Spin gap of the three-leg $S = \frac{3}{2}$ Heisenberg tube

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The ground-state properties of the three-leg $S = \frac{3}{2}$ straight Heisenberg tube are studied using the density-matrix renormalization group method. We find that the spin-excitation gap associated with a spontaneous dimerization opens for the whole coupling regime, as seen in the three-leg $S = \frac{1}{2}$ straight Heisenberg tube. However, in contrast to the case of the $S = \frac{1}{2}$ straight tube, the gap increases very slowly with increasing the rung coupling, and its size is only a few percent or less of the leg exchange interaction in the weak- and intermediate-coupling regimes. We thus argue that, unless the rung coupling is substantially larger than the leg coupling, the gap may be quite hard to be observed experimentally. We also calculate the quantized Berry phase to show that there exist three kinds of valence-bond-solid states depending on the ratio of leg and rung couplings.

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I. INTRODUCTION

For a long time, the exotic phenomena that have emerged from geometrical frustration have been fascinating but challenging subjects of research in condensed matter physics.[1] The peculiar dilemma of frustrated systems generally comes from a highly degenerate ground state in the classical sense. Here, we know that to resolve it comes essentially back to how the degeneracy is removed or how the frustration is minimized by taking the quantum fluctuations into account. The simplest example for the spin frustration may be the 120° structure of an antiferromagnetic triangle. In the context of a triangular-lattice $S = \frac{1}{2}$ antiferromagnet a spin-liquid state was proposed by Anderson.[2] As a related issue, the nine-leg "straight" spin tube system $Na_2V_3O_7$ (Ref. 3) has attracted much attention in the last few years. One could say that odd-leg straight spin ladders belong to the same universality class as a single chain does; thus, the ground state is comprehended as a gapless spin liquid or a Tomonaga-Luttinger (TL) liquid.[4] However, if the periodic boundary conditions (PBCs) are applied in the rung direction, i.e., a straight tube is shaped, the spin states are dramatically changed due to spin frustration in the polygonal ring with an odd number of rungs. In this sense, the three-leg "twisted" tube compound $[(CuCl_2\ tachH)_3Cl]Cl_2$ (Ref. 5) is also an intriguing system because it is regarded as a simple $S = \frac{3}{2}$ spin chain and no frustration occurs,[6] which is in contrast to the straight tube system $Na_2V_3O_7$.

Quite recently the hexagonal compound $CsCrF_4$ (Ref. 7), which is an ideal three-leg straight spin tube system formed by Cr^{3+} ions, has been reexamined experimentally from the point of view of spin frustration.[8] Since the magnetic moment comes from the e_g^2 state of Cr^{3+} ion, the magnitude of spin on each site is $S = \frac{3}{2}$. By performing magnetic susceptibility, heat capacity $C(T)$, and electron spin resonance measurements, it was reported that no magnetic long-range order is observed down to $T = 1.3$ K. In particular, a gapless spin-liquid state (or a TL liquid state) is indicated from the finite T-linear component of $C(T)$; this result raises a more absorbing question because a gapped ground state is expected in an odd-leg spin-half-integer straight spin tube system.[9] The need for an investigation of odd-leg straight spin tube systems with $S = \frac{3}{2}$ is therefore obvious in order to figure out this puzzle.

II. MODEL AND METHOD

In this paper, we thus consider a three-leg $S = \frac{3}{2}$ straight Heisenberg tube. The Hamiltonian is given as

