Ferrimagnetism induced by asymmetrical ferromagnetic next-nearest-neighbour exchange interactions in an antiferromagnetic spin-1/2 zigzag chain*

Wang Zhong-Long(王忠龙)[a)†] and Fu Hua-Hua(傅华华)[b)]

[a)] *Science College, China Three Gorges University, Yichang* 443002, *China*

[b)] *School of Physics, Huazhong University of Science and Technology, Wuhan* 430074, *China*

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The magnetic properties of an antiferromagnetic bond alternating spin-1/2 zigzag chain with asymmetrical ferromagnetic next-nearest-neighbour exchange interactions at finite temperature are investigated by using the many-body Green's function theory. It is found that the ferrimagnetic ordering does not appear in the symmetrical next-nearest-neighbour coupling case, and takes place only for the asymmetrical next-nearest-neighbour case at finite temperature rather than the ground state. Furthermore, as the asymmetry degree of the next-nearest-neighbour exchange interactions increases, the ferrimagnetism becomes more and more dominant. It is shown that the elementary excitation spectra are responsible for the observed magnetic behaviour.

Keywords: ferrimagnetism, Green's function theory, zigzag spin chain

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1. Introduction

Since the first discovery of the organic ferromagnet para-nitrophenyl nitronyl nitroxide (p-NPNN),[1] the design and preparation of ferrimagnets have attracted considerable attention both experimentally and theoretically. The ferrimagnetism has long been considered as an effective strategy to produce spontaneous magnetizations through the antiferromagnetic exchange coupling between two kinds of magnetic centres with different spin quantum numbers.[2] Experimentally, Xu *et al.*[3] found that for the Sr-substituted Zn_2–Y hexagonal ferrite, the ferrimagnetic transition to the antiferromagnetic phase was responsible for the magnetic and the dielectric properties. Fan *et al.*[4] observed that the magnetic behaviour in dilute magnetic ferroelectric $(Bi_{0.46}Na_{0.46}Ba_{0.08})TiO_3$:Co was attributed to the competition between the ferrimagnetic response and the spin–lattice coupling effect. It indicates that the ferrimagnetism in the inorganic material can be controlled by doping. In addition, theoretically, for the Heusler alloy Mn_2NiGe, the ferrimagnetism resulting from the antiparallel Mn(A) and Mn(B) atoms with different spin quantum numbers, was investigated by Luo *et al.*[5] using the first-principles theory. The elementary excitations responsible for the ferrimagnetism in the Heisenberg alternating-spin chain with $S = 1/2$ and 1 antiferromagnetically coupled to each other were investigated by Yamamoto *et al.*[2] by using the Monte Carlo technique as well as the exact-diagonalization method.

Likewise, the spin-1/2 zigzag chain with the nearest-neighbour (NN) and the next-nearest-neighbour (NNN) exchange interactions has also long attracted a great deal of attention.[6−9] One of the important reasons is that the competition between the NN and the NNN exchange interactions makes the spin systems present various intriguing phenomena and peculiar properties, giving rise to the investigation of some interacting quantum models profoundly, such as the Haldane state in a spin-1 chain,[10] a dimer state in a spin-1/2 chain with the bond alternation,[11] and a strong frustrated spin chain.[6] The spin-1/2 zigzag spin chain with competing exchange interactions has been investigated theoretically for many years based

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†Corresponding author. E-mail: wzlsanxia@yahoo.cn

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on the following Hamiltonian:[8,12,13]

