Finite Temperature RVB Mean-field Analysis of the Anisotropic Triangular Lattice Spin System

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Abstract. An anomalous non-magnetic insulator phase called “gapless spin liquid state” has been found in various materials with geometrically frustrated lattices. Organic conductor \( \kappa \)-(ET)\(_2\)Cu\(_2\)(CN)\(_3\), which has quasi-two-dimensional triangular lattice, is one of them. Mott insulator phase of this material is known by several experiments to show neither sign of magnetic ordering nor spin excitation gap in low temperatures. To clarify the physical origin of this peculiar state, we perform resonating-valence-bond mean-field analysis of the spin-1/2 Heisenberg antiferromagnet on a triangular lattice with one-dimensional (1D) anisotropy. We take an approach from 1D spin chains assuming it is likely that the gapless behavior observed in experiments descends from 1D spin excitations. As a result of calculation at zero temperature, almost 1D gapless excitation spectra are obtained in the wide range from the 1D limit. We further extend our analysis to finite temperatures and find that the anisotropy parameter range expands in which the “one-dimensionalization” occurs. The gapless features characteristic of 1D spin systems are also found in the temperature dependences of several thermodynamical quantities.

Recent experimental realization of anomalous quantum spin liquid states\cite{1, 2, 3, 4, 5} renewed interests in magnetism of frustrated spin systems. Since the proposal of a resonating valence bond (R VB) spin liquid state\cite{6}, a two-dimensional triangular lattice has been considered as a candidate to have spin disordered ground state. As a result of intensive theoretical studies so far, the spin-1/2 Heisenberg antiferromagnet on a triangular lattice turns out to have three-sublattice long-range magnetic order (LRMO)\cite{7, 8, 9}. If one assumes a disordered ground state, the mean-field theory of R VB state gives a spin-gap state with \( d_{x^2-y^2} + \text{i}d_{xy} \)-wave symmetry\cite{10, 11}.

From the experimental point of view, there has been considerable recent interests in a quasi-two-dimensional organic conductor \( \kappa \)-(ET)\(_2\)Cu\(_2\)(CN)\(_3\). In low temperatures, this system can be regarded as a nearly isotropic triangular lattice spin system with \( S = 1/2 \). NMR and susceptibility measurements\cite{1, 12} have shown that \( \kappa \)-(ET)\(_2\)Cu\(_2\)(CN)\(_3\) exhibits neither LRMO nor spin excitation gap. If we disregard this material as an equilateral triangular lattice spin system, the “gapless spin liquid” phase cannot be explained by the theories presented above.

In order to elucidate the physical origin of this gapless spin liquid state, we paid attention to a small anisotropy which resides in \( \kappa \)-(ET)\(_2\)Cu\(_2\)(CN)\(_3\)\cite{13}. According to the extended Hückel evaluation, one of the three hopping integrals between ET dimers (units of our triangular lattice model) is larger than the others. It means that the triangular lattice has quasi-one-dimensional (Q1D) anisotropy. Considering that the pure 1D spin system has no LRMO and has gapless spin excitations, it is likely that this Q1D anisotropy is concerned with the formation of the gapless spin liquid state.
Above considerations in mind, we study the spin-1/2 Heisenberg antiferromagnet on an anisotropic triangular lattice, which is equivalent to 1D spin chains (interaction $J$) coupled with zigzag bonds (interaction $J'$) as shown in Fig. 1. The Hamiltonian is given by

$$
\mathcal{H} = \sum_{<i,i'>} J S_i \cdot S_{i'} + \sum_{<i,j>} J' S_i \cdot S_j,
$$

where $<i,i'>$ and $<i,j>$ denote intrachain and interchain nearest-neighbor pairs, respectively (see Fig. 1). We investigate the anisotropy parameter range $J'/J = 0.0 - 1.0$, in which the model interpolates between the decoupled chains ($J' = 0$) and the isotropic triangular lattice ($J' = J$).