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

where $\vec{S}_{\alpha,i}$ is a spin-$\frac{3}{2}$ operator at leg $\alpha(= 1,2,3)$ and rung i. $J_{\parallel}$ and $J_{\perp}$ are the nearest-neighbor exchange interactions in the leg and rung directions, respectively (see Fig. 1). We take $J_{\parallel} = 1$ as the unit of energy hereafter. In order to investigate the ground-state properties of the system (1), the density-matrix renormalization group (DMRG) technique[10] is employed. As necessary, the PBCs or the open-end boundary conditions (OBCs) are chosen in the leg direction. Using the OBCs (PBCs), we study the tubes with several kinds of length $L = 12$ to 48 ($L = 8$ to 24) keeping $m = 1200$ to 2600 ($m = 1600$ to 3200) density-matrix eigenstates in the renormalization procedure; in this way, the typical truncation error, i.e., the discarded weight, is $2 \times 10^{-6} - 1 \times 10^{-5}$ ($3 \times 10^{-5} - 7 \times 10^{-5}$). We note that the system length must be even and is better to be kept in $L = 4l$ or $4l + 2$ with $l =$ an integer for systematic extrapolation of calculated quantities into the thermodynamic limit. Moreover, an extrapolation to $m \to \infty$ for each calculation is necessary because the DMRG trial wave function slowly approaches the exact one with increasing m due to the large degrees of freedom $\sim 4^{3L}$ and strong spin frustration in our system. All the calculated quantities in this paper are extrapolated to the limit $m \to \infty$; thus, for example, the maximum error in the ground-state energy is estimated to be less than 1×10^{-3}.

The first thing we think of when considering the system (1) might be the topological similarity to the other odd-leg half-integer-spin straight Heisenberg tubes. The simplest case,

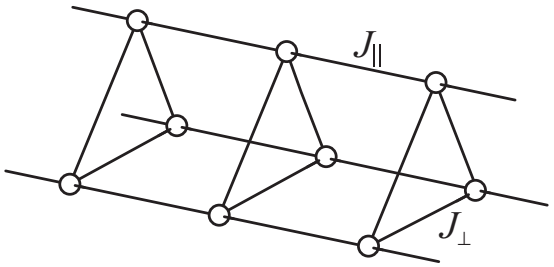

FIG. 1. Lattice structure of three-leg straight Heisenberg tube.

i.e., a three-leg $S = \frac{1}{2}$ straight Heisenberg tube, has been well studied, and the ground state is known to be gapped where the system is spontaneously dimerized in the leg direction to relax the intrarung spin frustration.[9,11–13] This can be naturally understood by analogy with the gap-opening mechanism in the one-dimensional (1D) $S = \frac{1}{2}$ spin-Peierls Heisenberg model.[14] Hence, in the case of $S = \frac{3}{2}$ as well, it would be best to start with a bond-alternated single chain problem, namely, the 1D $S = \frac{3}{2}$ spin-Peierls Heisenberg model. The Hamiltonian is written as $H = \sum_{i=1}^{L-1}[1-(-1)^i\delta]\vec{S}_i \cdot \vec{S}_{i+1}$ where $\vec{S}_i$ is a spin-$\frac{3}{2}$ operator at site i and $\delta(>0)$ is the strength of bond alternation. The low-energy physics of this system has been fundamentally elucidated:[15–17] Across the critical point $\delta \approx 0.42$, two kinds of valence-bond-solid (VBS) phases appear in the ground state; for the larger alternation ($\delta > 0.42$), the VBS state is essentially written as a direct product of "simple" spin-Peierls singlet bonds [Fig. 2(II)], whereas for the smaller alternation ($\delta < 0.42$), it is expressed as a combined state of the spin-Peierls singlet and $S = 1$ Haldane-like-gapped configurations [Fig. 2(I)], and the ground state is always gapped except at the critical point. Now, therefore, getting back to our system (1), if the spontaneous dimerization occurs as in the three-leg $S = \frac{1}{2}$ straight Heisenberg tube, it is likely that a gapped ground state is also obtained here. In fact, the 1D $S = \frac{3}{2}$ Heisenberg model with next-nearest-neighbor interactions, which is a similar model to our system (1), shows a spontaneous dimerized state when the frustration is strong.[18]

