Thermodynamic properties and excitation spectrum of spin chain with antiferromagnetic - ferromagnetic interactions

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Abstract. Thermodynamic properties of antiferromagnetic spin chain with various AFM nearest-neighbor and FM next-nearest-neighbor exchange interactions are studied using functional integral method in $J_1 - J_2$ Heisenberg model. Besides, the excitation spectrum of the system is also given. The results show that the system has two distinct excitations with no energy gap from the ground state for any permissible values of $J_1$ and $J_2$ without single-ion anisotropy. Changing the exchange parameters influences on the thermodynamic properties and the excitation spectra of the system.

1. Introduction
The properties of quantum spin chains have recently attracted a lot of interest in both of theoretical and experimental sides due to their potential applications. Moreover, the development in the synthesis of magnetic materials has made the study of these systems be more and more advanced. Theoretically, the thermodynamic properties of the spin chain have been investigated with nearest neighbor (NN) Heisenberg model using various methods such as general Jordan-Wigner transformation and integral Gaussian transformation [1], mean-field Jordan-Wigner transformation [2]; second-order Green-function theory with arbitrary spin $S$ and quantum Monte Carlo simulations [3], transfer-matrix renormalization-group method [4]. The properties of such systems are strongly affected by quantum thermal fluctuations. When dimensionality of the magnetic system is reduced, the thermal fluctuation becomes to be enhanced which makes the magnetic order of the system decrease. Mermin and Wagner theorem [5] has shown quite generally that the one and two-dimensional isotropic Heisenberg models with interactions of finite range can be neither ferromagnetic (FM) nor antiferromagnetic (AFM) at nonzero temperature.

Another model was developed from the Heisenberg model by adding next nearest neighbor (NNN) exchange interactions, $J_1 - J_2$ model, which describes magnetic behavior of a zigzag spin chain (see figure 1). Recently, there have been a lot of compounds which are found out being zigzag chains, such as frustrated zigzag chain In$_2$VO$_5$ or antiferromagnetic zigzag chains (NH$_4$)$_2$VOF$_4$, K$_2$VOF$_4$ [6]. In [7] the authors showed that competition of $J_1 - J_2$ interactions together with frustrated interchain interactions play a key role in stabilizing spin-liquid type correlations in SrHo$_2$O$_4$ and SrDy$_2$O$_4$. Changing $J_1$ and $J_2$ influence strongly on properties of
the zigzag chains. When $J_2 < 0$ (AFM), the spin chain is a frustrated system, with the FM NN coupling $J_1 > 0$, modified spin wave approach showed that susceptibility depend on temperature $T$ as $\chi \sim T^{-1.2}$ in quantum case and $\chi \sim T^{-4/3}$ for classical model at quantum critical point with $J_1/J_2 = -4$ [8]. Mean field theory [9] indicated that the system is incommensurate in region $-2(\pi+1)/\pi < J_1/J_2 < 2$ and when $-2(\pi+1)/(\pi-1) > J_1/J_2 > -4$ this ground state has a finite total spin magnitude. With $J_1 < 0$ and $J_2 < 0$ (AFM-AFM), in [10] the authors found magnetization plateau at $1/3$ of full moment accompanying spontaneous symmetry breaking of translation in strongly frustrated region ($J_2/J_1 > 0.6$). From the above given results, one can see that the frustrated cases have been extensively studied in recent years. The remaining case, AFM-FM ($J_1 < 0$ and $J_2 > 0$), has attracted quite less attention from scientists, however according to me the system also displays interesting properties. So in this paper, I will study the thermodynamic properties and the excitation spectrum of the zigzag spin chain with AFM-FM exchange interactions for arbitrary spin using the functional integral method.

2. Theory

Considering a chain of $N$ spins as given in figure 1. In the figure 1, $J_1$ is exchange interaction constant between the $j^{th}$ spin $S^A_{Aj}$ with nearest neighbors (NN) $S^0_{Aj+1}$, $J_2$ is the exchange constant between the $j^{th}$ spin $S^B_{Aj}$ with next nearest neighbors (NNN) $S^0_{Aj+2}$, here $\alpha = x, y, z$. Position of the $j^{th}$ spin are defined by vector $\vec{r}_j = x_j \vec{e}_x$.

