

Ferro- and Antiferromagnetic Aspects of Alternating Spin Systems

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ABSTRACT

Using linear spin-wave theory we have investigated the thermal properties of frustrated dimerized Heisenberg ferrimagnetic system with alternating spins s_1 and s_2 on one- and two-dimensional lattices. At intermediate temperature the susceptibility χT and the specific heat C_v shows a minimum and a Schottky-like peak respectively. Frustration enhances the antiferromagnetic aspect in the system by causing a left-shift in the peak and the minimum which indicates that the antiferromagnetic behavior overbalance the ferromagnetic one at earlier temperatures. The effect of dimerization is different for the two form of the coupling constants. While the expanded form; $J(1 \pm \delta)$, boosts the antiferromagnetic behavior of the system by making a left-shift of the peak and the minimum, the distance-variable coupling constant; $\frac{J}{1 \mp \delta}$, shifts them to the right opposing, for a while, the appearance of the antiferromagnetic aspect. The slope of χT after the minimum shows that the aspect of ferrimagnetic system with spins (3/2, 1) is more antiferromagnetic and the system with (3/2, 1/2) is ferromagnetic. Free energy and magnetization decreased by increasing dimerization as well as frustration. Both of them scales with T^2 . PACS numbers: 75.10.Jm, 75.50.Ge.

Keywords: Spin Wave Theory, Alternating Spins, Specific Heat, Schottky Peak, Susceptibility

1. Introduction

Many attentions were made towards the low-dimensional antiferromagnets after Halden made his prediction that the integer-spin chain is massive, whereas the half-odd-integer spin chain is massless. This stimulated also several attempts to investigate the quantum behavior of chains consisting of two types of spins. An integrable model of this type was constructed by De Vega and Woynarovich [1], which allows us to guess the essential consequences of chains with spins of different length S . Recently [2], several authors discussed in detail such a bimetallic chain with spins $s = 1$ and $1/2$ as the simplest case.

The thermal behavior was also investigated for ferrimagnetic chains [3-13]. Besides verifying the existence of two (gapped and gapless) excitation modes, the specific heat and magnetic susceptibility of ferrimagnetic chains were shown to depend upon temperature as $T^{1/2}$ and T^{-2} respectively at low temperatures using modified spin-wave theory (MSWT) [6,7], density matrix renormalize-

tion group (DMRG) [7], quantum Monte Carlo method (QMC) [6,7,11], and Schwinger boson mean field theory (SB) [5]. Susceptibility was shown to have a minimum at intermediate temperature [3-5,7]. Specific heat was having a maximum [3,4,7,9-13] at mid temperature showing a Schottky-like peak. The spin correlation length behaves as T^{-1} at low temperature [6,8]. Using those techniques, it was shown also that the ferromagnetic behavior vanishes by increasing T , while the antiferromagnetic one persists up to higher temperatures.

Modified spin wave theory, which includes Takahashi constraint or non-linear spin wave theory, was also shown to give results in surprisingly good agreement with those from quantum Monte Carlo method in the thermodynamic limit of this system [6,7].

In this paper we will study alternating spin systems formed with a spin values; (s_1, s_2) using linear spin wave theory. We would like to investigate the thermal properties of the lattice magnetization, specific heat, free energy, and susceptibility. We would like to see how their thermal behavior depends on dimerization and frustration.

We shall then study alternating spin systems on a square lattice.

2. One Dimensional Alternating System

The dimerization of chains with spins s_1 and s_2 ($s_1 > s_2$) on alternating sites with competing antiferromagnetic nearest and next nearest neighbor couplings, J_1 and J_2 respectively, may be described by the Hamiltonian

$$H = \sum_i J_1^i S_i^A \cdot S_{i+1}^B + J_2 \sum_i [S_i^A \cdot S_{i+2}^A + S_{i+1}^B \cdot S_{i+3}^B], \quad (1)$$

with S^A belongs to the first sublattice with spin s_1 and S^B to the second one with spin s_2 . J_1^i is the alternating coupling between two adjacent sites, it gives the displacement of the i th atom through $u_i = 1/2 \cdot (-1)^i \delta$. The exchange coupling constants defined in Equation (1) can have different forms. In order to take into account changing distances in the course of spin-Peierls distortions, we assume that the nearest-neighbor spin-spin exchange coupling is itself distance dependent [13]

$$J(r) = \frac{J}{r}. \quad (2)$$

instead of the often-used $J(1 \pm \delta)$, which in any case is an approximation of $\frac{J}{1 \pm \delta}$.