$$H = \sum_{i=1}^{N} (J_1 S_i \cdot S_{i+1} + J_2 S_i \cdot S_{i+2}). \tag{1}$$

Furthermore, many compounds, which can be described by the spin-1/2 zigzag chain with the NNN exchange interactions, have been synthesized experimentally, such as $SrCuO_2$,[14] $(N_2H_5)CuCl_3$,[15] $CuGeO_3$,[9] $LiCu_2O_2$,[16] $NaCu_2O_2$,[17] $Rb_2Cu_2Mo_3O_{12}$,[18] and In_2VO_5.[19] It seems that the magnetic properties of the spin-1/2 zigzag chain have been well established from the theoretical and the experimental points of view. In particular, many interesting low-dimensional quantum states, such as the frustrated spin state, the spin liquid state, the gapless antiferromagnetic (AFM) state, and the incommensurate state, have been found and described well in this spin-1/2 zigzag chain model with the NNN exchange interactions. It shows, however, that a ferrimagnetic ordered state seems to be excluded. Different from the alternating-spin chain, the AFM zigzag chain with uniform spin-1/2, wherein the asymmetrical NNN exchange interactions can be easily realized by chemical means, especially for organic compounds, will produce ferrimagnetic ordering, which is in favour of the design of the molecular device.[20] Thus, herein, we investigate the magnetic properties of the AFM spin-1/2 zigzag chain with the NNN exchange interactions to explore the possible occurrence of the ferrimagnetism by using the many-body Green's function theory (GFT).

2. Model and theoretical method

According to the structures of $Rb_2Cu_2Mo_3O_{12}$[18] and In_2VO_5,[19] the Hamiltonian of the one-dimensional spin-1/2 zigzag chain with the NNN exchange interactions (see Fig. 1) in an external magnetic field reads

$$\begin{aligned} H = \sum_{i}^{N} \Big\{ & \frac{J_1}{2}(S_{i-1,b} \cdot S_{i,a} + S_{i,b} \cdot S_{i+1,a}) \\ & + \frac{J_3}{2}(S_{i-1,a} \cdot S_{i,a} + S_{i,a} \cdot S_{i+1,a}) \\ & + \frac{J_4}{2}(S_{i-1,b} \cdot S_{i,b} + S_{i,b} \cdot S_{i+1,b}) \\ & + J_2 S_{i,a} \cdot S_{i,b} + h \cdot (S^z_{i,a} + S^z_{i,b}) \Big\}, \end{aligned} \tag{2}$$

where S_i is the spin operator at the i-th site, N is the total number of the spin repeating cells in the zigzag chain, and $h = g\mu_B B$ (Zeeman splitting), with g being the g factor, μ_B the Bohr magneton, and B an external magnetic field along the z-axis direction. Both J_1 and J_2 are the NN exchange interactions, and J_3 and J_4 are the NNN exchange interactions along the chain. If $J_3 = J_4$, the system will be reduced to a general one described by Hamiltonian (1). When $J_3 \neq J_4$, the system shows the asymmetrical NNN exchange interactions, which will be mainly focused on in this paper.

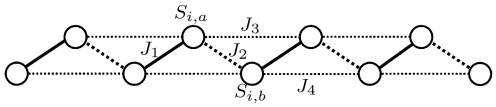


Fig. 1. Sketch of the spin-1/2 zigzag chain. Open circles represent spines with $S = 1/2$ coupled by bond exchange interactions J_1, J_2, J_3, and J_4, i denotes the superlattice number, and a unit cell is composed of two subcells $S_{i,a}$ and $S_{i,b}$.

The method we used is the many-body GFT, which has long been considered as a powerful tool to treat the Heisenberg model of spin systems.[20–24] The double-time retarded Green's functions, according to Bogolyubov and Tyablikov,[21,25] are defined as

$$\begin{aligned} G^{AB}_{ij}(t-t') &= \langle\langle A_i(t); B_j(t')\rangle\rangle \\ &= -\mathrm{i}\theta(t-t')\langle[A_i(t), B_j(t')]\rangle, \end{aligned} \tag{3}$$

where A and B are two operators, subscripts i and j label the lattice sites, $\theta(t-t')$ denotes the step function, and $\langle\cdots\rangle = \mathrm{Tr}(\mathrm{e}^{-\beta H}\cdots)/\mathrm{Tr}(\mathrm{e}^{-\beta H})$ is the average over the ensemble with the inverse temperature $\beta = 1/(k_B T)$, with k_B being the Boltzman's constant, and T the absolute temperature. For the time Fourier transformation, the retarded Green's function is put into the following equation of motion in energy space,

$$\omega G^{AB}_{i,j}(\omega) = \langle[A_i; B_j]\rangle + \langle\langle[A_i, H]; B_j\rangle\rangle_\omega. \tag{4}$$

