Dynamical structure factors of $S = 1/2$ two-leg spin ladder systems

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We investigate dynamical properties of $S = 1/2$ two-leg spin ladder systems. In a strong coupling region, an isolated mode appears in the lowest excited states, while in a weak coupling region, an isolated mode is reduced and the lowest excited states become a lower bound of the excitation continuum. We find in the system with equal intrachain and interchain couplings that due to a cyclic four-spin interaction, the distribution of the weights for the dynamical structure factor and characteristics of the lowest excited states are strongly influenced. The dynamical properties of two systems proposed for SrCu$_2$O$_3$ are also discussed.

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$S = 1/2$ two-leg spin ladder systems with antiferromagnetic interactions have attracted great attention both theoretically and experimentally [1]. Fascinating aspects of elementary excitation as well as thermodynamic properties have been revealed. Using several theoretical methods, it was shown that $S = 0$ and $S = 1$ two-triplet bound-states exist below the two-triplet continuum in addition to the one-triplet excitation [2, 3, 4, 5, 6, 7, 8]. Such an $S = 0$ two-triplet bound-state was identified in the optical conductivity measured for an $S = 1/2$ two-leg spin ladder material (Ca$_n$La)$_{1/4}$Cu$_{2/4}$O$_{11}$ [9]. In this experiment, the strength of the interchain coupling ($J_\parallel$) and the intrachain coupling ($J_\perp$) was estimated as $J_\parallel/J_\perp \sim 1 - 1.2$ with $J_\perp \sim 1020 - 1100$cm$^{-1}$ [10, 11].

In addition to the interchain and intrachain couplings, a cyclic four-spin interaction, which acts among four $S = 1/2$ spins forming a plaquette, has been introduced to explain experimental findings for cuprate two-leg spin ladder systems. The analysis of the one-triplet mode observed by inelastic neutron-scattering experiments for La$_n$Ca$_{8/4}$Cu$_{2/4}$O$_{11}$ revealed that a cyclic four-spin interaction is necessary to reproduce the observed dispersion relation [12, 13]. For a $S = 1/2$ two-leg spin ladder material SrCu$_2$O$_3$, two sets of coupling constants, (i) $J_\parallel = 86$meV, $J_\perp = 172$meV [12] and (ii) $J_\parallel = 150$meV, $J_\perp = 195$meV, $J_{cyc} = 18$meV, $J_{diag} = 3$meV [4] with $J_{cyc}$ and $J_{diag}$ being the coupling constants for a cyclic four-spin interaction and a diagonal interaction, have been proposed to reproduce temperature dependence of the susceptibility. It seems thus difficult to decide the proper model only from temperature dependence of the susceptibility. Detailed information on dynamical properties and low-lying excitations is desirable to discuss characteristics of $S = 1/2$ two-leg spin ladder systems.

In this paper, we calculate the dynamical structure factor (DSF) of $S = 1/2$ two-leg spin ladder systems using continued fraction method based on Lanczos algorithm. Dynamical properties of $S = 1/2$ pure two-leg spin ladder systems have already been studied in Refs. 15, 3, 16, 17, and 4. We perform systematic calculation for DSF and characteristics of the lowest excited states, laying stress on the effects of a cyclic four-spin interaction.

Let us first consider the $S = 1/2$ two-leg spin ladder systems described by the following Hamiltonian,

$$
\mathcal{H} = J_\parallel \sum_{i=1}^{N/2} (S_{1,i} \cdot S_{1,i+1} + S_{2,i} \cdot S_{2,i+1}) + J_\perp \sum_{i=1}^{N/2} S_{1,i} S_{2,i},
$$

where $S_{i,l}$ denotes the $S = 1/2$ spin operator in the $l$th rung of the $i = 1, 2$ chain, and $N$ is the total number of the site. The periodic boundary condition is applied along the chain. The energy is measured in units of $J_\parallel$. The DSF can be expressed in the form of a continued fraction [13] as

$$
S^\mu(q_x, q_y; \omega) = \langle \Psi_0 | S^\mu_{q_x, q_y} \frac{1}{\mathcal{H} - S^\mu_{q_x, q_y}} | \Psi_0 \rangle = S^\mu(q_x, q_y) C^\mu(q_x, q_y; \omega), \quad (\mu = +, -, z)
$$