The above Hamiltonian can be linearized using spin-wave analysis with the help of Holstein-Primakoff transformations to bosonic spin-deviation operators. The linearized Hamiltonian can be diagonalized using Bogoliubov transformations in terms of normal modes as

$$\tilde{H} = \sum_k [\varepsilon_g + E_1(k) \alpha_k^\dagger \alpha_k + E_2(k) \beta_k^\dagger \beta_k] \quad (3)$$

with ε_g is the ground state energy and $E_{1,2}$ are the acoustic and a optical modes for this system. A detailed study of the above system at zero temperature was discussed in our previous paper [2].

The free energy of the system is obtained by

$$F = \varepsilon_g + \sum_k [n_a E_1(k) + n_b E_2(k)]. \quad (4)$$

Static susceptibility χ is obtained by taking thermal averages [15] of the longitudinal correlations as

$$\chi = \frac{1}{3T} \sum_k [n_a (n_a + 1) + n_b (n_b + 1)] \quad (5)$$

The sublattice magnetization is calculated by taking the thermal average of the occupation number as

$$M_1 = s_1 - \sum_k \tilde{n}_k^a \quad (6)$$

$$M_2 = \sum_k \tilde{n}_k^b - s_2 \quad (7)$$

and

$$M_{tot} = M_1 + M_2 = s_1 - s_2 - \sum_k (n_a - n_b) \quad (8)$$

where

$$\tilde{n}_k^a = v^2 (n^a + n^b + 1) + n^a,$$

$$\tilde{n}_k^b = v^2 (n^a + n^b + 1) + n^b$$

and \sum_k runs over half of Brillouin zone and v_k^2 is the coefficient in Bogoliubov transformation; see Ref. [2]. n_a and n_b are bose distribution functions. Specific heat C_v is calculated by taking the differentiation of the free energy with respect to T .

The behavior of these physical quantities with temperature in presence of dimerization δ and frustration α parameters for different system (s_1, s_2) ; $(1, 1/2)$, $(3/2, 1/2)$ and $(3/2, 1)$, shall be discussed below. In chains we will investigate the thermal behavior using the expanded form of the coupling constant first and make a comparison with the behavior when the unexpanded form is used.

2.1. Dimerized Chains with $\alpha = 0$

2.1.1. Using Expanded Form

In a previous work [16] the thermal properties of chains were investigated for this kind exchange coupling constant and reported. We will mention briefly the results here for the sake of comparison. Free energy decrease with increasing temperature regardless the magnitude values of spins s_1 and s_2 . At $T = 0$ it shows the ground state energy of the system. It was founded that dimerization at $T = 0$ reduces ground state energy [2,14], it does the same with free energy. We found that the decrease of the free energy goes as T^2 .

Total magnetization decreases with increasing T and δ , with a slower decrease for higher dimerization. This decrease shows a power law behavior goes as T^2 .

It is known that ferromagnetic materials show a decaying susceptibility with temperature and increasing for antiferromagnet. Ferrimagnets are believed to have a combined effect of both behaviors [4,8,9,17,21,18], ferromagnetic and antiferromagnetic. Susceptibility times temperature decreases at low temperature showing a minimum and then starts to increase showing that the antiferromagnetic behavior is not smeared out after the minimum. Activating the effect of dimerization, we found that dimerization pushes χT up and causes a shift of this minimum to lower values of T at larger values of δ for all the three spin systems discussed here. The slope after the minimum varies from spin system to another. While the slope changes minutely with increasing dimerization parameter δ in the case of $(3/2, 1/2)$ it is much visible for $(3/2, 1)$. This behavior indicates that the spin system $(3/2, 1/2)$ acts like antiferromagnetic system

than a ferromagnetic one. It is vice versa for the system (3/2,1) This conclusion is in agreement with earlier one saying that the system with $s_1 > 2s_2$ is more ferromagnetic and the system with $s_1 < 2s_2$ is more antiferromagnetic.