III. RESULTS

A. Dimerization order parameter

Then, we will simply evaluate a dimerization-order parameter to check the presence or absence of long-ranged spin-Peierls ordering in our system. Since a spin-Peierls state is characterized as an alternating formation of spin-singlet pairs in the leg direction, we focus on the nearest-neighbor spin-spin correlations,

$$S(i) = -\langle \vec{S}_{\alpha,i} \cdot \vec{S}_{\alpha,i+1} \rangle, \tag{2}$$

where $\langle \cdots \rangle$ denotes an expectation value in the ground state. Note that this quantity is independent of α. With applying the OBCs, the translational symmetry is broken due to the Friedel oscillation, and the spin-Peierls state is directly observable as a ground state. In general, the Friedel oscillation in the center

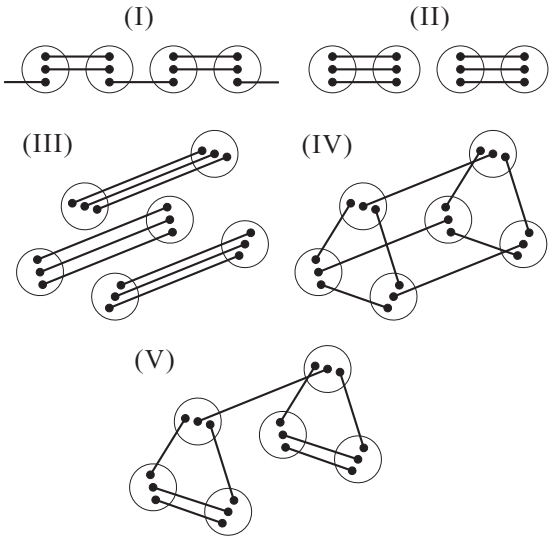

FIG. 2. Schematic pictures of the VBS configurations. Each small solid circle and connection with a line denote a spin-$\frac{1}{2}$ variable and a singlet pair, respectively. The large open circle represents a spin-$\frac{3}{2}$ operation that symmetrizes three spin-$\frac{1}{2}$ variables. The configurations (I) and (II) are VBS states in small and large bond-alternation regimes of the 1D $S = \frac{3}{2}$ Heisenberg model, respectively. The configurations (III)–(V) are possible candidates for VBS states in the three-leg $S = \frac{3}{2}$ straight Heisenberg tube (details are given in the text and Fig. 5).

of the system decays as a function of the system length. If the amplitude at the center of the system

$$D(L) = |S(L/2) - S(L/2+1)| \tag{3}$$

persists for an arbitrarily long system length, it corresponds to a long-range dimerization order, which indicates the spin-Peierls ground state. The order parameter is thus defined as an extrapolated value into the thermodynamic limit,

$$D = \lim_{L\to\infty} D(L). \tag{4}$$

In Fig. 3(a) we show the system-size dependence of the amplitude $D(L)$ for several $J_\perp$ values. We see that the values of $D(L)$ can be smoothly extrapolated to the thermodynamic limit $1/L \to 0$.

In Fig. 3(b) the extrapolated values D are plotted as a function of $J_\perp$. We see that it increases gradually at $J_\perp \lesssim 5$, almost linearly at $5 \lesssim J_\perp \lesssim 15$, and then goes into saturation at $D = 0.2764$ in the large $J_\perp$ limit. These different behaviors could be interpreted in terms of different VBS states for each $J_\perp$ regime, as in the $S = \frac{1}{2}$ straight Heisenberg tube.[12] This point will be clarified below by examining the Berry phase. A remaining question would be whether the order parameter remains finite for the small $J_\perp$ regime (we have not successfully obtained D for $J_\perp < 3$ due to its smallness). We find that the order parameter develops as $D = 0.25\exp(-13.2/J_\perp)$ from the single logarithmic plot (see the inset of Fig. 3); it implies that the order parameter is exponentially small but finite at $0 < J_\perp < 3$. Thus, we may expect a finite spin-excitation gap with dimerization order for the entire region of $J_\perp$. In this regard, it is worth noting that the behavior of D at small $J_\perp$ is quite different from that