![Figure 1. The zigzag spin chain with the $J_1 - J_2$ model.](image)

I divide the system into sub-lattice $A$ and sub-lattice $B$. Hamiltonian of the system with applied magnetic field $\vec{h} \uparrow \uparrow Oz$ has form:

$$H = -g \mu_B h \sum_{j'} S^z_{Aj} - \frac{1}{2} \sum_{a,j} J_1 S^z_{Aj} S^z_{Bj+1} - \frac{1}{2} \sum_{a,j} J_2 S^z_{Aj} S^z_{Aj+2}$$

$$-g \mu_B h \sum_{j} S^z_{Bj} - \frac{1}{2} \sum_{a,j} J_1 S^z_{Bj} S^z_{Aj+1} - \frac{1}{2} \sum_{a,j} J_2 S^z_{Bj} S^z_{Aj+2}$$

$$-\frac{1}{2} D \sum_j \langle S^z_{Aj} \rangle^2 - \frac{1}{2} D \sum_j \langle S^z_{Bj} \rangle^2,$$

(1)

When considering to the spin fluctuations I rewrite (1) using the Fourier transform for the spin operators as following:

$$H = H_0 + H_{\text{int}},$$

(2)

with

$$H_0 = - \sum_j (g \mu_B h + J_1 (k_x = 0) \langle S^z_B \rangle + J_2 (k_x = 0) \langle S^z_A \rangle + D \langle S^z_A \rangle) S^z_{Aj}$$

$$- \sum_j (g \mu_B h + J_1 (k_x = 0) \langle S^z_A \rangle + J_2 (k_x = 0) \langle S^z_B \rangle + D \langle S^z_B \rangle) S^z_{Bj} +$$

$$N J_1 (k_x = 0) \langle S^z_A \rangle \langle S^z_B \rangle + \frac{N}{2} [J_2 (k_x = 0) + D] \langle S^z_A \rangle \langle S^z_A \rangle + \frac{N}{2} [J_2 (k_x = 0) + D] \langle S^z_B \rangle \langle S^z_B \rangle$$

(3)
Consider with constants:

\[ H = J_1 (k_x) \delta S^\alpha_A (k_x) \delta S^\beta_B (-k_x) + J_1 (k_x) \delta S^\beta_B (k_x) \delta S^\alpha_A (-k_x) + [J_2 (k_x) + D \delta_{\alpha,z}] \delta S^\alpha_A (k_x) \delta S^\alpha_A (-k_x) + [J_2 (k_x) + D \delta_{\alpha,z}] \delta S^\alpha_B (k_x) \delta S^\alpha_B (-k_x) \]  

(4)

Rewriting \( H_{\text{int}} \) in form:

\[ H_{\text{int}} = -\frac{1}{2} \sum_{\alpha,k_x} \sum_{n,n'} J_{nn'} (k_x) \delta S^\alpha_n (k_x) \delta S^\alpha_{n'} (-k_x), \]  

(5)

here

\[ J = \begin{bmatrix} J_{AA} (k_x) & J_{AB} (k_x) \\ J_{BA} (k_x) & J_{BB} (k_x) \end{bmatrix} = \begin{bmatrix} J_2 (k_x) + D \delta_{\alpha,z} & J_1 (k_x) \\ J_1 (k_x) & J_2 (k_x) + D \delta_{\alpha,z} \end{bmatrix}, \]  

(6)

with

\[ J_{AB} (k_x) = J_1 (k_x) = J_{BA} (k_x), J_{AA} (k_x) = J_2 (k_x) + D \delta_{\alpha,z} = J_{BB} (k_x), \]  

\( n, n' = A, B \) and \( J_{nn'} (k_x) \) is the component of the \( 2 \times 2 \) matrix of the exchange interaction constants:

\[ J_1 (k_x) = J_1 (\exp (ik_x a) + \exp (-ik_x a)) = 2J_1 \cos (k_x a), \]

\[ J_2 (k_x) = J_2 (\exp (ik_x 2a) + \exp (-ik_x 2a)) = 2J_2 \cos (k_x 2a), \]  

(7)

with \( a = |x_j - x_{j+1}|. \)