Specific heat increases at low T , and goes as $T^{0.5}$, till it reaches a maximum where it shows a shoctky-like peak at intermediate temperature and then starts to decrease again. The antiferromagnetic excitation mode plays a major rule to this peak because it is gapped at $k = 0.0$, showing that after this peak the antiferromagnetic behavior is predominant. This peak is softening with the increase of dimerization parameter and shifts towards lower values of temperature. While at low temperature dimerization enhances the specific heat, δ reduces it at intermediate T for the three spin systems. We notice that at high δ specific heat become double peaked. This tell us that the ferromagnetic and antiferromagnetic modes are still gapless and gapped modes [19]. The double peak was not seen in the system with spins (3/2,1) at least in our range of temperature.

2.1.2. Using $\frac{J}{1 \mp \delta}$ as a Coupling Constant

As mentioned above, using $\frac{J}{1 \mp \delta}$ as a coupling constant is not of much important in the chains as it should be for the case of a square lattice. But we will investigate the thermal behavior of chains using the distance dependence coupling constant for the sake of having a complete picture of this form.

At zero temperature analysis[14] the acoustic mode showed a peculiar behavior with both forms of the coupling constants. While in the expanded form dimerization suppressed this mode the $\frac{J}{1 \mp \delta}$ used to enhance it. The same peculiar behavior was observed in the gapless mode at any finite temperature.

Free energy as well as total lattice magnetization show qualitatively a similar thermal behavior as the expand coupling constant one. Both were founded to decrease with increasing temperature, regardless the magnitude values of spins s_1 and s_2 , and both of them decrease with T as T^2 .

χT decreases at low temperature showing a minimum and then starts to increase showing that the antiferromagnetic behavior is not smeared out after the minimum. We found that dimerization in this case suppresses χT and shifts the minimum toward a higher values of T with the increase of δ for all the three spin systems. The slope after the minimum varies from spin system to another. While the slop changes minutely with increasing dimerization parameter δ in the case of (3/2,1/2), it is more visible for (3/2,1) and it is the

most for (1/2). This behavior indicates that the spin system (3/2,1/2) acts like antiferromagnetic system than a ferromagnetic one. It is vice versa for the system (3/2,1). Although this form delays the appearance of antiferromagnetic mode but the general behavior of the system remains unchanged. As a conclusion, we found an agreement with earlier calculations that the system with $s_1 > 2s_2$ is more ferromagnetic and the system with $s_1 < 2s_2$ is more antiferromagnetic.

Specific heat shows the same different behavior with dimerization. Shoctky-like peak at intermediate temperature shifts towards larger values of T . That means: the antiferromagnetic excitation mode, which plays a major rule to this peak, goes to a little higher temperature before it become predominant with the increase of dimerization. While at low temperature dimerization suppresses the specific heat, δ pushes it up at intermediate T for higher values of dimerization.

2.2. Frustrated Ferrimagnetic Chain with $\delta = 0$

The effect of frustration in ferrimagnetic systems was investigated [2,20] at zero temperature. Several characterization of the frustration parameter were reported; mainly, α_c which marks the complete destruction of the long range order in the system and α^* marks the onset of spiral phase transition. At any finite temperature, we found that they are temperature independent. We will study the thermal effect of a frustrated chain in the absence of dimerization.

Free energy decreases with frustration. This decrease is faster for higher α , and scales with T as T^2 . At the critical value of frustration, α_c , the free energy become imaginary and our LSW theory fails to go beyond this value of frustration. It was reported earlier [2,20] that at $T = 0.0$ the ground states energy increases with frustration parameter and then starts to decrease for values of frustration close to α_c .

Like the dimerization effect in ferrimagnetic chains, the total magnetization of the system decreases with increasing temperature and frustration. At $\alpha = \alpha^*$ we found that the reduction on magnetization is faster because of phase transition at this point.

The shoctky-like peak at intermediate temperatures is also observed in the specific heat with frustration. Frustration enhances antiferromagnetic mode by causing the peak to shift to lower values of temperature. Below this maximum, C_v is suppressed with the increase of frustration parameter α .

Susceptibility time temperature decreases at small T to a minimum after which it starts to increase for the three spin systems. As in the case of Schottky-like peak in specific heat, frustration makes this minimum in χT to shift towards smaller values of T verifying the en-

hancement of antiferromagnetic excitation mode due to frustration and it also pushes the χT to higher values for larger values of α after the minimum. The slope after the minimum varies from spin system to another and shows an agreement with the previous conclusion: the spin system $(3/2, 1/2)$ shows antiferromagnetic aspect more than a ferromagnetic one and the system $(3/2, 1)$ is more ferromagnetic than antiferromagnetic one.

