Spin dynamics of the half-magnetization plateau in $S=1$ bond-alternating spin chains

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Abstract. We investigate dynamical properties of the half-magnetization plateau in $S=1$ bond-alternating spin chains. Dynamical structure factors calculated by the exact diagonalization method are compared with the dispersion curves of low-lying excitations obtained by a cluster expansion technique. We find that in the half-magnetization plateau the quintet excitation forms the lowest-lying excitation. The field dependence of the critical exponents of the spin correlation functions are further investigated. On the basis of the results, characteristics of the half-magnetization plateau are discussed.

In $S=1$ bond-alternating spin chains, it was shown theoretically that the plateau appears at a half of the saturation value in the magnetization curve [1]. According to the condition for the appearance of the magnetization plateau [2], the spatial period of this half-magnetization plateau in the $S=1$ bond-alternating spin chain is the same as that of the Hamiltonian [3]. This feature is in contrast with that in the $S=1/2$ bond-alternating spin chain with the next-nearest-neighbor (NNN) interaction, where the spatial period of the half-magnetization plateau is twice as large as that of the Hamiltonian, indicating that the translational symmetry is broken [3, 4]. This difference may induce characteristic dynamical properties in the half-magnetization plateaus of both models.

For some $S=1$ bond-alternating spin-chain compounds in the dimer phase, $[\text{Ni}_2(\text{dpt})_2(\mu-\text{ox})(\mu-\text{N}_3)]\text{PF}_6$ (abbreviated to NDOAP), $[\text{Ni}(\text{Medpt})_2(\mu-\text{oxy})(\mu-\text{N}_3)]\text{ClO}_4 \cdot 0.5\text{H}_2\text{O}$ (NMOAP), and $[\text{Ni}(\text{C}_9\text{H}_{24}\text{N}_4)(\text{NO}_2)]\text{ClO}_4$ (NTENP), where dpt = bis(3-aminopropyl)amine, ox = $\text{C}_2\text{O}_4$ and Medpt=methyl-bis(3-aminopropyl)amine, the half-magnetization plateaus were observed at 30T-40T, 40T-55T and $\sim70$T, respectively [5, 6]. Quite recently, inelastic neutron-scattering measurements for NTENP were performed in magnetic fields up to 14.5T. Characteristic spin dynamics distinct from those of the Haldane-gap compound NDMAP were revealed in the gapless regime as well as in the gapful regime [7]. Although spin dynamics in the plateau state are expected to be observed in the near future, the corresponding theoretical studies for the half-magnetization plateau have not been well studied yet. Detailed investigations are desirable.

In this paper, we investigate dynamical properties of the half-magnetization plateau in $S=1$ bond-alternating spin chains. We calculate the dynamical structure factor (DSF), using a continued fraction method based on the Lanczos algorithm. For the analysis of the low-lying excitations, the dispersion curves are calculated by using the cluster expansion technique [8]. Comparing both results, we discuss characteristics of the low-lying excitation in the half-magnetization plateau. We further investigate the critical exponents of the spin correlation

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functions in magnetic fields. The results are compared with those of the $S=1/2$ bond-alternating spin chains with the NNN interaction in magnetic fields. We discuss features of the half-magnetization plateau in a viewpoint of critical properties.

Let us consider the $S = 1$ bond-alternating spin chain with a single-ion anisotropy in magnetic fields described by the following Hamiltonian,

$$
\mathcal{H} = J \sum_{i=1}^{N/2} (S_{2i-1} \cdot S_{2i} + \alpha S_{2i} \cdot S_{2i+1}) + D \sum_{i=1}^{N} (S_{i}^{z})^{2} - g\mu_{B}H \sum_{i=1}^{N} S_{i}^{z},
$$

where $N$ and $H$ denote the total number of spins and the magnetic field, respectively. We set the coupling constant $J = 1$ and adopt $\hbar = g\mu_{B} = 1$. The periodic boundary condition is applied. The lattice constant between neighboring two sites is set to be unity and thus $q = 0.5\pi$ corresponds to the boundary of the Brillouin zone. We calculate the transverse DSF $S^{+}(q,\omega) = [S^{+-}(q,\omega) + S^{-+}(q,\omega)])/2$ up to $N = 20$. Applying the method discussed in Ref. 9, we analyze the finite-size effects and determine whether the low-lying excitation forms the isolated mode or the excitation continuum in the thermodynamic limit.