For further Fourier transformation into the k-space, the Green's function can be expressed as

$$G^{AB}_{i,j}(\omega) = \frac{1}{N}\sum_k g(k,\omega)\,\mathrm{e}^{\mathrm{i}k\cdot(i-j)}, \tag{5}$$

where lowercase letter k represents the wavevector, and the sum of wavevector k runs over the first Brillouin zone. Now the Green's function g is a function of wavevector k and frequency ω. According to the well-known spectral theorem,[21,26] the expectation values and the correlation functions can be calculated by

$$\begin{aligned} & \langle B_j A_i\rangle \\ &= \frac{\mathrm{i}}{2\pi N}\sum_k \mathrm{e}^{\mathrm{i}k\cdot(i-j)} \end{aligned}$$

$$\times \int \frac{\mathrm{d}\omega}{\mathrm{e}^{\beta\omega}-1}[g(k,\omega+\mathrm{i}0^+)-g(k,\omega-\mathrm{i}0^+)]. \quad (6)$$

There are two sub-lattices in each repeating unit, so operator A in the above formula is taken as $S^+_{i,a}$ and $S^+_{i,b}$; correspondingly, operator B is taken as $S^-_{i,a}$ and $S^-_{i,b}$, where $S^\pm = S^x \pm \mathrm{i}S^y$ are the usual spin raising and lowering operators, respectively. Then, a set of linear equations are obtained as

$$\sum_\lambda [\omega\delta_{\mu\lambda} - P_{\mu\lambda}(k)]g_{\lambda\beta}(\omega,k) = \langle[S^+_\mu, B_\beta]\rangle, \quad (7)$$

where $P_{\mu\lambda}(k)$ is a parameter matrix given by

$$P_{\mu\lambda}(k) = \begin{bmatrix} -\{h+(J_1+J_2)\langle S^z_b\rangle + 2J_3[1-\cos(k)]\langle S^z_a\rangle\} & \langle S^z_a\rangle(J_2+J_1\,\mathrm{e}^{-\mathrm{i}k}) \\ \langle S^z_b\rangle(J_2+J_1\,\mathrm{e}^{\mathrm{i}k}) & -\{h+(J_1+J_2)\langle S^z_a\rangle + 2J_4[1-\cos(k)]\langle S^z_b\rangle\} \end{bmatrix}, \quad (8)$$

and $\langle S^z_\alpha\rangle$ ($\alpha = a,\ b$) indicates the sublattice magnetization. In the above matrix equations, the high-order Green functions are decoupled by the random phase approximation (RPA) or by the Tyablikov decoupling scheme,[20–22] such as $\langle\langle S^z_\alpha S^+_\alpha; S^-_\beta\rangle\rangle \approx \langle S^z_\alpha\rangle\langle\langle S^+_\alpha; S^-_\beta\rangle\rangle$ ($\alpha = a,\ b$). Due to the translation symmetry, $\langle S^z_{i,\alpha}\rangle = \langle S^z_\alpha\rangle$ is independent of site i, and the lattice constant is set to be unity.

In Eq. (7), $g_{\lambda\beta}$ is a Green's function matrix corresponding to the operators $S^+_{i,a}$, $S^+_{i,b}$, $S^-_{i,a}$, and $S^-_{i,b}$ in the momentum space, which can be written as

$$g_{\lambda\beta} = \begin{bmatrix} \langle\langle S^+_a; S^-_a\rangle\rangle & \langle\langle S^+_a; S^-_b\rangle\rangle \\ \langle\langle S^+_b; S^-_a\rangle\rangle & \langle\langle S^+_b; S^-_b\rangle\rangle \end{bmatrix}. \quad (9)$$

By solving Eq. (7), we can obtain the detailed expressions for the Green's function $G_{\alpha\beta}(\omega) = \langle\langle S^+_\alpha; S^-_\beta\rangle\rangle_\omega$, $(\alpha,\beta = a,b)$ as follows:

$$G_{aa}(k,\omega) = \frac{\langle S^z_a\rangle[\omega+E_2(k)]}{[\omega+E_1(k)][\omega+E_2(k)]-\gamma_1(k)\gamma_2(k)}, \quad (10)$$

$$G_{ab}(k,\omega) = \frac{\langle S^z_a\rangle\gamma_2(k)}{[\omega+E_1(k)][\omega+E_2(k)]-\gamma_1(k)\gamma_2(k)}, \quad (11)$$