3. Frustration on a Square Lattice

In a two-dimensional lattice, different phonons produce different types of dimerizations. A lattice distortion that involves two phonons, one with wavevector $(\pi, 0)$ the other with wavevector $(0, \pi)$, forming a plaquette lattice, was found to be the most favored one [14,22,23]. We will restrict our investigation to this kind of dimerization.

Using the untruncated exchange coupling defined in Equation (2) for both nearest neighbor and the next nearest neighbor interactions, the Hamiltonian of a ferrimagnetic square lattice can be written as a sum of the nearest neighbor and the next nearest neighbor parts:

$$H = H_1 + H_2 \quad (9)$$

$$H_1 = \sum_{i,j} \sum_{\lambda=\pm 1}^{\sqrt{N}} J_{\lambda} \left[S_{1,i,j} \cdot S_{2,i+\lambda,j} + S_{1,i,j} \cdot S_{2,i,j+\lambda} \right] \quad (10)$$

$$H_2 = \sum_{i,j} \sum_{\lambda,\lambda'=\pm 1}^{\sqrt{N}} J_{\lambda,\lambda'} \left[S_{1,2i,2j} \cdot S_{1,2i+\lambda,2j+\lambda'} + S_{2,2i,2j} \cdot S_{2,2i+\lambda,2j+\lambda'} \right] \quad (11)$$

with $J_{\lambda} = \frac{1}{(1+\lambda\delta)}$, and

$$J_{1,1} = J_{-1,-1} = \frac{1}{\sqrt{2}(1+\delta^2)} \quad (12)$$

$$J_{-1,1} = \frac{1}{\sqrt{2}(1+\delta)} \quad (13)$$

$$J_{1,-1} = \frac{1}{\sqrt{2}(1-\delta)} \quad (14)$$

Using HP transformation and Bogliubov transformation the above Hamiltonian can be diagonalized. The procedure of working out the physical quantities was discussed in detail in Ref. [2]. After that all the thermal quantities defined above are reproducible for the case of 2D.

3.1. Dimerization in a Non-Frustrated Square Lattice

Like in the chain, free energy and total magnetization,

defined in Equations (4) and (8) respectively, were found to decrease with increasing temperature as shown for the system $(1, 1/2)$ in **Figures 1** and **2** respectively. This is true regardless the magnitude values of spins s_1 , and s_2 . At $T = 0$ the free energy reduces [13,14] to the zero temperature ground state energy. We found that the decrease in both goes as T^2 in the three spin systems. Along with temperature, we found that dimerization suppress free energy with larger values of δ , while it increases the total magnetization at a fixed value of T .

Specific heat shows a shock-like peak at intermediate temperatures. At low temperature specific heat goes as $T^{0.5}$. This peak is softening with increasing of dimerization parameter and shifts towards higher values of temperature. While at low temperature, say below the peak temperature, dimerization reduces the specific heat, δ enhances specific heat at intermediate T as shown in **Figure 3** for the case of spin $(1, 1/2)$. The double peak is observed, not for the system $(3/2, 1)$, at least within our T range, which suggests that even at a high values of

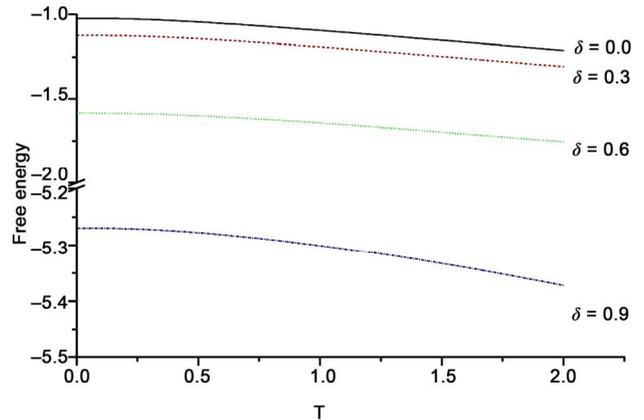


Figure 1. The free energy of a square lattice $(1, 1/2)$ ferrimagnet vs T for different values of δ .