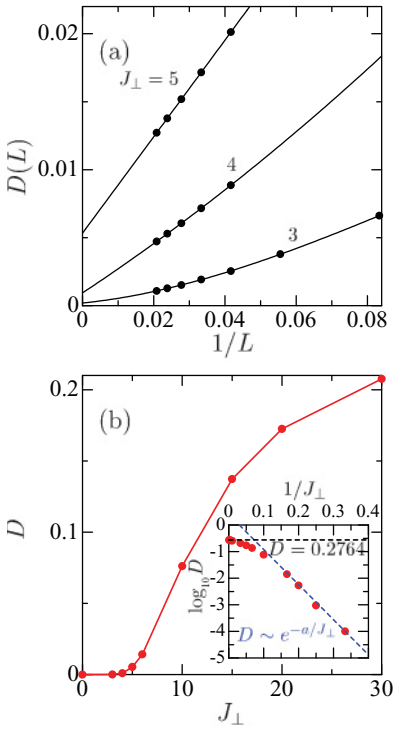

FIG. 3. (Color online) (a) Finite-size scaling of $D(L)$ as a function of $1/L$. The lines are the polynomial fittings. (b) Dimerization-order parameter D as a function of $J_\perp$. Inset: Semilog plot of D vs $1/J_\perp$. D saturates to 0.2764 in the large $J_\perp$ limit. The data for small $J_\perp$ are fitted by a function $D = \alpha \exp(-\beta/J_\perp)$ with $\alpha = 0.25$ and $\beta = 13.2$.

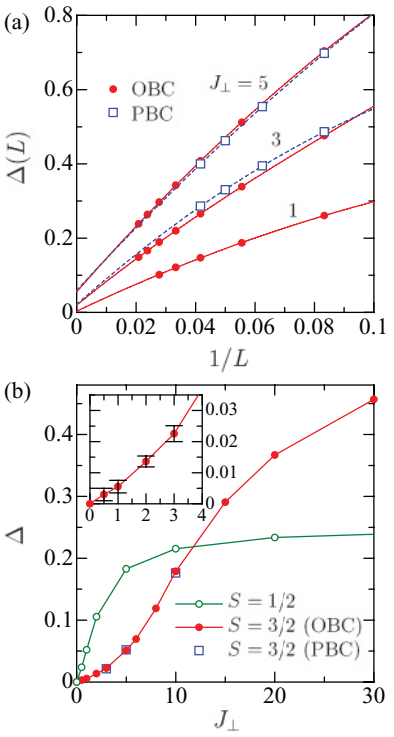

FIG. 4. (Color online) (a) Finite-size scaling of $\Delta(L)$ as a function of $1/L$. The lines are the polynomial fittings. (b) Extrapolated values of $\Delta(L)$ to the thermodynamic limit $1/L \to 0$. Inset: Extended figure for $0 \leqslant J_\perp \leqslant 4$. The error bars give differences between the second-order and cubic polynomial fittings for $\Delta(L)$.

in the case of the $S = \frac{1}{2}$ straight tube,[19] which would indicate a slower opening of the gap with increasing $J_\perp$ in our system.

B. Spin-excitation gap

Let us then estimate the spin-excitation gap. Of particular interest is the evolution of the gap onto the ratio between leg and rung couplings. The gap is defined as

$$\Delta = E_1(L) - E_0(L), \quad \Delta = \lim_{L\to\infty} \Delta(L), \tag{5}$$