The total fields act on the spins:

\[ y_A = \beta g \mu_B h + 2 \beta J_1 \langle S_B^z \rangle + (2 \beta J_2 + \beta D) \langle S_A^z \rangle, \]

\[ y_B = \beta g \mu_B h + 2 \beta J_1 \langle S_A^z \rangle + (2 \beta J_2 + \beta D) \langle S_B^z \rangle. \]  

(8)

Consider

\[ \exp (-\beta H) = \exp (-\beta H_0) \exp (-\beta H_{\text{int}}) = \exp (-\beta H_0) \tilde{T} \exp \left\{ -\int_0^\beta H_{\text{int}} (\tau) d\tau \right\} \]

\[ = \exp (-\beta H_0) \times \int (d\varphi) \tilde{T} \exp \left\{ \sum_{n,\alpha, \bar{q}} \left( \frac{-1}{2} \varphi_n^\alpha (\bar{q}) \varphi_n^\alpha (-\bar{q}) + \sum_{n'} (\beta J_{nn'} (k_x))^{1/2} \varphi_n^\alpha (\bar{q}) \delta S^\alpha_{n'} (-\bar{q}) \right) \right\}, \]  

(9)

with \( \bar{q} \) is two-component wave vector \( \bar{q} = (k_x, \omega) \), \( \delta S^\alpha_n (\bar{q}) = \beta^{-1} \int_0^\beta e^{i\tau \omega} \delta S^\alpha_n (k_x, \tau) d\tau \) and

\[ \int (d\varphi) = \prod_{\alpha, \bar{q}, -\infty}^{+\infty} \int \frac{d\varphi_n^\alpha (\bar{q})}{\sqrt{2\pi}} = 0 \prod_{k_x \neq 0, -\infty}^{+\infty} \int \frac{d\varphi_n^{\alpha,c} (\bar{q})}{\sqrt{\pi}} \int_0^\infty \frac{d\varphi_n^{\alpha,s} (\bar{q})}{\sqrt{\pi}}, \]  

(10)

here \( \varphi_n^{\alpha,c} (\bar{q}) \) and \( \varphi_n^{\alpha,s} (\bar{q}) \) are real part and image part of the field variable \( \varphi_n^\alpha (\bar{q}) \),

\[ \varphi_n^\alpha (\bar{q}) = \varphi_n^{\alpha,c} (\bar{q}) + i\varphi_n^{\alpha,s} (\bar{q}). \]  

(11)
From that I can find functional integral form for free energy:

\[ F = -\frac{1}{\beta} \ln \text{Tr} \left( e^{-\beta H} \right) = -\frac{1}{\beta} \ln \text{Tr} \left( e^{-\beta H_0} e^{-\beta H_{int}} \right) = F_0 - \frac{1}{\beta} \ln \left\langle e^{-\beta H_{int}} \right\rangle, \quad (12) \]

with

\[ F_0 = -\frac{1}{\beta} \ln \left( \text{Tr} e^{-\beta H_0} \right) = 2N J_1 \langle S_A^z \rangle \langle S_B^z \rangle + \frac{N}{2} (2J_2 + D) \sum_n \langle S_n^z \rangle \langle S_n^z \rangle - \frac{N}{\beta} \sum_n \ln \left( \frac{\text{sh}(S_n+1/2)y_n}{\text{sh}(y_n/2)} \right) \quad (13) \]

and

\[
\ln \left\langle e^{-\beta H_{int}} \right\rangle_0 = \ln \int (d\varphi) \exp \left\{ -\frac{1}{2} \sum_{n,\alpha,\vec{q}} \varphi_n^\alpha(\vec{q}) \varphi_n^{-\alpha}(\vec{q}) \right\} \\
\times \left\langle \hat{T} \exp \left\{ \sum_{n,n',\alpha,\vec{q}} \left( \beta J_{nn'}(k_x) \right) \varphi_n^\alpha(\vec{q}) \delta S_{n'}^\alpha(\vec{q}) \right\} \right\}_0. \quad (14)
\]