We transform spin operators into fermion operators as $S_i = \frac{1}{2} c_{i\alpha}^\dagger \sigma_{\alpha\beta} c_{i\beta}$, where $\sigma_{\alpha\beta}$ ($\alpha, \beta = \uparrow$ or $\downarrow$) are Pauli matrices. By introducing mean fields $\xi_{\tau_i} \equiv \langle c_{i\uparrow}^\dagger c_{i+\tau_i \downarrow} \rangle \equiv \langle c_{i\uparrow}^\dagger c_{i+\tau_i \dagger} \rangle$ and $\Delta_{\tau_i} \equiv \langle c_{i\uparrow}^\dagger c_{i+\tau_i \downarrow} \rangle$, where $\tau$ denotes a lattice vector, we can obtain self-consistent equations on the analogy of BCS theory as

$$
\begin{cases}
\xi_{\tau_i} = -\frac{1}{2N} \sum_k e^{ik \cdot \tau_i} \frac{\xi_k}{E_k} \tanh \frac{\beta E_k}{2}, \\
\Delta_{\tau_i} = \frac{1}{2N} \sum_k e^{-ik \cdot \tau_i} \frac{\Delta_k}{E_k} \tanh \frac{\beta E_k}{2},
\end{cases}
$$

where $\beta \equiv 1/T$ (we set $k_B = 1$) and $E_k = \sqrt{\xi_k^2 + |\Delta_k|^2}$ denotes a quasiparticle excitation spectrum. $\xi_k$ and $\Delta_k$ are given by

$$
\begin{cases}
\xi_k \equiv -3J \xi_{\tau_1} \cos(k \cdot \tau_1) - 3J' \xi_{\tau_2} \cos(k \cdot \tau_2) + \xi_{\tau_3} \cos(k \cdot \tau_3), \\
\Delta_k \equiv 3J \Delta_{\tau_1} \cos(k \cdot \tau_1) + 3J' \Delta_{\tau_2} \cos(k \cdot \tau_2) + \Delta_{\tau_3} \cos(k \cdot \tau_3),
\end{cases}
$$

and $\tau_1 = (1,0)$, $\tau_2 = (1/2,\sqrt{3}/2)$, $\tau_3 = (1/2,-\sqrt{3}/2)$ as shown in Fig. 1. We determine the mean-fields $\Delta_{\tau_1}$, $\xi_{\tau_i}$ ($i = 1, 2, 3$) as a function of temperature $T$, by solving self-consistent equations (2) numerically.

First, we define two order parameters $D_1 \equiv \sqrt{\xi_{\tau_1}^2 + |\Delta_{\tau_1}|^2}$ and $D_{23} \equiv \sqrt{\xi_{\tau_2}^2 + |\Delta_{\tau_2}|^2} = \sqrt{\xi_{\tau_3}^2 + |\Delta_{\tau_3}|^2}$. These determine the dispersion relations along the chains ($\tau_1$) and between the chains ($\tau_2, \tau_3$), respectively. These are the only parameters determined uniquely regardless of

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{Figure1.png}
\caption{Anisotropic triangular lattice with $J' < J$.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{Figure2.png}
\caption{Temperature dependence of the order parameters $D_1$ and $D_{23}$ for $J'/J = 0.2, 0.6$.}
\end{figure}
the SU(2) degeneracy at half-filling[10]. At zero temperature, we have already found that $D_{23}$ becomes almost zero, in spite of the comparatively large $J'$ up to $J'/J \sim 0.25$[13]. This means that practically 1D state is realized in the parameter range $0 < J'/J \lesssim 0.25$, since the system reduces to a set of pure 1D chains when $D_{23} = 0$. We consider this “one-dimensionalization” occurs owing to the frustrated couplings between 1D chains.

The temperature dependences of $D_1$ and $D_{23}$ calculated for two anisotropy values $J'/J = 0.2$ and 0.6 are shown in Fig. 2. When $J'/J = 0.2$, $D_{23}$ keeps zero for all temperature range, and $D_1$ decreases monotonically to zero at a finite temperature $T_c^1$. This temperature dependence of $D_1$ is the same as that of the BCS gap, and its critical temperature can be calculated analytically as $T_c^1 = 3J/8$. In temperatures above $T_c^1$, both order parameters vanishes and the system becomes paramagnetic. Precisely speaking, there is a finite $D_{23}$ at zero temperature in the thermodynamic limit and should vanish at a finite temperature $T_{c}^{23}$. However, the value of $D_{23}$ at zero temperature is exponentially small (about 4 orders of magnitude smaller than $J$), and so $T_{c}^{23}$ is inevitably very small. On the other hand, $D_{23}$ of $J'/J = 0.6$ is found to vanish at a temperature $T_{c}^{23} \sim 0.08J$. Above $T_{c}^{23}$, only $D_1$ survives and it traces perfectly the same curve as that of $J'/J = 0.2$ case. As mentioned above, $D_{23} = 0$ means the realization of pure 1D state, and thus the 1D-originated behavior of various physical quantities are expected in the temperature range $T_{c}^{23} < T < T_c^1$.