$$G_{ba}(k,\omega) = \frac{\langle S^z_b\rangle\gamma_1(k)}{[\omega+E_1(k)][\omega+E_2(k)]-\gamma_1(k)\gamma_2(k)}, \quad (12)$$

$$G_{bb}(k,\omega) = \frac{\langle S^z_b\rangle[\omega+E_1(k)]}{[\omega+E_1(k)][\omega+E_2(k)]-\gamma_1(k)\gamma_2(k)}, \quad (13)$$

where $E_1(k)$, $E_2(k)$, $\gamma_1(k)$, and $\gamma_2(k)$ can be written as

$$\tau_1(k) = h + (J_1+J_2)\langle S^z_b\rangle + 2J_3[1-\cos(k)]\langle S^z_a\rangle,$$
$$\tau_2(k) = h + (J_1+J_2)\langle S^z_a\rangle + 2J_4[1-\cos(k)]\langle S^z_b\rangle,$$
$$\gamma_1(k) = J_2 + J_1\,\mathrm{e}^{-\mathrm{i}k}, \quad \gamma_2(k) = \gamma_1(k)^*. \quad (14)$$

Thus we find that the two energy spectra $E_1(k)$ and $E_2(k)$ can be obtained through solving the equation $[\omega+\tau_1(k)][\omega+\tau_2(k)] - \gamma_1(k)\gamma_2(k) = 0$ as

$$E_1(k) = -\frac{1}{2}[\tau_1(k)+\tau_2(k)] + \sqrt{\frac{1}{4}[\tau_1(k)-\tau_2(k)]^2 + \gamma_1(k)\gamma_2(k)}, \quad (15)$$

$$E_2(k) = -\frac{1}{2}[\tau_1(k)+\tau_2(k)] - \sqrt{\frac{1}{4}[\tau_1(k)-\tau_2(k)]^2 + \gamma_1(k)\gamma_2(k)}. \quad (16)$$

From Eqs. (10), (13), (15), and (16), we can obtain the Green's function as

$$G_{aa}(k,\omega) = \frac{\langle S^z_a\rangle}{E_1(k)-E_2(k)}\left[\frac{E_1(k)+\tau_2(k)}{\omega-E_1(k)} - \frac{E_2(k)+\tau_2(k)}{\omega-E_2(k)}\right], \quad (17)$$

$$G_{bb}(k,\omega) = \frac{\langle S^z_b\rangle}{E_1(k)-E_2(k)}\left[\frac{E_1(k)+\tau_1(k)}{\omega-E_1(k)} - \frac{E_2(k)+\tau_1(k)}{\omega-E_2(k)}\right]. \quad (18)$$

Then the values of sublattice magnetization $\langle S^z_\alpha\rangle$ ($\alpha = a,\ b$) are easily obtained from the spectral theorem and are given by

$$\langle S^z_\alpha\rangle = \frac{1}{2} - \Phi_\alpha, \quad (\alpha = a,b), \quad (19)$$

where

$$\Phi_a = \frac{2}{N}\sum_k \frac{1}{E_1(k)-E_2(k)}\left\{[E_1(k)+\tau_2(k)]n\left(\frac{E_1(k)}{k_\mathrm{B}T}\right) - [E_2(k)+\tau_2(k)]n\left(\frac{E_2(k)}{k_\mathrm{B}T}\right)\right\}, \quad (20)$$

$$\Phi_b = \frac{2}{N}\sum_k \frac{1}{E_1(k)-E_2(k)}\left\{[E_1(k)+\tau_1(k)]n\left(\frac{E_1(k)}{k_\mathrm{B}T}\right)\right.$$

$$- [E_2(k) + \tau_1(k)] n\left(\frac{E_2(k)}{k_{\rm B}T}\right)\Big\}, \quad (21)$$

$$n\left(\frac{E_{1,2}(k)}{k_{\rm B}T}\right) = \left\{\exp\left[\frac{E_{1,2}(k)}{k_{\rm B}T}\right] - 1\right\}^{-1}.$$

Then the average magnetization in each repeating cell $M_{\rm tot}$ and the magnetic susceptibility χ are defined as

$$\langle M_{\rm tot}\rangle = \frac{1}{N}\sum_{i}^{N}(\langle S_{i,a}^{z}\rangle + \langle S_{i,b}^{z}\rangle),$$

$$\chi = \frac{\partial\langle M_{\rm tot}\rangle}{\partial h}. \quad (22)$$

The above equations can be solved self-consistently. In the calculation, an initial state composed of a set of magnetizations $\{\langle S_{\alpha}^{z}\rangle\}$ is put into the equations to produce the resultant magnetization. The iteration goes on until the convergence is reached.

