Dynamical Structure Factors of $S = 1$ Bond-Alternating
Heisenberg Chains at Finite Temperatures

Takahumi SUZUKI and Sei-ichiro SUGA

Department of Applied Physics, Osaka University, Suita 565-0871, Japan

Dynamical structure factors of the $S = 1$ bond-alternating spin chains in the dimer state are calculated at finite temperature using the pair dynamical correlated-effective-field approximation. At $T = 0$, the delta-function-type peak corresponding to the one-magnon mode appears. When temperature increases, such a sharp peak is broadened and the additional weak peak caused by the excitation from the triplet state to the quintet state emerges in the higher energy region. The results are discussed in comparison with those obtained by the exact diagonalization method.

$S = 1$ bond-alternating Heisenberg chains have attracted attention both theoretically and experimentally. It was shown that the ground state of the system is in the Haldane phase in $\alpha_c < \alpha < 1$ and in the dimer phase in $\alpha < \alpha_c$, where the bond-alternation ratio is $1 : \alpha$.\(^1,2\) Recent progress on material synthesis has made it possible to study the elementary excitation as well as thermodynamic properties of $S = 1$ bond-alternating Heisenberg chains experimentally. Furthermore, dynamical properties have been investigated by inelastic neutron-scattering experiments for a typical compound NTENP.\(^3\) Note that compounds synthesized so far are in the dimer phase. Therefore, it may be desirable to investigate dynamical properties of $S = 1$ bond-alternating Heisenberg chains in the dimer phase at finite temperatures. In this paper, we calculate dynamical structure factors and static susceptibility in the dimer phase by pair dynamical correlated-effective-field approximation (Pair-DCEFA).\(^4\) Pair-DCEFA was successfully applied to calculate the dynamical structure factor as well as the static susceptibility of the $S = 1/2$ bond-alternating Heisenberg chain.\(^4\)

Let us consider the $S = 1$ bond-alternating Heisenberg chain described by the following Hamiltonian,

$$
\mathcal{H} = J \sum_i \left( S_{i,1} \cdot S_{i,2} + \alpha S_{i-1,2} \cdot S_{i,1} \right), \quad (J > 0)
$$

(1)

where $S_{i,1}(S_{i,2})$ denotes the operator of the spin on the left side (right side) in the unit cell $i$. We assume $|\alpha| < 1$. In Pair-DCEFA, we first consider the singlet, triplet, and quintet states in an isolated spin pair on the strong bond. In thermal equilibrium, the excitation from the singlet state to the triplet state and that from the triplet state to the quintet state take place.

Effective Hamiltonian in Pair-DCEFA is given by

$$
\mathcal{H}_{\text{eff}} = J \sum_i S_{i,1} \cdot S_{i,2}
$$

$$
+ \alpha J \sum_i \left[ S_{i,1} \cdot (\langle S_{i-1,2} \rangle - \lambda (S_{i,1})) + S_{i,2} \cdot (\langle S_{i+1,1} \rangle - \lambda (S_{i,2})) \right],
$$

(2)
where \(\langle S_{\mu\nu}\rangle\) (\(\nu = 1, 2\)) denotes the spontaneous moment and \(\lambda\) is a correlation parameter determined by the self-consistency equation derived from the fluctuation-dissipation theorem. We confine ourselves to consider the paramagnetic state. On the basis of the effective Hamiltonian, the self-consistency equation for \(\lambda\) is obtained as

\[
\frac{8}{3} = \frac{1}{N} \sum_q \frac{J}{(\hbar \omega_+)^2 - (\hbar \omega_-)^2} \left[ \coth \left( \frac{\beta \hbar \omega_+}{2} \right) \left( \frac{d_1 \hbar \omega_+ - d_2 J^2}{\hbar \omega_+} \right) - \coth \left( \frac{\beta \hbar \omega_-}{2} \right) \left( \frac{d_1 \hbar \omega_- - d_2 J^2}{\hbar \omega_-} \right) \right],
\]

(3)

where \(N\) is the total number of unit cells, \(\beta = 1/k_B T\), \(d_1 = (4/3)(2\Delta \rho_0 + 5\Delta \rho_1)\), and \(d_2 = (4/3)(8\Delta \rho_0 + 5\Delta \rho_1)\) with \(\Delta \rho_0 = \rho_s - \rho_{t} \) and \(\Delta \rho_1 = \rho_t - \rho_q\). Note that \(\rho_s(= \exp(2\beta J)/Z), \rho_t(= \exp(\beta J)/Z), \) and \(\rho_q(= \exp(-\beta J)/Z)\) are the contributions from the singlet, triplet, and quintet states with \(Z = \exp(2\beta J) + 3 \exp(\beta J) + 5 \exp(-\beta J)\), and take the values 1, 0, and 0, respectively, when \(T \to 0\). The dispersion relation of the one-magnon mode is given by

\[
\omega_{\pm}(q) = \frac{J}{\sqrt{2}} \left[ (5 - \alpha d_1 \{\lambda + \cos[q(c + c')]) \right] \\
\quad \pm \sqrt{\left( 5 - \alpha d_1 \{\lambda + \cos[q(c + c')]) \right)^2 - 4 \left( 4 - \alpha d_2 \{\lambda + \cos[q(c + c')]) \right)^2 },
\]

(4)

where \(\omega_+(q)\) and \(\omega_-(q)\) denote the dispersion relations of the excitations from the singlet state to triplet state and from the triplet state to quintet state, and \(c\) and \(c'\) are the lengths between the neighboring spins along the strong bond and the weak bond, respectively.