where $E_0(L)$ and $E_1(L)$ are energies of the ground state ($S = 0$) and first triplet excited state ($S = 1$) for the system with length L, respectively. In Fig. 4(a) we plot the system-size dependence of the spin-excitation gap calculated with the OBCs in full circles. We see that the values of $\Delta(L)$ can be smoothly extrapolated to the thermodynamic limit $1/L \to 0$. The extrapolated values Δ, using a cubic polynomial extrapolation for $\Delta(L)$, are shown in Fig. 4(b) as a function of $J_\perp$. As expected, $J_\perp$ dependence of Δ looks rather similar to that of D; namely, it increases slowly at $J_\perp \lesssim 5$, rapidly at $5 \lesssim J_\perp \lesssim 15$, and then saturates in the strong-coupling limit $J_\perp = \infty$. This is because the spin-excitation gap is essentially equivalent to a binding energy of most weakly bounded spin-singlet pair in the system and it scales approximately with the dimerization strength. For comparison, we obtained $\Delta = 0.6661$ in the large $J_\perp$ limit, and it is in good agreement with a value $D = 0.624 \pm 0.024$ obtained from the exact-diagonalization analysis of a strong-coupling effective model within the first-order approximation.[20]

Here it is very instructive to compare the gap with that of the $S = \frac{1}{2}$ straight tube, which is also shown in Fig. 4(b). Two remarks are made about the comparison: (1) Although it may be rather natural, the gap in the strong-coupling limit seems to scale with the magnitude of spin, $\Delta(J_\perp \to \infty) \propto S$, and (2) the gap for $S = \frac{3}{2}$ increases much more slowly with increasing $J_\perp$ in the weak-coupling regime. As a result, only a few percent of the leg exchange interaction remains even at $J_\perp/J_\parallel = 5$. Hence, we argue that, unless the ratio $J_\perp/J_\parallel$ is sizably large, it may be difficult to detect the gap experimentally. For $CsCrF_4$, the leg exchange interaction is estimated to be a few 10 to 100 K by comparing the experimental peak position of magnetic susceptibility to numerical analysis for the 1D $S = \frac{3}{2}$ Heisenberg chain,[21] and the gap is only a few K at the outside even for $J_\perp/J_\parallel = 5$.

As described above, we obtain the gapped ground state for the whole $J_\perp$ region by applying the OBCs. It would mean that our system never contains the Haldane-type VBS state [Fig. 2(I)] in the three chains. This is because the gap cannot open due to free edge spins created by the OBCs if the state (I) is included. But to be sure, we shall confirm it by estimating the gap with the PBCs. The obtained results are shown with open squares in Fig. 4. We see that the extrapolated values are in good agreement with those with the OBCs, and it is confirmed that the state (I) does not exist at any VBS state in our system. Now, it is a fair question then to ask which kind

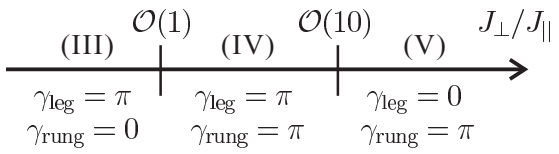

FIG. 5. Schematic "phase diagram" of the three-leg $S=\frac{3}{2}$ straight Heisenberg tube, classified by the Berry phases on the leg bond ($\gamma_{\rm leg}$) and rung bond ($\gamma_{\rm leg}$). The Roman numbers correspond to the VBS states shown in Fig. 2.

of VBS state is formed and how it changes with varying $J_\perp$, which we will discuss below.

C. Quantized Berry phase

Finally, we investigate the quantized Berry phase for determining the topological configuration of the VBS ground state. The Berry phase is defined by

