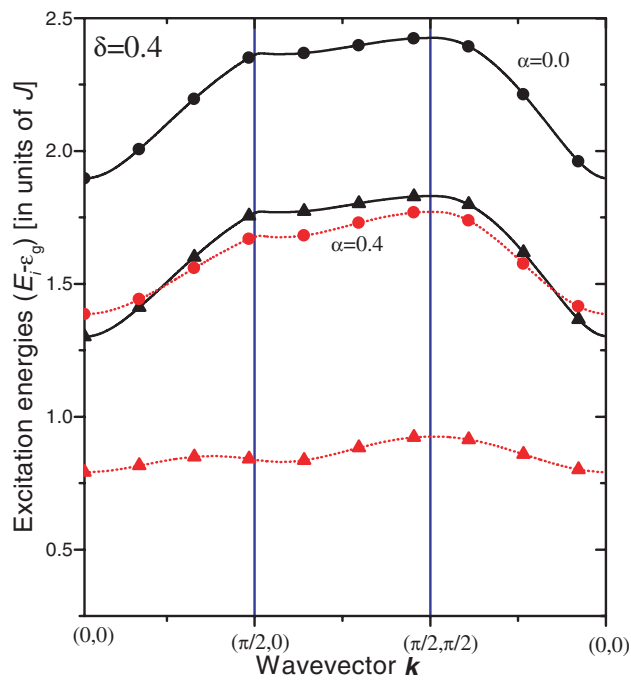
$$\zeta_\sigma^{(\tau)} = 2s_\tau [1 - \cos(k_x + \sigma k_y)] \quad \tau = 1, 2; \sigma = \pm 1.$$

The ground-state energy per site ε_g defined in equation (5), the energies of the two excitation modes $E_i(k)$ in equations (6) and (7), the staggered magnetization M_i defined in equations (10), and the correlation length defined in equation (15) can now be calculated as functions of the dimerization parameter δ and frustration parameter α . Setting $\alpha = 0$ we reproduce the results for the unfrustrated dimerized ferrimagnetic square lattice [5].

The linear spin-wave theory shows that, like that of the chain, the ground-state energy of a square lattice decreases with δ and increases with α . As reported earlier [5], an unfrustrated ferrimagnetic square lattice has a dependence of its ground-state energy on δ as $\delta^{1.5}/|\ln(\delta)|$. We now also find that the ground-state energy scales as $\alpha^{0.5}$ for any fixed value of dimerization. This is true for all pairs of spins forming the ferrimagnet.



(a)



(b)

Figure 6. The elementary excitation dispersion relations of the ferrimagnetic system $(1, \frac{1}{2})$ on a square lattice. The spectra are shown for different α and for (a) $\delta = 0.0$ and (b) $\delta = 0.4$. Curves with circles represent optical modes while curves with triangles represent acoustic ones.

The elementary excitation spectra are plotted for the system $(1, \frac{1}{2})$ in figure 6 along the principal symmetry directions in the irreducible Brillouin zone. The same schematic dispersion relations were found for the other two systems. The acoustic and optic modes again have ferromagnetic and antiferromagnetic characters respectively, and both of them are pushed up by dimerization and pulled down by frustration. The optic mode at $\mathbf{k} = (0, 0)$ is δ -dependent, a result not of the dimensionality of the lattice but of the interaction $J^\pm = J_1/(1 \mp \delta)$, as explained earlier [5].

As in the chains, the competing second-neighbour interaction also causes a transition from a Néel-like state to a spiral state, indicated by softening of the excitation modes. α^* , the critical value at which the transition takes place, in the case of a square lattice is also δ -dependent:

$$\alpha^* = \frac{s_1 s_2}{\sqrt{2}(s_1^2 + s_2^2)} (1 + \delta^2)(2 - \delta^2). \quad (23)$$

This is different from the α^* for chains in that it is the same for both ferromagnetic and antiferromagnetic modes. This relation also shows that for $\delta = 0$, the value of α^* for a square lattice is $2\sqrt{2}$ times larger than that for a chain. Moreover, unlike a monotonically decreasing α^* for a chain, it is a function that is peaked towards higher values of δ , as shown in figure 7. This indicates that while the transition to a spiral state in a square lattice is initially opposed by dimerization, it is facilitated at larger magnitudes of lattice deformation. This turnaround in behaviour occurs at $\delta = 1/\sqrt{2}$. Softening of the acoustic (ferromagnetic) mode is quite clearly discernible.