In the 1st order of the Gaussian approximation, we have:

\[
\ln \left\langle e^{-\beta H_{int}} \right\rangle_0 = \ln \int (d\varphi) \exp \left\{ -\frac{1}{2} \sum_{n,\alpha,\vec{q}} \varphi_n^\alpha(\vec{q}) \varphi_n^{-\alpha}(\vec{q}) \right\} \\
\times \exp \left\{ -\frac{1}{2} \sum_{n,n',k_x} \beta J_{nn'}^1(k_x) J_{nn'}^1(-k_x) y_n \varphi_n(k_x,0) \varphi_n^*(k_x,0) \right\} \\
\times \exp \left\{ -\frac{1}{2} \sum_{\alpha,\beta,\vec{q}} \delta S_{nn'}^\alpha(\vec{q}) \delta S_{nn'}^{-\alpha}(\vec{q}) \right\}. \quad (15)
\]

Calculating the functional integrals in (15), I receive the expression for the free energy:

\[
F = 2N J_1 \langle S_A^z \rangle \langle S_B^z \rangle + \frac{N}{2} (2J_2 + D) \sum_n \langle S_n^z \rangle \langle S_n^z \rangle - \frac{N}{\beta} \sum_n \ln \left( \frac{\text{sh}(S_n+1/2)y_n}{\text{sh}(y_n/2)} \right) \\
+ \frac{1}{2\beta} \sum_{k_x,\omega} \ln \left\{ \left( 1 - \beta J_2(k_x) \right) b'(y_A) \left( 1 - \beta J_2(k_x) + D \right) b'(y_B) \right\} \\
+ \frac{1}{2\beta} \sum_{k_x,\omega} \ln \left\{ \left( 1 - \beta J_2(k_x) b(y_A) \right) \left( 1 - \beta J_2(k_x) b(y_B) \right) \text{cth} \frac{y_{A,B}}{2} \right\} \quad (17)
\]

with \( b(y_{A,B}) \) and \( b^{(m)}(y_{A,B}) \) is Brillouin function and its \( m \)th order derivative:

\[
b(y_{A,B}) = (S_{A,B} + \frac{1}{2}) \text{cth} (S_{A,B} + \frac{1}{2}) y_{A,B} - \frac{1}{2} \text{cth} \frac{y_{A,B}}{2}.
\]

In ordered phase, mean spin moment per site of the system can be found from the free energy (17):

\[
m_{zA} = \langle S_A^z \rangle = \frac{1}{N} \frac{\partial F}{\partial h_A} = \frac{1}{N} \frac{\partial y_A}{\partial h_A} \frac{\partial F}{\partial y_A}, \\
m_{zB} = \langle S_B^z \rangle = \frac{1}{N} \frac{\partial F}{\partial h_B} = \frac{1}{N} \frac{\partial y_B}{\partial h_B} \frac{\partial F}{\partial y_B}, \\
m_z = m_{zA} + m_{zB}.
\]
To investigate the spectra of the excitations of the system, I give transverse correlation functions between the fluctuations of the spin components (or time-ordered spin Green functions):

$$\chi^{\pm}_{n,j_1,n',j_2}(\tau_1,\tau_2) = \chi^{\pm}_{n,n'}(x_{j_1} - x_{j_2}, \tau_1 - \tau_2) = \langle \hat{T} S^{-}_{n,j_1}(\tau_1) S^{+}_{n',j_2}(\tau_2) \rangle.$$  \hspace{1cm} (18)

The Fourier transformation of the transverse correlation functions are given as following:

$$\tilde{\chi}_{AB}^{\pm}(k_\omega) = \frac{1}{2} \beta J_2(k_\omega) \begin{bmatrix}
 b(y_A) b(y_B) (\beta J_1(k_\omega))^3 + \\
 - (\beta J_2(k_\omega))^2 b(y_A) b(y_B)
\end{bmatrix} \begin{bmatrix}
 (\beta J_1(k_\omega))^2 - (\beta J_2(k_\omega))^2 \\
 z^2 - (y_A + y_B + \beta J_2(k_\omega) (b(y_A) + b(y_B))) z + y_A y_B
\end{bmatrix}, \hspace{1cm} (19)
$$

here $z = i \beta \omega$.

