Gapped state in three-leg $S = \frac{1}{2}$ Heisenberg tube

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We study the ground state of three-leg $S = \frac{1}{2}$ Heisenberg tube using the density-matrix renormalization group method. The dimerization order-parameter and spin-excitation gap are calculated in a wide range of leg exchange interactions. We confirm that a gapped state with the dimerization order is realized even when the leg exchange interactions are ferromagnetic. Furthermore, the topological configuration of spin-singlet pairs in the ordered state is determined from the results of the Berry phase of each spin coupling as well as the structure factor of singlet-singlet correlation functions. We find that there exist three kinds of configurations of the valence-bond states depending on the ratio of leg and rung exchange interactions.

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Odd-leg spin ladder belongs to the same universality class as single chain; thus, the ground state is precluded as a gapless spin-liquid (or a Tomonaga-Luttinger liquid). However, if the periodic boundary conditions are applied in the rung direction, i.e., a tube is shaped, the spin states are dramatically changed. It is due to the occurrence of a geometric property called frustration, which is today a hot topic in condensed matter physics. In general, it would appear that the fundamental low-energy physics of any odd-leg spin tube is essentially epitomized by that of three-leg one. So far, it has been recognized that, as long as all the exchange interactions are antiferromagnetic, the three-leg spin tube can be spontaneously dimerized to avoid (or to reduce) the frustration and the spin excitations are gapped.

An ideal nanotubular material with odd number of legs is vanadium oxide Na$_2$V$_3$O$_7$, which may be regarded as a $S = \frac{1}{2}$ nine-leg Heisenberg spin tube system. In experiments (Ref. 2), the $^{23}$Na NMR response, the dc- and ac-magnetic susceptibilities, and the specific heat reveal that above 100 K the system is considered as paramagnetic; whereas, below 100 K most of the localized V magnetic moments ($S = \frac{1}{2}$) form a collection of spin-singlet dimers with gaps $\Delta \sim 0 - 350$ K and the remaining small fraction of them forms spin-triplet bound states with gaps $\Delta \sim 0 - 15$ K; and, the degeneracy of the triplet ground states is lifted by a phase transition at 0.086 K. The mechanism of the gap opening is still open issue. Besides, this material has attracted considerable attention from a standpoint of entanglement in spin systems. Moreover, it is of great interest to seek a relevance to a spin-liquid state observed in a three-leg $S = \frac{1}{2}$ spin tube system CsCrF$_3$.

The low-energy spin Hamiltonian of Na$_2$V$_3$O$_7$ has been proposed by some theoretical groups, nevertheless, it is still controversial. Both the rung ($J_\perp$) and leg ($J_{||}$) exchange interactions seem to be very sensitive because the estimated values are quite different, even in ferro- and antiferromagnetic characteristics, depending on the approaches. An overview of the results are as follows: (i) The ab initio microscopic analysis both $J_\perp$ and $J_{||}$ are ferromagnetic, but $J_{||}$ is frustrated and the magnitude is much smaller than $J_\perp$; (ii) the first-principle calculations both $J_\perp$ is antiferromagnetic, while $J_{||}$ is ferromagnetic, and they have the same order of magnitude; and, (iii) the first-principles density functional theory the $J_{||}$ is ferromagnetic and $J_\perp$ is ferro- or antiferromagnetic. Possibly, the point to be grasped next is whether the dimerization order with finite spin gap can occur when ferromagnetic exchange interactions are contained in the odd-leg spin tube.

We thus consider the ground state and low-lying excited states of three-leg $S = \frac{1}{2}$ Heisenberg spin tube and provide new insights especially for the case that the ferromagnetic exchange interactions are taken into account. The Hamiltonian is given by

$$H = J_\parallel \sum_{\alpha=1}^{3} \sum_{i=1}^{L} \vec{S}_{\alpha,i} \cdot \vec{S}_{\alpha,i+1} + J_\perp \sum_{\alpha(\neq \alpha')}^{3} \sum_{i=1}^{L} \vec{S}_{\alpha,i} \cdot \vec{S}_{\alpha',i}$$

where $\vec{S}_{\alpha,i}$ is a spin-$\frac{1}{2}$ operator at rung $i$ and leg $\alpha$. We here restrict the rung interactions $J_\perp$ to be antiferromagnetic because it is obvious that no dimerization order occurs with ferromagnetic rung interactions. The leg interactions $J_{||}$ are varied from ferro- to antiferromagnetic range. Although several theoretical studies have been carried out on this and similar models, only antiferromagnetic interactions are considered.