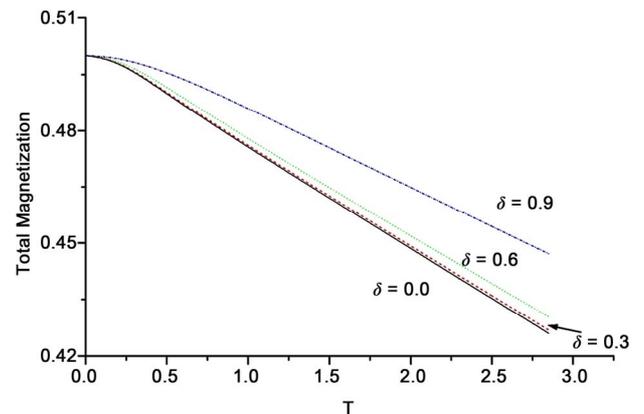


Figure 2. The total magnetization as a function of temperature in 2D $(1, 1/2)$ system for different values of δ .

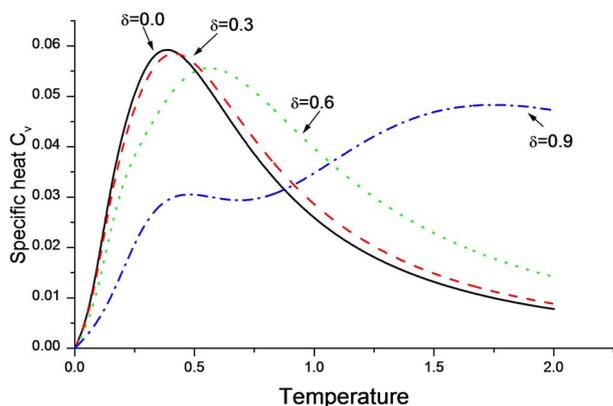


Figure 3. The specific heat of a square lattice with spin system (1, 1/2) against T in the presence of dimerization.

dimerization there is still a gapped and pagless modes in the systems. The above result again shows that the anti-ferromagnetic aspect is predominant in the case of intermediate temperature.

The susceptibility times temperature was also obtained using the Equation (5) for the square lattice. At zero dimerization it was reported by Yoshihiro *et al.* [24] that the uniform susceptibility shows an increasing behavior with temperature; although his diagrams started at $0.4T$, which we found there is a decrease before this increase as shall be discussed below.

We found that in χT , the ferromagnetic mode smeared out at T much earlier than the case of chains. The minimum, in support to the specific heat observations, shifts toward higher temperature, and after the minimum, χT further suppressed with the increase of dimerization parameter. This behavior is observed in the three spin systems and illustrated in **Figure 4**. From the slope of χT , for different δ values after this minimum, we found that even in 2D the system with $(3/2,1/2)$ shows anti-ferromagnetic behavior more than ferromagnetic and it is vis versa for the spin system $(3/2,1)$ as clear from **Figure 4**.

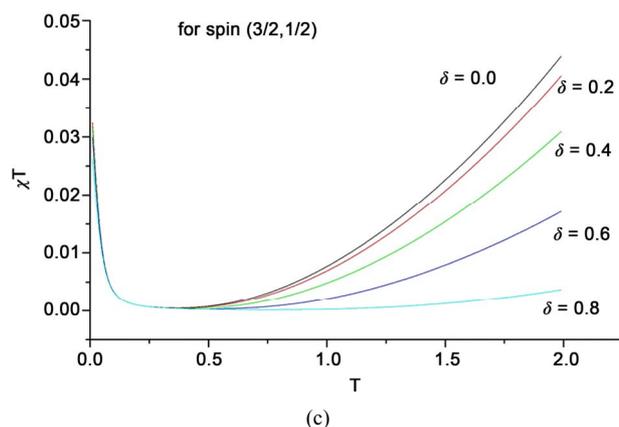
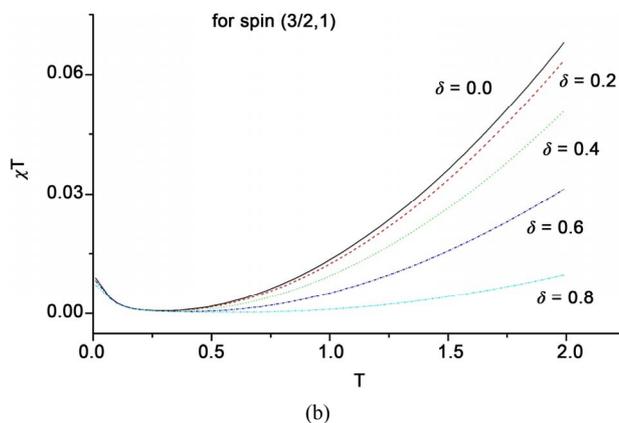
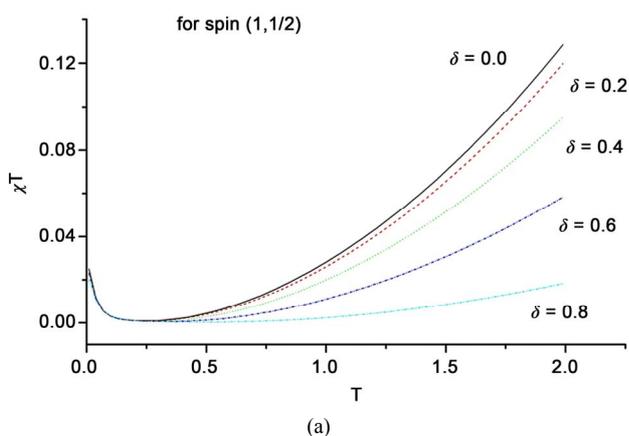