The excitation spectra can be received from poles of the transverse correlation function (19), the results show that the system has two distinct excitations:

$$\epsilon_{k_\omega} = \frac{y_A}{2} + \frac{y_B}{2} - \frac{\beta J_2(k_\omega) b(y_A) - \beta J_2(k_\omega) b(y_B)}{2} \pm \left( 4(\beta J_1(k_\omega))^2 b(y_A) b(y_B) + (\beta J_2(k_\omega))^2 (b(y_A)^2 - 2b(y_A) b(y_B) + b(y_B)^2) \right)^{1/2}$$

$$\hspace{5cm} + \frac{-2\beta J_2(k_\omega) (b(y_A) y_A - b(y_A) y_B - b(y_B) y_A + b(y_B) y_B + y_A^2 - 2y_A y_B + y_B^2)}{2}.$$

3. Numerical results and discussion

In this section, I present the thermodynamic quantities in unit of the exchange interaction $J$, here $J_1 = a J$, $J_2 = b J$ and $D = c J$. Thus I have the external magnetic field of $\frac{g \mu_B h}{J}$, the susceptibility of $\frac{\chi^J}{N(g \mu_B)^2}$, the temperature of $\frac{\kappa_B T}{N(g \mu_B)^2}$ and specific heat of $\frac{C}{N k_B}$.

At large positive value of the anisotropic parameter $D$ and without the magnetic field $g \mu_B h = 0$, the system is in a AFM Neel phase, with the spins aligned in the $z$ direction on the alternating sites $A$ and $B$ (see figure 2). When $g \mu_B h$ is very large, the system has a FM order, with all spins aligned in the direction of the applied field (see figure 3).

When $D \neq 0$ and $g \mu_B h \neq 0$, the system shows a spin-reorientation transition. Figure 4 and figure 5 point out the results on the temperature dependence of the susceptibility $\frac{\chi^J}{N(g \mu_B)^2}$ and the specific heat $\frac{C}{N k_B}$ at the various values of the exchange interaction parameters. There are two peaks in the curve of the susceptibility. Sharp peaks of $\frac{\chi^J}{N(g \mu_B)^2}$ and $\frac{C}{N k_B}$ show the reorientation of the spins $B$, tend to the direction of the applied field. Broad peaks in figure 4 show magnetic behavior of the system after the reorientation. The peaks in these figures moves to the higher $k_B T$ with the increase of $J_2$. This indicates that the spin fluctuations are suppressed by the NNN FM interaction. However, after the reorientation the order-disorder phase transition of the system is not observed due to the appearance of the external magnetic field (the broad peaks).

The total field which acts on the sub-lattice $B$ causes the magnetic moments of this sub-lattice are decreased as the applied field increases and approach zero. After that the magnetic moments align with the direction of the applied field and then reach the saturation value (see figure 6).
Figure 2. The temperature dependence of the magnetization at large positive value of $D$ and $g\mu_B J = 0$, here $a = -1.0$, $b = 0.6$, $c = 3.0$ and $S = 1$.

Figure 3. The temperature dependence of the magnetization when $g\mu_B h J$ is very large, here $a = -1.0$, $b = 0.1$, $c = 0.3$, $g\mu_B h J = 1.5$ and $S = 1/2$.

Figure 4. The temperature dependence of the susceptibility. The inset shows the temperature dependence of the magnetization when $D \neq 0$ and $g\mu_B h J \neq 0$.

Figure 5. The temperature dependence of the specific. The inset shows the temperature dependence of the magnetization when $D \neq 0$ and $g\mu_B h J \neq 0$.

Figure 7 shows the magnetic excitations of the system. The branches concerns combination of the two NN spins $J_1$, the two NNN spins $J_2$ and the anisotropy $D$. When $J_2 = 0$ and $D = 0$, the system is the uniform AFM Heisenberg chain having the gapless excitation. In the case of $J_1 < 0$, $J_2 > 0$ and $D = 0$, the state ground of the spin chain which is AFM, carries $S_z = 0$ ($... \uparrow\uparrow\downarrow\uparrow\downarrow ...$) and is non-degenerate. The excitation spectra in this case have no the energy gap from the ground state when changing $J_2$. According to me the results are logical, because when $J_2 > 0$ will support the arrangement $... \uparrow\uparrow ...$ of the sub-lattice A and $... \downarrow\downarrow ...$ of the sub-lattice B, the system is similar to the uniform AFM chain. When $D \neq 0$, the anisotropy causes appearance of the gapped excitations in the system. The energy gap is induced by the single-ion anisotropy due to the change of the spin ordering. In [12], using perturbation theory the authors also indicate that the gap increases with the anisotropic parameter $D$. 
4. Comparison with experimental result