where $| \Psi_0 \rangle$ is the eigenfunction corresponding to the lowest eigenvalue $E_0$. $q_x$ and $q_y$ denote the wave numbers along the leg and the rung, respectively, and $\omega = \omega + i\varepsilon + E_0$ with $\hbar = 1$. We set $\varepsilon = 3.0 \times 10^{-2}$. The Fourier transform of the spin operator $S^\mu_{q_x, q_y}$ is given by

$$
S^\mu_{q_x, q_y} = (1/\sqrt{N}) \sum_{l,j} \exp[i(q_x l + q_y j)] S^\mu_{l,j} \quad S^\mu(q_x, q_y; \omega) = \text{constant structure factor and } C^\mu(q_x, q_y; \omega) \text{ is represented in the form of the continued fraction. Since rotational symmetry around the } x, y, \text{ and } z \text{ axes remains, we only calculate } S^x(q_x, q_y; \omega).
$$

In Figs. 1(a) and 1(b), we show $S^x(q_x, \pi; \omega)$ and $S^x(q_x, 0; \omega)$ for $N = 28$, respectively, in $J_\perp/J_\parallel = 1/5$, 1, and 2. The weight of $S^x(q_x, q_y; \omega)$ is proportional to the area of the full circle. The accuracy of the continued fraction becomes worse with decreasing $J_\perp/J_\parallel$ and/or increasing $\omega$. In $J_\perp/J_\parallel = 0.5$ the convergences have the relative errors about $O(10^{-3})$ for $\omega \geq 4$ and about $O(10^{-10})$ for $\omega < 3$, whereas in $J_\perp/J_\parallel = 2$ they have the relative errors about $O(10^{-10})$ even for $\omega \sim 6$.

In Fig. 1(a), the largest weights in given $q_x$ lie in the lowest excited states, and the largest weight at $q_x = \pi$ becomes dominant among them with decreasing $J_\perp/J_\parallel$. The solid line represents the dispersion relation of the one-triplet excitation adequate for $J_\perp/J_\parallel \gg 1$ [14]. The
Inelastic one-triplet mode below the excitation continuum. The intensity of each pole is proportional to the area of the circle. The solid and broken lines represent the dispersion relations of the one-exciton triplet mode and the lowest excited states, respectively, adequate for \( J_\perp/J_\parallel \gg 1 \).

Results show that the lowest excited states form an isolated mode. In the figures, the intensity of each pole is proportional to the area of the circle. The solid and broken lines represent the dispersion relations of the one-exciton triplet mode and the lowest excited states, respectively, adequate for \( J_\perp/J_\parallel \gg 1 \).

To discuss characteristics of the lowest excited states in given \( q_x \) lie in the lowest excited states. As shown in Fig. 2(b), the residue in \( J_\perp/J_\parallel = 2 \) decreases drastically around \( q_x \approx 0.6\pi \) with decreasing \( q_x \). This result means that the weights spread in many excited states in \( q_x \approx 0.6\pi \). Therefore, in \( J_\perp/J_\parallel = 2 \), the lowest excited states in \( q_x > 0.6\pi \) form an isolated mode of the \( S = 1 \) two-triplet bound state, while those in \( q_x < 0.6\pi \) become a lower bound of the two-triplet continuum. In \( J_\perp/J_\parallel \leq 1 \), the size dependence of the residue becomes noticeable except for the region \( q_x \approx 0 \). Thus, the lowest excited states in \( J_\perp/J_\parallel \leq 1 \) are the lower bound of the continuum formed by dressed spinons.

We now investigated the effects of a cyclic four-spin interaction described by the following expression,

\[
H_{cycl} = J_{cycl} \sum_{i=1}^{N/2} \left\{ (S_{i,i} \cdot S_{i+1,i+1})(S_{2,i+1} \cdot S_{2,i}) + (S_{i,i} \cdot S_{i+1,i+1})(S_{1,i+1} \cdot S_{2,i}) + (S_{2,i+1} \cdot S_{2,i}) + (S_{1,i+1} \cdot S_{2,i}) \right\},
\]