3. Results and discussion

First of all, in order to clarify the effect of the ferromagnetic (FM) NNN exchange interactions on the magnetic ordering of the AFM spin-1/2 zigzag spin chain and to make a comparison, we investigate the magnetic properties of an AFM spin-1/2 zigzag chain without the NNN exchange interactions, i.e., $J_3 = J_4 = 0$. In the calculation, the bond alternation along the chain is taken into account. To make a comparison with different magnetic interactions, we choose parameters J and δ to describe the strength of the magnetic interaction and the bond alternation. Thus the interactions J_1 and J_2 can be set as $J(1+\delta)$ and $J(1-\delta)$, respectively. The temperature dependence of the sublattice magnetizations $\langle S_{\alpha}^{z}\rangle$ ($\alpha = a$, b) for different values of δ under an external magnetic field $h = 0.002|J|$ is shown in Fig. 2. It can be seen that sublattice magnetization $\langle S_{\alpha}^{z}\rangle$ decreases slowly at low temperature and then drops rapidly to zero at high temperature as expected. Meanwhile, sublattice magnetizations $\langle S_{a}^{z}\rangle$ and $\langle S_{b}^{z}\rangle$ are symmetrical with respect to the temperature axis, indicating the spin alignments of the neighbouring subspins are antiparallel, which results in zero net total magnetization and no ferrimagnetic ordering. We can also find that at zero temperature, the sublattice magnetizations are much smaller than their classical values, i.e., $|\langle S_{\alpha}^{z}\rangle| < 0.5$, which is attributed to the zero-point quantum fluctuation and the formation of the singlet-like state connected by the strong bond, resulting in the nonmagnetism of the system. Besides, the larger the dimerization δ is, the more dominant the singlet-like state at zero temperature is, and the smaller the sublattice magnetizations are.

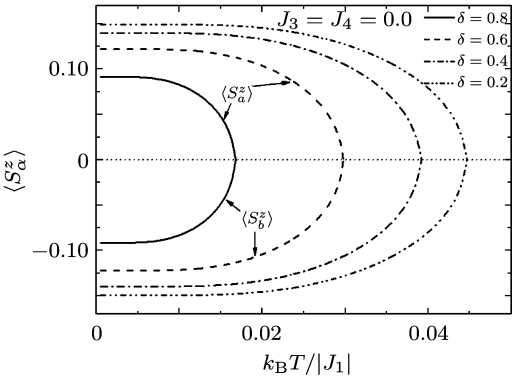


Fig. 2. Sublattice magnetizations versus temperature for an AFM spin-1/2 bond alternating chain without the NNN exchange interactions, under the field $h = 0.002|J|$. The δ denotes the bond alternation parameter, $J_1 = J(1+\delta)$, and $J_2 = J(1-\delta)$.

To further investigate the magnetic behaviour, we present the elementary excitation spectra for the AFM bond alternating spin-1/2 chain, as shown in Fig. 3. There are two branches of energy bands, $E_1(k)$ and $E_2(k)$, both of which have no energy gap at $k = 0$. From Eqs. (15) and (16), it is apparent that $E_1(k) > 0$ while $E_2(k) < 0$. In order to ascertain whether the energy bands are symmetrical with respect to the k-axis, we plot $|E_2(k)|$ for intuition in Fig. 3, wherein we find that $E_1(k) = |E_2(k)|$ for any k. At zero temperature, lower band E_2(k) is occupied by magnons, while upper band $E_1(k)$ is empty. Accordingly, from Eqs. (19)–(21), we can obtain $\langle S_{a}^{z}\rangle = -\langle S_{b}^{z}\rangle$, which results in no ferrimagnetic ordering at the ground state. Nevertheless, at low temperature, the excitations with minus energies can be regarded as the hole type, yielding the thermally activated particle–hole symmetry. Equations (19)–(21) also result in $\langle S_{a}^{z}\rangle = -\langle S_{b}^{z}\rangle$ without any ferrimagnetic ordering at finite temperature. In addition, it is found that the dispersion relations behave linearly as $k \to 0$, which is in agreement with the result of Yamamoto *et al.*[2] obtained by using the spin-wave theorem.