In this paper, we calculate the dimerization order-parameter and the spin-excitation gap to clarify in which range of the exchange interactions the ordered state is realized. Also, the Berry phase of each coupling and the structure factor of singlet-singlet correlation functions are calculated to check topological configuration of the spin-singlet pairs in the ordered state. For those calculations, the density-matrix renormalization group (DMRG) technique is applied. We investigate tubes with several kinds of length up to $L = 312$, i.e., $312 \times 3$ cluster, under
the open boundary conditions (OBC) in the leg direction, unless otherwise stated. The density-matrix eigenstates up to \( m = 2400 \) are kept in the renormalization procedure and all quantities are extrapolated to the limit \( m \to \infty \). In this way, the discarded weight is less than \( 1 \times 10^{-7} \), while the maximum error in the ground-state energy is less than \( 10^{-7} - 10^{-6} \).

Let us first evaluate the dimerization order-parameter in a wide range of \( J_\perp/J_\parallel \) for exploring the presence or absence of long-range dimerized state. The order parameter is featured by an alternation of nearest-neighbor spin-spin correlations, \( S(i) = -\langle \vec{S}_{\alpha,i} \cdot \vec{S}_{\alpha,i+1} \rangle \), where \( \langle \cdots \rangle \) denotes the ground-state expectation value. As the OBC breaks translational symmetry in our calculation, the dimerized state is observed as a Friedel oscillation. Generally, the amplitude of the Friedel oscillation at system center decays as a function of system size. In case that it persists for arbitrarily large system size, we can judge that a long-range order exists. Thus, the dimerization order-parameter is defined as

\[
D = \lim_{L \to \infty} |S(L/2) - S(L/2 + 1)|.
\]

Nonzero value of \( D \) indicates the presence of long-range dimerization order in our model. In Fig. 1 (a), the results of \( D \) are shown as a function of \( J_\perp/J_\parallel \). We find that the dimerization order appears over the entire range of \( J_\perp/J_\parallel (\neq 0) \). For both positive and negative \( J_\perp/J_\parallel \) values, \( D \) increases rapidly at \( J_\perp/J_\parallel \lesssim 10 \) and keeps almost constant at \( J_\perp \gtrsim 10 \); in the limit of \( J_\perp/J_\parallel \to \infty \), it is saturated to \( D \approx 0.0673 \) (0.0183). However, quite different behaviors are seen if we take a closer look at small \( J_\perp/J_\parallel \) regime [see Fig. 1 (b)]. At \( J_\perp/J_\parallel > 0 \), \( D \) is discontinuously enhanced when \( J_\perp \) is switched on, then goes through a minimum around \( J_\perp = 0.1 \), and increases almost linearly from \( J_\perp \approx 0.2 \) to \( 5 \); while at \( J_\perp/J_\parallel < 0 \), \( D \) increases gradually with increasing \( J_\perp \), like \( D \sim \exp[-\mathcal{O}(1/J_\perp)] \).

The dimerization order implies a complete occupation of the system with spin-singlet pairs. Which poses a question on the topological configuration of the pairs. In order to solve it, we investigate the quantized Berry phase in the ordered state. The Berry phase is defined by

\[
\gamma = -i \int_0^{2\pi} A(\phi) d\phi,
\]

where \( A(\phi) \) is the Abelian Berry connection, \( A(\phi) = \langle \psi_\phi | \partial_\phi | \psi_\phi \rangle \) with the ground state \( |\psi_\phi \rangle \). The Berry phase is quantized as \( 0 \) or \( \pi \) (mod 2\( \pi \)) if the system has spin gap during the adiabatic continuation and time reversal symmetry; and “undefined” if a gapless excitation exists. We introduce a local perturbation by a twist of the nearest-neighbor connection, \( \tilde{S}_{\alpha,i} \cdot \tilde{S}_{\alpha',j} \to \frac{1}{2} (e^{-i\phi} \tilde{S}_{\alpha,i}^z \tilde{S}_{\alpha',j}^z + e^{i\phi} \tilde{S}_{\alpha,i}^- \tilde{S}_{\alpha',j}^-) + \tilde{S}_{\alpha,i}^z \tilde{S}_{\alpha',j}^- \). We here pick up a dimerized pair of triangles, i.e., including six spins, and evaluate the Berry phases of the leg bond (\( \gamma_{\text{leg}} \)) for \( \alpha = \alpha', j = i + 1 \) and of the rung bond (\( \gamma_{\text{rung}} \)) for \( \alpha \neq \alpha', j = i \). Note that the dimerized pair of triangles must include three spin-singlet pairs.