In Pair-DCEFA, the static susceptibility \(\chi(T)\) and dynamical structure factor \(S(q, \omega)\) are expressed as

\[
\chi(T) = \frac{2(g \mu_B)^2 (\rho_t + 5 \rho_q)}{1 - \alpha \xi \rho_t (\rho_t + 5 \rho_q)},
\]

(5)

\[
S(q, \omega) = \frac{2J}{1 - e^{-\beta \hbar \omega}} \frac{2[1 - \cos(qc)]}{(\hbar \omega_+)^2 - (\hbar \omega_-)^2} \left[ \frac{d_1 (\hbar \omega_+)^2 - d_2 J^2}{\hbar \omega_+} \frac{\delta}{(\omega - \omega_+)^2 + \delta^2} - \frac{d_1 (\hbar \omega_-)^2 - d_2 J^2}{\hbar \omega_-} \frac{\delta}{(\omega - \omega_-)^2 + \delta^2} \right].
\]

(6)

In the following, we set \(c = c' = 1, k_B = \hbar = 1, \) and \(\delta = 1.0 \times 10^{-2}\). The energy is measured in units of \(J\). We solve Eqs. (3) and (4) numerically to obtain \(\lambda\) in given \(T\) for a fixed \(\alpha\). Using \(\lambda\) thus obtained, we calculate \(\omega_{\pm}(q), \chi(T), S(q, \omega)\).

We first show the dispersion curve \(\omega_-(q)\) at \(T = 0\). The results are shown in the extended zone scheme. In Fig. 1, the dispersion curves obtained by Pair-DCEFA are expressed by the solid lines for \(\alpha = -0.3, 0.1, 0.3,\) and 0.4. As a reference, the excitation energies obtained by the exact diagonalization method for 20 spin systems\(^5\) are plotted by dots. For \(\alpha = 0.1\) and 0.3 the agreement between both
Fig. 1. The dispersion curves $\omega(q)$ at $T = 0$ for $\alpha = -0.3, 0.1, 0.3$, and 0.4. The results obtained by Pair-DCEFA are expressed by the solid lines and those obtained by the exact diagonalization method for 20 spin systems are plotted by dots.

Fig. 2. The static susceptibility $\chi(T)$ for $\alpha = -0.1, 0.1$, and 0.3. The results obtained by Pair-DCEFA are expressed by the solid lines and those obtained by the exact diagonalization (ED) method are expressed by the broken lines.

results is excellent, while for $\alpha = -0.3$ and 0.4 the deviation becomes noticeable in $q \sim 0$ and $\pi$. Thus, Pair-DCEFA may be effective in $-0.3 < \alpha \leq 0.3$ for the $S = 1$ bond-alternating Heisenberg chain.

We show the static susceptibility $\chi(T)$ in Fig. 2. The results for $\alpha = -0.1, 0.1$, and 0.3 are compared with those obtained by the exact diagonalization method for 8 spin systems. Note that $\chi(T)$ by the exact diagonalization method exhibits little size dependence. The agreement between both results becomes better in low temperatures.

In Fig. 3, dynamical structure factors at finite temperatures are shown. Accord-
Fig. 3. $S(q, \omega)$ at $T = 0, 0.4$, and $0.8$ for $\alpha = -0.1, 0.1$, and $0.3$.

According to the finite-size analysis for the $S(q, \omega)$ obtained by the exact diagonalization method, the lowest excited states in $\alpha = -0.1, 0.1$, and $0.3$ form the one-magnon mode in $0 \leq q \leq \pi$. Since we turn our attention to the behavior of the isolated mode located around $\omega \sim 1.0$ and $2.0$, we set $T < 1.0$. At $T = 0$ the intensity in $\alpha = -0.1$ takes the maximum at $q = 0.95\pi$, while in $\alpha = 0.1$ and $0.3$ the intensity at $T = 0$ takes the maximum at $q = \pi$. We have confirmed that the wave number of the maximum intensity at $T = 0$ shifts towards $\pi/2$, as $\alpha$ decreases in $\alpha < 0$. The results are consistent with those obtained by the exact diagonalization method. When temperature increases, the intensity of the one-magnon mode around $\omega \sim 1.0$, which corresponds to the singlet-triplet excitation, is reduced. At finite temperatures, the additional mode with weak intensity appears around $\omega \sim 2.0$. At $T = 0.8$, its intensity takes the maximum at $q \sim \pi$ in $\alpha = 0.1$ and at $q = 0.78\pi$ in $\alpha = 0.3$, respectively, whereas the intensity of the lower-energy mode in $\alpha > 0$ takes the max-
imum at \( q = \pi \) even at finite temperatures. The excitation mode around \( \omega \sim 2.0 \) is caused by the triplet-quintet excitation in thermal equilibrium, which vanishes at \( T = 0 \) because \( \rho_t \) and \( \rho_q \to 0 \) as \( T \to 0 \).

As shown in Fig. 1, the excitation continuum is located around \( \omega \sim 2.0 \) in \( \alpha = 0.3 \).\(^5\) Thus, the triplet-quintet mode becomes unstable in the continuum and disappears. In \( \alpha = 0.1 \), by contrast, the excitation continuum is located in the higher energy region.\(^5\) Therefore, the triplet-quintet mode may be stable. Recently, it was determined experimentally that a compound abbreviated NDOAP is well described by the isotropic \( S = 1 \) bond-alternating Heisenberg chain with \( \alpha \sim 0.1 \).\(^6\) When inelastic neutron-scattering experiments are performed on NDOAP, such a triplet-quintet mode with weak intensity may be observed around \( \omega \sim 2.0 \) in addition to the noticeable singlet-triplet mode around \( \omega \sim 1.0 \). According to our results, the ratio of the intensity of the higher-energy mode to that of the lower-energy mode at \( q = \pi \) is evaluated to be 22\% in \( T = 0.8 \).

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