![Figure 3](image)

**Figure 3.** The temperature dependence of the specific heat $C$ (left panel) and static susceptibility $\chi$ (right panel) for $J'/J = 0.2$ and 0.6. The low temperature behavior of $C$ up to $T/J = 0.1$ are expanded in the inset of the left panel. The solid line in the right panel represents a curve obeying Curie’s law fitted as $\chi = 0.5/T$.

Using the mean-fields and quasiparticle excitation spectrum $E_k$ determined by the self-consistent equations (2), we calculate the temperature dependences of the specific heat and static susceptibility defined as

$$C = -2k_B T \frac{\partial}{\partial T} \sum_k \left[ f(E_k) \ln f(E_k) + \left( 1 - f(E_k) \right) \ln \left( 1 - f(E_k) \right) \right],$$

$$\chi = -2\mu_B^2 \frac{1}{N} \sum_k \frac{\partial f(E_k)}{\partial E_k},$$

where $f(E) \equiv 1/(e^{\beta E} + 1)$ represents Fermi distribution function. The results are shown in Fig. 3. When $J'/J = 0.2$, only one transition at $T = T_c^1$ can be found. In high temperatures above $T_c^1$, the specific heat $C$ keeps zero since the entropy becomes constant, and the susceptibility $\chi$ obeys the Curie’s law $\chi \propto 1/T$ in this region, reflecting a paramagnetic character. Note
that in low temperatures, $C$ decreases linearly (see the inset in the left panel of Fig. 3), and $\chi$ remains finite toward zero temperature. These indicate the existence of gapless spin excitations, which is consistent with the realization of pure 1D state ($D_{23} = 0$). Actually, the temperature dependence of $\chi$ for $J'/J = 0.2$ is qualitatively agree with that of an exact behavior observed in a 1D spin system[14]. Its quantitative disagreement is considered to be due to the strong quantum fluctuation in 1D system. In the case of $J'/J = 0.6$, we can see two clear transitions at $T = T_c^2$ and $T_{23}^c$, corresponding to the emergence of intrachain and interchain order parameters, respectively. In low temperatures below $T_{23}^c$, both quantities decays exponentially, indicating spin-gap opening. We should notice that the behavior of $C$ and $\chi$ above $T_{23}^c$ for $J'/J = 0.6$ are identical with those for $J'/J = 0.2$. This means that, despite of the large spin-gap in the ground state, the physical quantities behave as if the 1D state is realized when $T > T_{23}^c$. Interpreting this from a different point of view, we can say that the anisotropy parameter $(J'/J)$ range in which “one-dimensionalization” occurs expands as the temperature increases.

We would like to discuss here the relation to $\kappa$-$(ET)_2Cu_2(CN)_3$. As mentioned at the beginning of this letter, the static susceptibility measurements suggest that it remains finite in temperatures as low as 2 orders of magnitude smaller than the coupling $J$. Furthermore, low-temperature specific heat proportional to the temperature is found in a recent experiment[15]. These low temperature behavior can be explained within our one-dimensionalization picture. However, in the nearly isotropic regime, for example $J'/J = 0.9$, relatively large spin-gap exists and $D_{23}$ does not vanish in low temperatures. This difficulty may not be overcome in the reach of the RVB mean-field approximation, and other sophisticated methods which can treat one-dimensional fluctuation will be needed.

In conclusion, we analyzed a triangular lattice Heisenberg antiferromagnet with Q1D anisotropy by use of RVB mean-field approximation in order to reveal the physical origin of the gapless spin liquid state observed in $\kappa$-$(ET)_2Cu_2(CN)_3$. As a result of finite-temperature calculations for $J'/J = 0.2$, typical second-order transition with the intrachain order parameter $D_1$ is found. Since the interchain order parameter $D_{23}$ is zero for whole temperature range, linear decrease of the specific heat $C$ and finite residue of the susceptibility $\chi$ are found in low temperatures, as evidences to support the realization of pure 1D state. In case of $J'/J = 0.6$, we found clear two-step transitions corresponding to the intrachain and interchain order parameters. Above $T_{23}^c$ where the interchain order parameter $D_{23}$ vanishes, $C$ and $\chi$ behave completely the same way as those of $J'/J = 0.2$. In other words, the “one-dimensionalization” can be seen in finite temperatures, even if the anisotropy parameter $J'/J$ becomes comparatively large.

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