$$\gamma = -i\int_0^{2\pi} A(\phi)d\phi, \tag{6}$$

where $A(\phi)$ is the Abelian Berry connection, $A(\phi)=\langle\psi_\phi|\partial_\phi\psi_\phi\rangle$ with the ground state $|\psi_\phi\rangle$.[22] The Berry phase is quantized as 0 or π (mod 2π) if the system has a spin gap during the adiabatic continuation and time-reversal symmetry, and "undefined" if a gapless excitation exists. We introduce a local perturbation by a local twist of the nearest-neighbor connection, $\vec{S}_{\alpha,i}\cdot\vec{S}_{\alpha',j}\to\frac{1}{2}(e^{-i\phi}S^+_{\alpha,i}S^-_{\alpha',j}+e^{i\phi}S^-_{\alpha,i}S^+_{\alpha',j})+S^z_{\alpha,i}S^z_{\alpha',j}$. In this paper a couple of clusters with $L=2$ and 4 are analyzed for this quantity. A dimerized pair of triangles, including six spins, from the clusters are picked up (it is the cluster itself for $L=2$), and the Berry phases of the leg bond ($\gamma_{\rm leg}$) for $\alpha=\alpha', j=i+1$ and of the rung bond ($\gamma_{\rm rung}$) for $\alpha\neq\alpha', j=i$ are evaluated. We call the spin-singlet pair on the leg (rung) bond "on-leg (on-rung) pair."

The Berry phases are obtained as follows: $(\gamma_{\rm leg},\gamma_{\rm rung})=(\pi,0)$ at $0<J_\perp<1$ $(0<J_\perp<0.5)$, $(\gamma_{\rm leg},\gamma_{\rm rung})=(\pi,\pi)$ at $1<J_\perp<15.3$ $(1<J_\perp<18)$, and $(\gamma_{\rm leg},\gamma_{\rm rung})=(0,\pi)$ at $J_\perp>15.3$ $(J_\perp>18)$ for $L=2$ $(L=4)$ cluster. Accordingly, we find three different VBS states depending on $J_\perp/J_\parallel$, as shown in Fig. 5. The boundary between VBS states is described by a crossover with recombination of spin-singlet bonds. In the large-coupling regime $J_\perp/J_\parallel>\mathcal{O}(10)$, we can easily imagine that the on-rung pair is more stable than the on-leg pair and as many pairs as possible prefer to be formed on the rung bond [Fig. 2(V)]. The spin gap therefore scales with the binding energy of an on-leg pair, i.e., $\Delta\propto J_\parallel$, which is consistent with the saturating behavior of Δ for $J_\perp\gg J_\parallel$. On the other hand, in the weak-coupling regime $J_\perp/J_\parallel<\mathcal{O}(1)$, all spin-singlet pairs are formed on the leg bond [Fig. 2(III)] because the binding energy of the on-leg pair is much larger than that of the on-rung pair. And, in the intermediate regime $\mathcal{O}(1)<J_\perp/J_\parallel<\mathcal{O}(10)$, the spin-singlet pairs seem to be distributed *in a balanced manner* [Fig. 2(IV)]. Here we notice an interesting relation to the VBS-state crossover in the $S=\frac{1}{2}$ straight tube.[19] If we ignore a spin-singlet pair on each rung in the states (IV) and (V) of our system, the remaining degrees of freedom are equivalent to those of the $S=\frac{1}{2}$ straight tube. As it turns out, the recombination of the remaining VBS bonds between (IV) and (V) can be essentially equivalent to the VBS-state crossover in the $S=\frac{1}{2}$ straight tube. Then the (ignored) extra degrees of freedom yields the additional state (III) in our $S=\frac{3}{2}$ system.

IV. CONCLUSIONS

We study the ground-state properties of the three-leg $S=\frac{3}{2}$ straight Heisenberg tube with the DMRG method. It is confirmed that a spontaneous dimerization occurs and the spin-excitation gap opens for the whole coupling region. This may be a common feature of odd-leg half-integer-spin straight Heisenberg tube systems. We find that the gap for $S=\frac{3}{2}$ increases very slowly with increasing $J_\perp$, and it remains very small compared with $J_\parallel$ in the weak- to intermediate-coupling regions. For $CsCrF_4$, the gap is estimated to be only a few K or less at normal pressures, and, for example, an additional condition such as applying pressure might be required to enlarge the ratio $J_\perp/J_\parallel$ in order to detect the gapped state. Moreover, by calculating the quantized Berry phase, it is shown that two VBS-state crossovers as recombination of spin-singlet bonds occur with varying the ratio $J_\perp/J_\parallel$, although further work is desirable for quantitative evaluation of the crossovers.

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