Now, we turn our attention to the effect of the FM NNN exchange interactions on the magnetic properties of the spin-1/2 zigzag chain. In order to give prominence to the effect of the asymmetrical FM NNN exchange interactions, it is necessary to discuss the effect in the symmetrical NNN exchange case, i.e., $J_3 = J_4$. The sublattice magnetizations $\langle S_{\alpha}^{z}\rangle$ ($\alpha = a$, b) of the system, each as a function of the temperature for $J_3 = J_4 = -0.1$ and -5.0 labeled by square

and circle respectively, are presented in Fig. 4. It is clearly seen that $\langle S_a^z \rangle$ and $\langle S_b^z \rangle$ are well symmetrical with respect to the temperature axis, implying that the total magnetization is still zero and no ferrimagnetism occurs, which is very similar to the case for the AFM spin-1/2 zigzag chain without the NNN exchange interactions discussed above. This behaviour was observed on compounds $Rb_2Cu_2Mo_3O_{12}$[18] and In_2VO_5.[19] As $h \to 0$, the net total magnetization $M \to 0$. It was also shown that with the temperature decreasing, the magnetic susceptibility displayed a round maximum, indicating the AFM feature of the low-dimensional magnetic materials.[18,27] Meanwhile, we can find that the sublattice magnetizations increase with J_3 and J_4 increasing, which is attributed to the fact that the strong FM NNN exchange interactions can enhance the FM excitations of the system and predominate over the AFM NN spin correlations responsible for the zero-point quantum fluctuation. Although the strong and symmetrical FM NNN exchange interactions can enhance the sublattice magnetizations, no net magnetization of the system can be produced at any temperature.

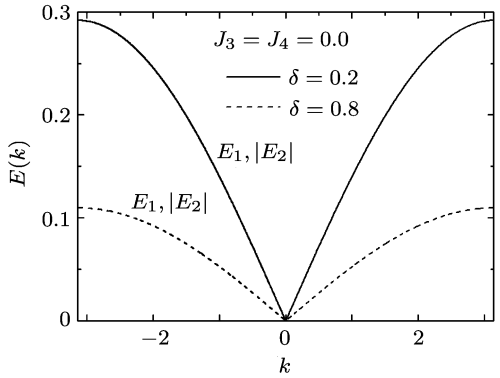


Fig. 3. Elementary excitation spectra of the AFM spin-1/2 zigzag chain without the NNN exchange interactions for $\delta = 0.2$ and 0.8.

If the FM NNN exchange interactions are asymmetrical, i.e., $J_3 \neq J_4$, the magnetic properties of the AFM spin-1/2 zigzag chain are much different. The temperature dependence of sublattice magnetizations $\langle S_\alpha^z \rangle$ with the asymmetrical FM NNN exchange interactions are also shown in Fig. 4. Here, in order to change the asymmetrical degree of the NNN exchange interactions, we set $J_4 = -0.1$ and make J_3 change from -1.0 to -5.0. As shown by the various lines in Fig. 4, we can see that both sublattice magnetizations $\langle S_a^z \rangle$ and $\langle S_b^z \rangle$ approach zero gradually with the increasing temperature and are no longer symmetrical with respect to the temperature axis. This implies that the magnetic system will exhibit a nonzero net magnetization and will be in a ferrimagnetic ordered state. It is interesting that with the temperature increasing, the total magnetization $\langle S_{\rm tot}^z \rangle$ $(= |\langle S_a^z \rangle + \langle S_b^z \rangle|)$ increases from zero to a maximum and then decreases to zero again slowly, which makes each of the curves of $\langle S_{\rm tot}^z \rangle$ as a function of the temperature appear as a round peak. Meanwhile, as the asymmetrical degree of the NNN exchange interactions increases, the total magnetization of the system is enhanced, and the round peak of the total magnetization shifts towards the high temperature. This implies that the asymmetrical FM NNN exchange interactions are responsible for the occurence of the ferrimagnetic ordering in the spin-1/2 zigzag chain. It is well known that a common ferrimagnetism is the resultant of two or more mixed spins carrying different magnetic moments with antiparallel spin alignments. Thus, the ferrimagnetic behaviour, which occurs in this quantum spin system with equal spin values and is induced by the asymmetrical FM NNN exchange interactions, is very different from the traditional one. Furthermore, another important point which should be stressed, is that as the temperature approaches zero, the total magnetization of the system vanishes, indicating that this ferrimagnetic behaviour occurs only at finite temperature rather than the ground state.