In Fig. 2 the \( J_\perp/J_\parallel \)-dependence of \( \gamma_{\text{leg}} \) and \( \gamma_{\text{rung}} \) are shown and the corresponding configurations are also schematically described. We find that there are three kinds of the spin-singlet configurations along \( J_\perp/J_\parallel \). Let us now see the case that \( J_\parallel \) is antiferromagnetic. A reconstruction of the valence bonds is seen at \( J_\perp/J_\parallel \approx 5 \). This would correspond to the crossover between constant-\( \Delta \) and proportional-\( \Delta \) regions around \( J_\perp/J_\parallel = 5 \). For \( J_\perp/J_\parallel \geq 5 \), the Berry phases of leg and rung bonds are denoted by \( \pi \) and 0, respectively. It means that all singlet pairs are formed on the leg bond [Fig. 2(iii)]. Whereas, for \( 0 < J_\perp/J_\parallel \lesssim 5 \), both \( \gamma_{\text{leg}} \) and \( \gamma_{\text{rung}} \) are denoted by 0. It may be interpreted if we assume that both the leg and rung bonds are involved to form spin-singlet pairs: one pair is formed in either one of three legs and the other four spins form a couple of pairs in the two rungs [Fig. 2(ii)]. This valence-bond state is threefold degenerate, so that the spin-singlet pairs would be not detected as local ones. Then, we turn to the case that \( J_\parallel \) is ferromagnetic. A valence-bond state is detected on the rung bond and a gapless excitation is found in the leg bond.
Interaction is denoted by triangles [Fig. 2(i)]. We call the last pair "diagonal spin-singlets" are formed on rungs of each triangle and the other one is between two sites on different legs and triangles [Fig. 2(i)]. We call the last pair "diagonal spin-singlets" and the effective antiferromagnetic exchange interaction is denoted by $J_3$ hereafter.

And now, of particular interest is the magnitude of spin-excitation gap especially in the ferromagnetic $J_\parallel$ case. It will be clarified by calculating an energy difference between the singlet ground state and the first triplet excited state,

$$\Delta = \lim_{L \to \infty} |E(L,1) - E(L,0)|, \quad (4)$$

where $E(L, S_z)$ is the ground-state energy of the system of length $L$ with the $z$-component of total spin $S_z$. We note that the system length must be taken as $L = 2l$, with $l (> 1)$ being an integer to maintain total spin of the ground state as $S = 0$. In Fig. 3 we show the results of $\Delta$ as a function of $J_\perp/J_\parallel$. We see that the gap opens except at $J_\perp = 0$, as anticipated from the results of dimerization order-parameter. For both $J_\parallel > 0$ and $J_\perp < 0$, roughly speaking, $\Delta$ starts to increase proportionally to $J_\perp$ and shift into almost constant for larger $J_\perp$. It means that the lowest singlet-triplet excitations for small and large $J_\perp/J_\parallel$ are scaled by distinct exchange interactions. When $J_3$ is antiferromagnetic, as suggested in our previous paper, $\Delta$ is approximately scaled by $J_\perp$ ($J_\parallel$) in the small (large) $J_\perp/J_\parallel$ regime. On the other hand, $\Delta$ seems to be always scaled by $J_3$ since the diagonal spin-singlet pair has the smallest binding energy when $J_\parallel < 0$. It may be interpreted as follows: in the perturbative sense, $J_3$ is proportional to $J_\perp$ ($|J_\parallel|$) for $J_\perp \ll |J_\parallel|$ ($|J_\parallel| \gg J_\parallel$); accordingly, $\Delta$ is scaled by $J_\perp$ ($|J_\parallel|$) in the small (large) $J_\perp/J_\parallel$ regime as in the case of antiferromagnetic $J_3$. In fact, $\Delta$ for $J_\perp/J_\parallel < 0$ is about a factor of 6 smaller than that for $J_\perp/J_\parallel > 0$.