In [11], the authors measured the magnetic susceptibility of Rb$_2$Cu$_2$Mo$_3$O$_{12}$ powder. Solodovnikov and Solodovnikova first synthesized Rb$_2$Cu$_2$Mo$_3$O$_{12}$ and determined its crystal structure [13]. According to [11], the localized spins exist only on Cu$^{2+}$ ions ($S = 1/2$). There are two crystallographic Cu sites. Slightly distorted chains formed by edge-shared CuO$_6$ octahedral parallel to the Ox axis correspond to $S = 1/2$ zig-zag chains (see the figure 1). The sign of $J_1$ cannot be determined from the crystal structure because both cases (AFM or FM) are allowed in such Cu-O-Cu angles.

In [11], the authors propounded that Rb$_2$Cu$_2$Mo$_3$O$_{12}$ could be described by a FM-AFM $J_1 > 0$ and $J_2 < 0$ model at first approximation. However, I find that the AFM-FM model with $J_1 < 0$ and $J_2 > 0$ of this paper gives the numerical results which quite fits the experimental data (see figure 8) when choosing the parameters $\frac{J_1}{k_B} = -15K$, $a = -1$, $b = 0$ and $c = 0.1$. Here I am unable to determine values of $J_1$ and $J_2$ uniquely when comparing the calculated result with the experimental result, so I only estimate the values for $a$, $b$ and $c$ from reasons as following:

- The value of the experimental susceptibility curve at starting point is not equal to zero ($\chi$(emu/Cu mol)$\approx 0.01$, see in the inset of the figure 8), which corresponds to a minimum point right below the sharp peak (the reorientation transition) of the theoretical curve at $h \neq 0$ and low temperature in my model (see more in the figure (4)). Therefore, the broad peak of the theoretical solid curve fits the peak of the experimental curve.

- Because the temperature at the starting point is very low ($2K < T < 10K$) and Rb$_2$Cu$_2$Mo$_3$O$_{12}$ is slightly distorted into a zigzag chain, I guess the spin fluctuations of the system are sufficient so that they don’t break the magnetic order in the system and the anisotropic interaction also becomes more important, so I choose the NN, NNN and anisotropic parameters suitably $\frac{J_1}{k_B} = -15K$, $a = -1$, $b = 0.1$ and $c = 0.1$. When increasing $b$ or $c$ (the dashed curve: $\frac{J_1}{k_B} = -15K$, $a = -1$, $b = 3$ and $c = 0.1$) which leads to decrease of the spin fluctuations, so the peak of the susceptibility move to the higher temperature region and the height of the broad peak is lowered.

5. Conclusion

In this paper, I studied the thermodynamic properties and the excitation spectra of the zigzag spin chain at the various values of the AFM NN ($J_1$) and the FM NNN ($J_2$) exchange interactions using the functional integral method. Increase of NNN $J_2$ and the singe ion anisotropy $D$ causes decrease of the spin fluctuations and correspondingly the magnetic order of the system increases.
Figure 8. The temperature dependence of the susceptibility (the solid curve: \(a=-1, b=0.1, c=0.1\); the dashed curve: \(a=-1, b=3, c=0.1\)): comparison with the experimental result for \(\text{Rb}_2\text{Cu}_2\text{Mo}_3\text{O}_{12}\) measured in [11], (the curve is numbered 3 in the inset).

The existence of the gapless and gapped excitations when changing the NNN and anisotropic parameters were discussed. The numerical results for the susceptibility were compared with the experimental data of \(\text{Rb}_2\text{Cu}_2\text{Mo}_3\text{O}_{12}\) powder when choosing the values of \(J_1\) and \(J_2\) fittingly. I also pointed out that the NN and NNN interactions and the spin fluctuations play important roles in the edge-sharing copper oxide chain.

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