Figure 4. The behavior of χT vs T for several values on dimerization parameter shown for: (a) for the spin system (1, 1/2), (b) for (3/2, 1) and (c) for (3/2, 1/2).

3.2. Zero Dimerization in Frustrated 2D Ferrimagnetic Lattice

The free energy found to decrease with increasing frustration and also scales with T as T^2 . While temperature decreases the free energy it got increased with frustration parameter. This behavior is illustrated in **Figure 5** for the case of spin $(1, 1/2)$.

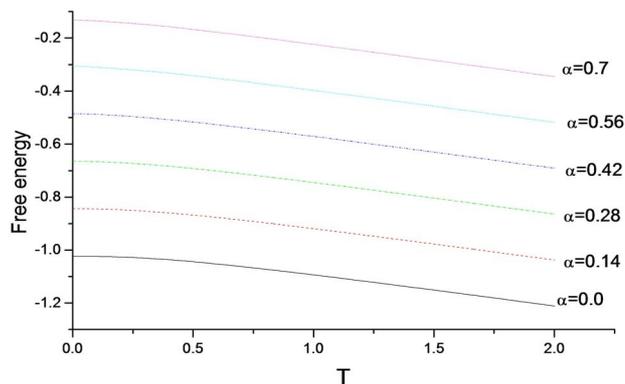


Figure 5. The free energy of a square lattice (1, 1/2) ferrimagnet vs T for different values of α .

Total magnetization shows a decrease with frustration and temperature. This decrease goes with T as T^2 . Unlike the case of free energy, frustration increases the reduction in total magnetization as a function of temperature until it reaches the transition value α^* , which after it will make a jump to higher values of magnetization and then starts again to decrease with increasing α as clear from **Figure 6**.

Specific heat shows a shoctky-like peak at intermediate temperatures for different values of frustration. While dimerization at low temperatures reduces the specific heat, frustration push it up. At intermediate T , frustration decreases C_v . The shoctky-like peak reduced in height with frustration and shifts towards smaller values of temperatures as can be seen from **Figure 7**. A double peak is observed in the three spin systems.

As shown in **Figure 8**, χT shows a minimum which shifts towards smaller T by introducing more frustration in the system indicating that frustration supports the enhancement of antiferromagnetic mode in these systems. After this minimum frustration increases the susceptibility. From this increase we reach to the same conclusion

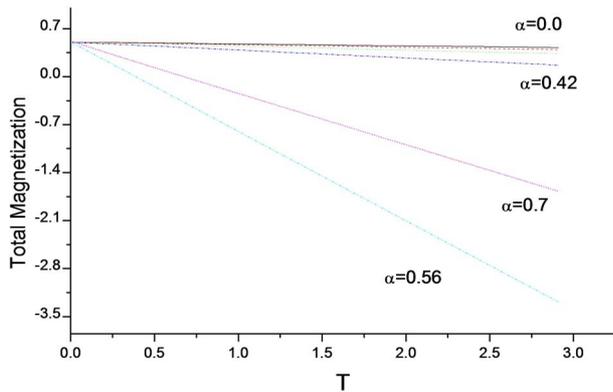


Figure 6. The total magnetization as a function of temperature in 2D (1, 1/2) system for different values of α .