As a consequence, we obtain $\Delta = 0.0437$ in the limit of $J_\perp/J_\parallel = -\infty$; $\Delta = 0.254$ in the limit of $J_\perp/J_\parallel = \infty$. We note that the $J_\perp$-dependence of $\Delta$ looks similar to that of $D$ but except when $J_\perp/J_\parallel$ is very small.

Lastly, in order to determine the periodicity of alignment of the valence-bond configurations, we calculate singlet-singlet correlation functions

$$\mathcal{S}(\vec{k}) = \frac{1}{3L} \sum_{\alpha \alpha', ij} \langle \mathcal{S}_{\alpha, i} \mathcal{S}_{\alpha', j} \rangle \exp[i\vec{k} \cdot (\vec{r}_{\alpha, i} - \vec{r}_{\alpha', j})] \quad (5)$$

with spin-singlet number operator

$$\mathcal{S}_{\alpha, i} = \frac{1}{4} - \vec{S}_{\alpha, i} \cdot \vec{S}_{\alpha', j}, \quad (6)$$

where $(\alpha', j) = (\alpha, i + 1)$, $(\alpha + 1, i)$, and $(\alpha + 1, i + 1)$ are chosen for the leg-leg (LL), rung-rung (RR), and diagonal-diagonal (DD) correlations, respectively. Here, the system size is fixed at $L = 24$ using the periodic boundary conditions. Thus, relations $\alpha + 3 \equiv \alpha$ and $i + 24 \equiv i$ are fulfilled.

In Fig. 4 the correlation functions $\mathcal{S}(\vec{k})$ at (a) $J_3 = 1, J_\perp = 1$, (b) $J_3 = 1, J_\perp = 10$, and (c) $J_3 = -1, J_\perp = 10$ for $L = 24$ are shown [the deviation from those for $L = 12$ has been confirmed to be negligibly-small]. We initially point out that large values of $\mathcal{S}(\pi, \pi)$ in the LL and of $\mathcal{S}(0, 0)$ in the RR correlations jointly indicate a straight line of the state (ii) in Fig. 2 which is described as alignment (II) in Fig. 3 whereas, large value of $\mathcal{S}(\pi, 0)$ in the LL correlation suggests a link
results of the Berry phase. As alignment (III) must be less dominant considering the agreements (II) and (III) coexist [see Fig. 4(a)]; however, the alignment of state (iii), which corresponds to alignment (III). For $J_\parallel = 1, J_\perp = 1$, therefore, it would appear that the alignments (II) and (III) coexist [see Fig. 1(a)]; however, the alignment (III) must be less dominant considering the results of the Berry phase. As $J_\perp$ increases, in the LL correlation $\mathcal{S}(\pi, \frac{3}{2}\pi)$ is rapidly diminished and $\mathcal{S}(\pi, 0)$ is rather enhanced [see Fig. 1(b)]; as a result, the alignment (III) becomes dominant for large $J_\perp/J_\parallel$ region. Then, we turn to the case of $J_\parallel < 0$ [see Fig. 1(c)]. As expected, $\mathcal{S}(\tilde{k})$ in the LL correlation is always zero indicating no spin-singlet pair in the leg bond; instead, the DD correlations are significant. The enhancements of $\mathcal{S}(\pi, \frac{3}{2}\pi)$ in the RR and $\mathcal{S}(\pi, \frac{5}{2}\pi)$ in the DD correlations are seen. It indicates a straight alignment of the state (i), as shown in Fig. 3(1). We argue that a zigzag chain, which is denoted as a dotted line in Fig. 5 is dimerized to remove the frustration instead of being dimerized in the leg direction for antiferromagnetic $J_\parallel$.

In conclusion, we study the ground-state properties of three-leg $S = \frac{1}{2}$ Heisenberg tube with antiferromagnetic rung exchange interactions. Using the DMRG method, the dimerization order-parameter, spin-excitation gap, Berry phase, and structure factor of singlet-singlet correlation functions are calculated. We confirm that the dimerization order occurs for both ferro- and antiferromagnetic leg exchange interactions, and the spin excitations are always gapped. Also, we find that there are three kinds of phases according to topological configuration of the spin-singlet pairs: (I) diagonal-singlet regime for $J_\parallel/J_\perp < 0$, (II) rung-singlet regime for $0 < J_\perp/J_\parallel \lesssim 5$, and (III) leg-singlet regime for $J_\perp/J_\parallel \gtrsim 5$.

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