To calculate the dispersion curve of the low-lying excitation by the cluster expansion technique, the second term in the first parenthesis of the Hamiltonian is treated as the perturbation term. In the half-magnetization plateau, the ground state of the unperturbed state is expressed by the direct product of triplet dimers with $S^{z}=1$. For the low-lying excitation, three excited states are considered: the quintet state with $S^{z}=2$, the singlet state, and the triplet state with $S^{z}=0$, which are abbreviated to $q^{++}$, $s^{0}$, and $t^{0}$, respectively. We calculate the dispersion curves for the $q^{++}$ mode, the $s^{0}$ mode and the $t^{0}$ mode up to the seventh order and apply the $[3/4]$ Pade approximation to make the dispersion curves precise.

In Fig. 1, $S^{+}(q,\omega)$ and the dispersion curves thus obtained are shown. The parameter sets used are $(\alpha, D) = (0.25, 0.08)$, $(\alpha, D) = (0.45, 0.25)$, $(\alpha, D) = (0.45, 0.0)$, and $(\alpha, D) = (0.8, 0.0)$. The first two correspond to NMOAP [6] and NTENP [7], respectively. The area of the circle is proportional to the scattering intensity. The full circles and the open circles indicate the isolated mode and the excitation continuum, respectively.

In NMOAP, the energy gap and the largest intensity appear at $q = \pi$. From the finite-size analysis, we have verified that the lowest-lying excitation becomes the isolated mode in $0 \leq q \leq \pi$. To decide the lowest-lying mode, the excitation energies of $S^{+-}(q,\omega)$ and $S^{-+}(q,\omega)$ are compared for a given $q$. Since $S^{+-}(q,\omega)$ and $S^{-+}(q,\omega)$ describes the excitation from the ground state with the magnetization $M = N/2$ to the excited state with $M = N/2 + 1$ ($M = N/2 - 1$), the lowest-lying excitation in $S^{+-}(q,\omega)$ in $S^{-+}(q,\omega)$ corresponds to the $q^{++}$ ($s^{0}$) mode. We find that the energies of the lowest-lying excitation for $S^{+}(q,\omega)$ are smaller than those of $S^{-+}(q,\omega)$ in $0 \leq q \leq \pi$. Therefore, in $S^{+}(q,\omega)$ the $q^{++}$ mode is the lowest-lying excitation. Indeed, the dispersion curve of the $q^{++}$ mode obtained by the cluster expansion technique is in agreement with the peaks of the lowest-lying excitation. Note that the deviation of the $q^{++}$ and $s^{0}$ excitations is so small that both excitations are expressed by the same line in the figure. The other isolated mode appears at $\omega \sim 1.7$ in $0 < q < \pi$. This isolated mode agrees well with the dispersion curve of the $t^{0}$ mode. Its intensity becomes large as $q$ approaches $0$. This behavior is in contrast with that of the $q^{++}$ mode, where the intensity increases as $q$ approaches $\pi$.

In NTENP, the energy gap and the largest intensity appear at $q = \pi$. As $\alpha$ increases (or the effect of the bond alternation becomes weak), the intensity at $q = \pi$ is enhanced. From the finite-size analysis, we have verified that the lowest-lying states in $0.2\pi < q < \pi$ form the isolated mode and those in $0 < q < 0.2\pi$ become the excitation continuum. The lowest excitation energies of $S^{+-}(q,\omega)$ are smaller than those of $S^{-+}(q,\omega)$ in $0 \leq q \leq \pi$. Therefore, the $q^{++}$ excitation form the lowest-lying state in $S^{+}(q,\omega)$, which well describes the lowest-lying excitation of the numerical results for $S^{+}(q,\omega)$. When $D$ is decreased down to $(\alpha, D) = (0.45, 0.0)$, the
Figure 1. The transverse DSF $S^\perp(q,\omega)$ in the half-magnetization plateau for $N=20$. The intensity is proportional to the area of the circle. Full circles denote the isolated mode and open circles denote the excitation continuum. The solid and broken curves represent the $q^{++}$ and $t^0$ excitations, respectively.