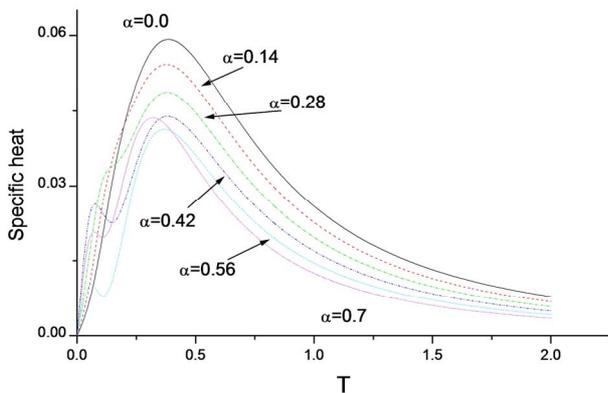


Figure 7. The specific heat of a square lattice with spin system (1, 1/2) against T in the presence of frustration.

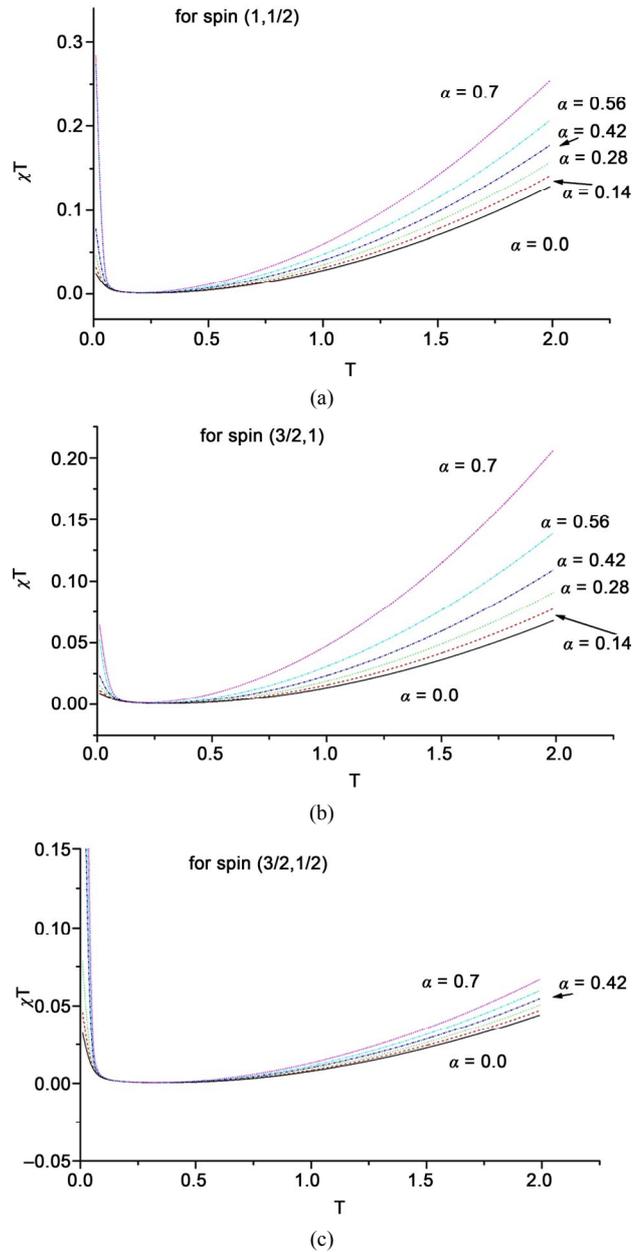


Figure 8. The behavior of χT vs T for several values on frustration parameter shown for: (a) for the spin system (1, 1/2), (b) for (3/2, 1) and (c) for (3/2, 1/2).

that in a square ferrimagnetic lattice the spin system (3/2, 1/2) shows antiferromagnetic behavior more than ferromagnetic and it is vis versa for the spin system (3/2, 1).

In summery we have used linear spin wave theory to study the thermal properties of ferrimagnetic system in the presence of frustration as well as dimerization parameters. In a chain, we found that dimerization enhances the antiferromagnetic aspect by taking coupling constat as $J(1 \pm \delta)$, it delays the mode in the case of

$\frac{J}{1 \mp \delta}$. In chains as well as in square lattices we found that the spin system $(3/2, 1/2)$ shows antiferromagnetic behavior more than ferromagnetic and the spin system $(3/2, 1)$ has a ferromagnetic one more than the antiferromagnetic aspect. We also found that both magnetization and free energy get decreased with frustration as well as dimerization and scales to square law of behavior with T .

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