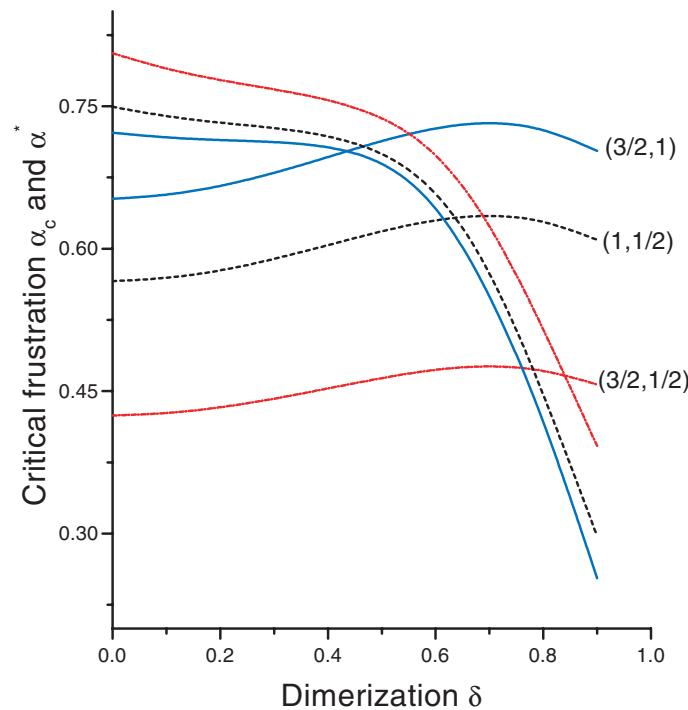


Figure 7. The dependence of α^* and α_c on the dimerization parameter δ for the three spin systems on the square lattice. The value of δ at which the peak occurs is independent of the spin components of a ferrimagnetic system. The lower three curves at $\delta = 0$ show α^* and the upper three α_c .

The sublattice magnetization M_i decreases with both α and δ as shown in figure 8. The

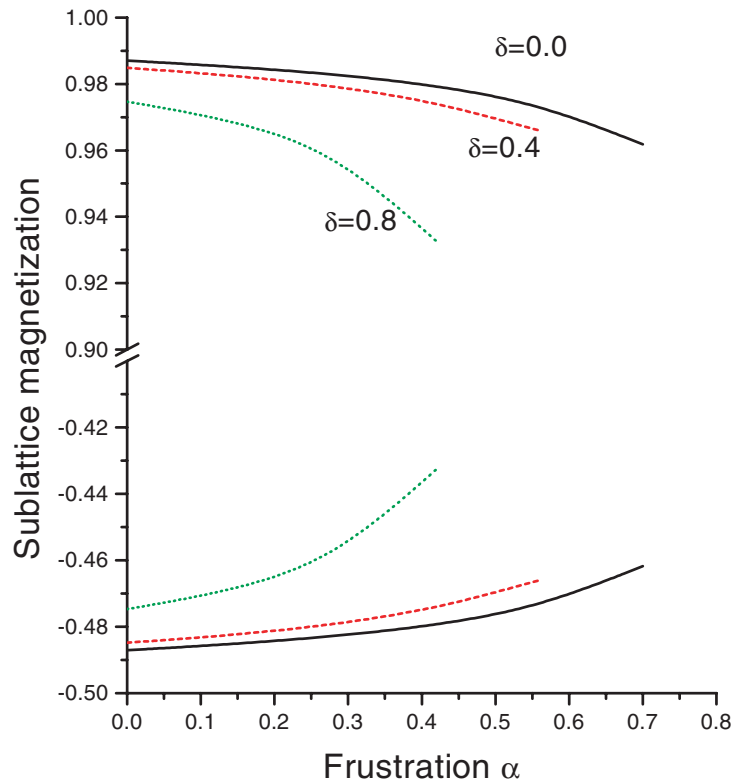


Figure 8. The α - and δ -dependence of the staggered magnetization of a square-lattice $(1, \frac{1}{2})$ ferrimagnet. The behaviour is schematically the same for the other spin systems.

δ -dependence for an unfrustrated square lattice was given in an earlier report [5]. For a non-dimerized square lattice, the magnetization has a logarithmic power-law scaling behaviour with the frustration parameter $\alpha^{1.5}/|\ln \alpha|$. The same scaling law was found for a dimerized plaquette.