From inelastic neutron-scattering experiments for La\(_6\)Ca\(_8\)Cu\(_{24}\)O\(_{41}\), the coupling constants were estimated as \( J_\perp = 110\) meV and \( J_{cycl} = 16.5\) meV. Thus, we set \( J_\perp/J_\parallel = 1 \) and \( J_{cycl}/J_\parallel = 0.1 \). Combining the Hamiltonian (1) with the contribution (3), we calculate DSF. The results for \( N = 28 \) are shown in Fig. 3. In the poles and their residues for \( q_y = \pi \), noticeable size dependence comes out in contrast with the case of \( J_\perp/J_\parallel = 1 \) and \( J_{cycl} = 0 \) shown in Fig. 2. Therefore, in \( q_y = \pi \), the isolated mode is reduced by a cyclic four-spin interaction, since it may act as a frustration term: The lowest excited states in \( q_x < 0.7\pi \) become a lower bound of the excitation continuum, while those in \( q_x > 0.7\pi \) keep an isolated mode.

In Fig. 4, the weights of the lowest excited states in \( J_{cycl}/J_\parallel = 0.1 \) are compared with those in \( J_{cycl} = 0 \). In \( q_y = \pi \), the weights in \( q_x \approx \pi \) are enhanced by \( J_{cycl} \), while those in \( q_x < \pi \) are suppressed. In \( q_y = 0 \), on the contrary, the weights in \( q_x > \pi/2 \) are suppressed, while those in \( q_x < \pi/2 \) are enhanced. It is apparent that the distribution of the weights of DSF is drastically rearranged by a small cyclic four-spin interaction.

We next investigate dynamical properties of the systems described by the parameter set (ii) for SrCu\(_2\)O\(_3\) mentioned previously. We add the contribution of a diagonal interaction described as \( J_{diag} \sum (S_{1,i} \cdot S_{2,i+1} + S_{1,i+1} \cdot S_{2,i}) \) to the expressions (1) and (3). The results for \( N = 28 \) are shown in Fig. 5. Note that the re-
and the residues. Therefore, the lowest excited states
0, noticeable size dependence appears both in the poles
and their residues in $J_{\perp}/J_1$ been already presented in Figs. 1 and 2. In $q_y = \pi$, the dispersion relation
for the parameter set (i) ($J_{\perp}/J_1 = 0.5$) have been already presented in Figs. 1 and 2. In $q_y = \pi$ and 0, noticeable size dependence appears both in the poles and the residues. Therefore, the lowest excited states
in $0 \leq q_x \leq \pi$ are the lower bounds of the excitation continuum as in the case of the parameter set (i).

In Fig. 6, the dispersion relations and the weights of the lowest excited states for both parameter sets (i) and (ii) are compared. In $q_y = \pi$, the dispersion relation for the parameter sets (ii) becomes flatter in $q_x < 0.7\pi$, whereas the weights for both parameter sets are almost the same. The results are qualitatively the same as those for $N = 24$. In $q_y = 0$, the dispersion relation for the parameter set (ii) becomes flatter with suppressed weights in $q_x > \pi/2$. The difference in the distribution of the weights between two parameter sets may be difficult to observe experimentally. On the other hand, it has been shown that the effects of a flat dispersion relation of the lowest excited states appear in phonon-assisted optical absorption spectrums [8, 9, 10]. To decide the proper model for SrCu$_2$O$_3$, such experiments may be effective.
In summary, we have investigated dynamical properties of $S = 1/2$ two-leg spin ladder systems. It has been shown that the largest weights in given $q_x$ lie in the lowest excited states. The distribution of the weights is influenced by a cyclic four-spin interaction. We have further shown that due to a cyclic four-spin interaction, the lowest excited states for $q_y = \pi$ in La$_6$Ca$_8$Cu$_{24}$O$_{41}$ become a lower bound of the excitation continuum in $q_x < 0.7\pi$, while those in $q_x > 0.7\pi$ may form an isolated mode. We have also discussed the difference in dynamical properties between two systems proposed to describe SrCu$_2$O$_3$.

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FIG. 5: $S^z(q_x, q_y, \omega)$, and the size dependence of the poles and their residues in $J_\perp/J_\parallel = 0.770, J_{yc}/J_\parallel = 0.0923$, and $J_{dya}/J_\parallel = 0.0154$ for $N = 28$. Those values correspond to the parameter set (ii) for SrCu$_2$O$_3$ [1].

FIG. 6: The dispersion relations and the weights of the lowest excited states in the parameter sets (i) and (ii) for SrCu$_2$O$_3$.

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