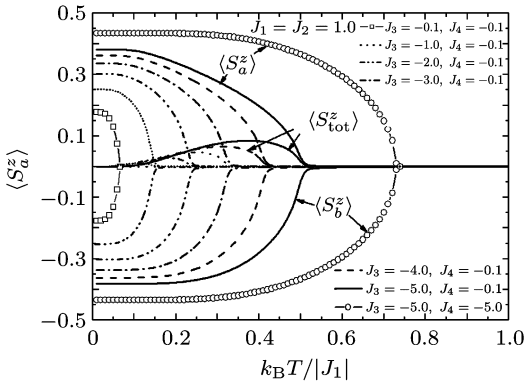


Fig. 4. Sublattice and total magnetizations versus temperature for the AFM spin-1/2 zigzag chain with the symmetrical and the asymmetrical FM NNN exchange interactions, where circle and square lines denote the symmetrical case, the other lines denote the asymmetrical case.

In order to give a further insight into the ferrimagnetic feature of the AFM spin-1/2 zigzag chain with the asymmetrical FM NNN exchange interactions, the magnetic susceptibilities multiplied by temperature χT each as a function of the temperature are shown in Fig. 5. It can be seen that in the low-temperature region, each of the χT values increases

rapidly to a maximum and then decreases with the temperature increasing, which makes each of the χT curves behave as a sharp peak. The peak is attributed to a small gap opened by a small external magnetic field. With the temperature further increasing, the χT value decreases to a minimum and then increases to a limiting value in the high-temperature region. The minimum, i.e., the upturn behaviour in the χT, suggests the possible occurrence of the ferrimagnetism in the low-dimensional spin system. It is also found that as the asymmetrical degree of the FM NNN exchange interactions increases, the minimum of the χT curve increases and moves towards the high-temperature region, which makes the ferrimagnetic behaviour of the system become clearer and clearer. On the contrary, as the asymmetry degree of the FM NNN exchange interactions decreases, the low-temperature sharp peak and the minimum in the χT curve are both suppressed, implying that the χT curve of the magnetic system approaches the AFM one, which was clearly observed in AFM spin ladder systems.[28] Accordingly, the asymmetrical FM NNN exchange interaction plays a crucial role for the occurrence of the ferrimagnetic ordering in the AFM spin-1/2 zigzag chain.

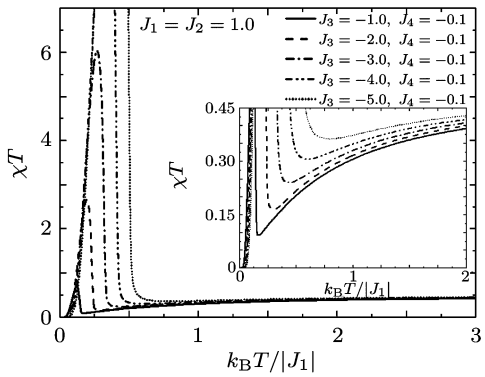


Fig. 5. Temperature dependences of magnetic susceptibility multiplied by temperature χT for the zigzag chain with asymmetrical FM NNN exchange interactions, in which J_3 varies from -1.0 to -5.0 and J_4 is kept a constant. The inset shows the low-temperature part of the plot.