The correlation function defined in equation (15) is calculated with

$$\langle O \rangle = \frac{1}{N} \sum_{\mathbf{k}} [\cos(k_x n_x) + \cos(k_y n_y)] u(\mathbf{k}) v(\mathbf{k}). \quad (24)$$

These correlations were found to have a more rapid decay with distance than in a chain lattice. The correlation length ξ in a square lattice also decreases with both δ and α as shown in figure 9. There is a clear minimum in the correlation length at a certain α that shifts to higher values with δ .

In summary, a simple linear spin-wave theory brings out quite a few new features in ferrimagnetic systems under the combined effects of dimerization and frustration. The effects in both one- and two-dimensional ferrimagnetic systems are most visible in the elementary excitation spectra. Besides the critical value α_c of the frustration parameter at which the long-range order is destroyed, there is another critical value α^* at which the elementary excitations undergo a mode softening, indicating a transition from a Néel-like to a spiral state. The LSWT shows that dimerization facilitates this transition. Both of the critical values of α are δ -dependent. While the ground-state energy initially increases with increasing magnitude of frustration, it reaches a maximum and then decreases just before α reaches its critical value

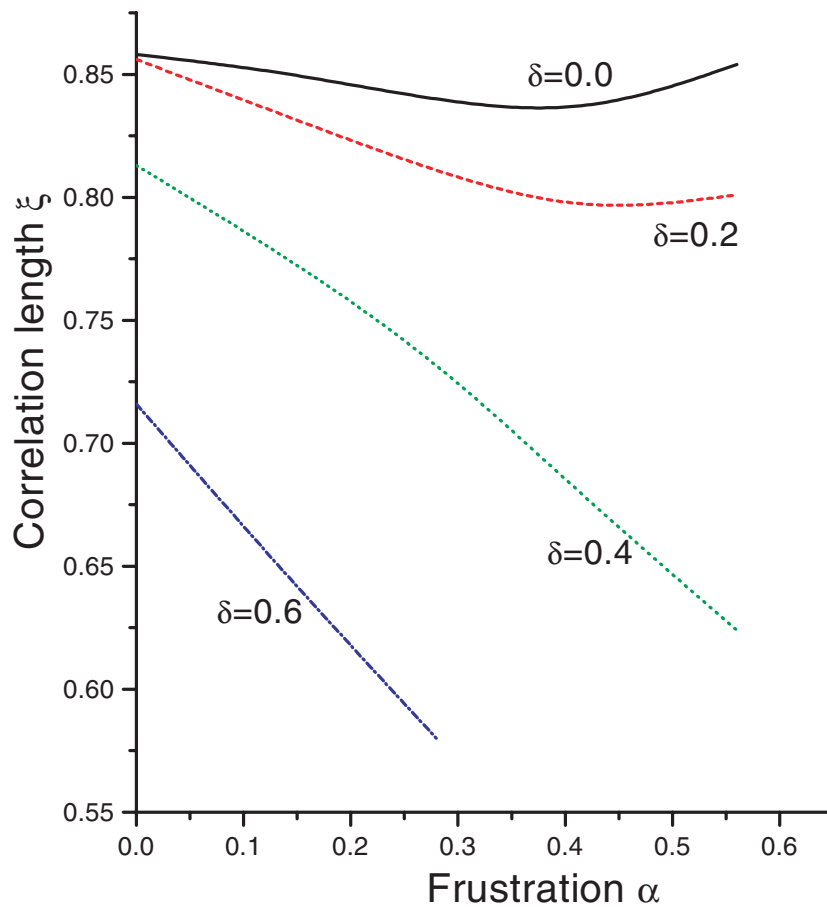


Figure 9. Correlation length ξ versus frustration parameter α for different values of the dimerization parameter δ for a square-lattice $(1, \frac{1}{2})$ ferrimagnet.

α_c . Both sublattice magnetization and correlation decrease as the strength of dimerization and frustration increases, indicating the loss of order. The theory also shows that on a square lattice, dimerization initially opposes the transition to a spiral state, but then beyond a certain critical value δ_c of the dimerization parameter, it facilitates the transition. The correlation length when fitted to the Ornstein–Zernike form shows a minimum with respect to α , at least for small values of δ .

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