The excitation continuum shifts to the low energy region and spreads in the wider wave-number region $0 < q < 0.7\pi$ than in NTENP. When $\alpha = 0.45$, the excitation energy of the $s^0$ is slightly larger than that of the $q^{++}$, in particular, around $q \sim 0.5\pi$.

When $(\alpha, D) = (0.8, 0.0)$ where the system is in the Haldane-gap phase at $H = 0$, the gapped isolated $q^{++}$ mode appears only in the vicinity of $q = \pi$. This results may reflect the fact that the half-magnetization-plateau region is reduced as $\alpha$ approaches 1 [1]. The parameter set $(\alpha, D) = (1, 0)$ is the critical point concerning the appearance of the half-magnetization plateau. As the system approaches the critical point, the excitation gap is reduced and the excitation continuum shifts to the low energy region, which makes the isolated mode unstable. Note that in $q < 0.4\pi$ there are invisibly small peaks of the excitation continuum for $0 < \omega < 2$. In this parameter set, unfortunately, the dispersion curves by the cluster expansion technique cannot be obtained in satisfying accuracy, since the region is far from the unperturbed state $\alpha = 0$.

We have shown that the low-lying excitation in the half-magnetization plateau is expressed by the particle-hole type excitation. This is in contrast with the low-lying excitation of the half-magnetization plateau in $S = 1/2$ bond-alternating spin chain with the NNN interaction, where the low-lying excitation is a superposition of the massive kinks and antikinks [10].

We next investigate critical properties in magnetic fields to discuss features of the low-lying excitation in the half-magnetization plateau in another point of view. When the spin gap vanishes in magnetic fields, it is expected that the low-lying excitation is described by the Tomonaga-Luttinger liquid (TLL). We calculate the critical exponents $\eta_z$ and $\eta$ of the longitudinal and transverse spin-correlation functions for the long-distance decay.

In Fig. 2, the field dependences of $\eta_z$ and $\eta$ for $(\alpha, D) = (0.45, 0.25)$ and $(\alpha, D) = (0.8, 0.3)$ are shown as functions of the normalized magnetization $m$. The former parameter set describes NTENP and the latter one is in the Haldane-gap phase. The critical exponents for both parameter sets take the values $\eta_z = 2$ and $\eta = 1/2$, when $m=0$ and 1. When $0 < m < 1$ except for $m=1/2$, they satisfy the relations $\eta < 1 < \eta_z$ and $\eta \cdot \eta_z \sim 1$, indicating that the
system is in the TLL. In the $S=1/2$ bond-alternating spin chain with the NNN interaction, the longitudinal incommensurate spin correlation becomes dominant around the half-magnetization plateau, making $\eta_z$ smaller so as to satisfy the relation $\eta_z < 1 < \eta$ [11, 12]. The dominant longitudinal incommensurate spin correlation is a manifestation of the instability of the TLL toward the magnetization plateau which can be regarded as the $4k_F$ CDW state [11]. Also in the 1D Hubbard model, it was shown exactly that the critical exponent of the $4k_F$ oscillation of the charge-density correlation function decreases and approaches the value at $U \to \infty$, as the change in the filling-factor drives the system to the Mott insulator [13, 14, 15]. In the $S=1$ bond-alternating spin chain, however, the critical exponent $\eta_z$ never decreases below 1 even when $m \sim 1/2$. This fact suggests that the half-magnetization plateau in the present system cannot be regarded as the Mott insulator in the correlated fermion language.

In summary, we have investigated dynamical properties of the half-magnetization plateau in $S=1$ bond-alternating spin chains. We have found that the lowest-lying excitation is the $q^{+\pm}$ state. As the system approaches the critical point concerning the half-magnetization plateau, the region for the isolated mode is reduced. On the basis of the field dependence of the critical exponents, characteristics of the half-magnetization plateau are discussed.

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