It is also instructive to investigate the appearance of the ferrimagnetic ordering by tracing the change of the elementary excitation spectra for the AFM spin-1/2 zigzag chains with the symmetrical and the asymmetrical NNN exchange interactions (see Fig. 6). It shows that when $J_3 = J_4$, $E_1(k) = |E_2(k)|$, i.e., $E_1(k)$ and $E_2(k)$ are symmetrical with respect to the k-axis (see Figs. 6(a) and 6(b)), which is very similar to the case of the AFM spin-1/2 zigzag chain without the NNN exchange interactions described above. Furthermore, as J_3 and J_4 increase, we can find that the dispersion of the elementary excitation spectra becomes more apparent due to the enhancement of the FM excitations, which causes the quantum fluctuation of the spins to be suppressed and makes the sublattice magnetizations increase as shown by the circle and the square lines in Fig. 4.

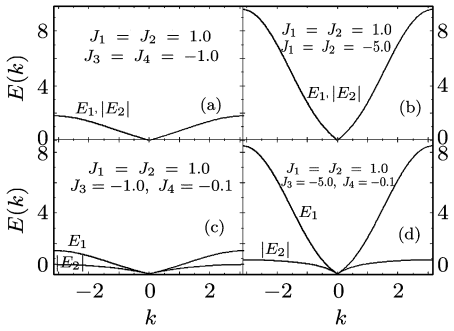


Fig. 6. Elementary excitation spectra of the spin-1/2 zigzag chain with the symmetrical (panels (a) and (b)) and the asymmetrical (panels (c) and (d)) NNN exchange interactions.

However, when the FM NNN exchange interactions are asymmetrical, i.e., $J_3 \neq J_4$, it is interesting to find out that the elementary excitation spectra of the system are also asymmetrical ($E_1(k) \neq |E_2(k)|$) with respect to the k-axis except for $k = 0$, as shown in Figs. 6(c) and 6(d), leading to the fact that the absolute values of sublattice magnetizations $\langle S_a^z \rangle$ and $\langle S_b^z \rangle$ are no longer equal to each other, which makes the spin system exhibit a nonzero total magnetization at finite temperature. Thus, a ferrimagnetic ordered state occurs in this case at finite temperature because of the asymmetrical particle–hole excitations. Meanwhile, as the asymmetrical degree of the NNN exchange interactions increases, i.e., $|J_4|$ is kept a constant and $|J_3|$ increases, it is obvious that the dispersion of the two branches of the spectra is enhanced and the relative difference $\Delta E = E_1(k) - |E_2(k)|$ increases, which results in the fact that the total magnetization increases and the ferrimagnetic ordered state of the system becomes more and more dominant. However, no matter how J_3 and J_4 vary at $T = 0$, energy band $E_1(k)$ is always empty, while energy band $E_2(k)$ is always filled by magnons, the relative difference ΔE still stays at zero at $k = 0$, so the AFM spin-1/2 zigzag chain does not exhibit any ferrimagnetic ordering at the ground state. At low temperature, due to the competition between the FM NNN and the AFM NN interactions and the asymmetrical FM NNN couplings, which result in the appearance of the asymmet-

rical particle-hole excitations, the ferrimagnetic ordering takes place.

4. Conclusion

The magnetic properties of an AFM spin-1/2 zigzag chain with the FM NNN exchange interactions are investigated by using the GFT to show the possible occurrence of the ferrimagnetism. We focus mainly on the effect of the FM NNN exchange interactions on the ferrimagnetic behaviour of the system. The results show that if the FM NNN exchange interactions are symmetrical, the magnetic system does not show any ferrimagnetic ordering at any temperature. When the FM NNN exchange interactions are asymmetrical, the absolute values of the sublattice magnetizations are no longer equal to each other, resulting in a nonzero total magnetization and the occurrence of the ferrimagnetic ordering at finite temperature, which is due to the asymmetrical particle–hole excitations. If the asymmetry degree of the FM NNN exchange interactions increases, the ferrimagnetic behaviour of the magnetic system becomes clearer and clearer. It should be stressed that the mechanism of the ferrimangnetism investigated here is very different from that of the common one stemming from the alternating or mixed spins with unequal spin quantum numbers in an antiparallel spin alignment. Our study would deepen further the understanding of the ferrimagnetic properties in the low-dimensional spin systems and be quite helpful for designing and synthesizing some ferrimagnetic compounds.

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