Dynamically dominant excitations of string solutions in the spin-1/2 antiferromagnetic Heisenberg chain in magnetic fields

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Using Bethe-ansatz solutions, we uncover a well-defined continuum in dynamical structure factor $S^{++}(k, \omega)$ of the spin-1/2 antiferromagnetic Heisenberg chain in magnetic fields. It comes from string solutions which continuously connect the mode of the lowest-energy excitations in the zero-field limit and that of bound states of overturned spins from the ferromagnetic state near the saturation field. We confirm the relevance to real materials through comparisons with experimental results.

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The spin-1/2 antiferromagnetic Heisenberg chain exhibits intriguing quantum many-body effects, associated with modern concepts in condensed-matter physics, such as spin liquids, quantum criticality and fractionalization. Also, this system is an excellent platform to make precise comparisons between experiments and theories: Various intriguing features predicted by exact solutions [1] can be confirmed by accurate experiments on quasi-one-dimensional materials. Actually, it is established that dynamical properties in the absence of magnetic field are characterized by quasiparticles called spinons [2, 3] through precise comparisons between theoretical predictions and experimental results [4, 5, 6].

In magnetic fields, dynamical properties are more complicated. Some basic features of dominant excitation spectra can be understood by modifying the 2-spinon continuum in zero field according to the strength of magnetic fields in the $k-\omega$ plane [7, 8, 9]. The distributions of spectral weights in $S^{++}(k, \omega)$ and $S^{zz}(k, \omega)$ are effectively expressed by low-order particle-hole excitations in Bethe-ansatz solutions [9, 10, 11]: The dominant excitations in $S^{++}(k, \omega)$ and $S^{zz}(k, \omega)$ are known as 2-spinon ($2\psi$) excitations and spinon-antispinon ($\psi\psi^*$) excitations, respectively [10, 11]. Their properties have been investigated in detail by using Bethe-ansatz solutions [10, 11].

As for $S^{+-}(k, \omega)$, the situation is much more complicated. Except the low-energy modes near momentum $k=0$ [3, 10, 18, 19, 20] and $k=\pi$ [3, 10, 19, 20], behaviors of $S^{+-}(k, \omega)$ have not been clarified: In Ref. [9], the continuum of $\psi\psi^*$ excitations was predicted to persist in the thermodynamic limit based on the classification by the Bethe formalism. However, numerical results indicated that the spectral weight in the $\psi\psi^*$ continuum is rather small except near the edges, and there exists a large fraction of spectral weights above the continuum [3, 20].

In this Letter, mainly focusing attention on behaviors of $S^{+-}(k, \omega)$ in magnetic fields, we identify excitations having large spectral weights to clarify overall dynamical features of the Heisenberg chain in magnetic fields.

We consider the spin-1/2 antiferromagnetic Heisenberg chain with $L$ sites, $M$ down spins ($M \leq L/2$) and periodic boundary conditions. The Hamiltonian is defined as

$$\mathcal{H} = J \sum_{x=1}^{L} \mathbf{S}_x \cdot \mathbf{S}_{x+1} - H S_z^z,$$

(1)

where $\mathbf{S}_x$ is the spin-1/2 operator at site $x$, and $J > 0$. Magnetic field $H$ at $S^z/L=1/2-M/L$ in the thermodynamic limit is obtained in Ref. [21]. In the Bethe ansatz [1], the wave function, energy and momentum of an eigenstate are expressed by a set of rapidities $\{\Lambda_j\}$ which is obtained from the Bethe equation: $L \arctan(\Lambda_j) = \pi I_j + \sum_{l=1}^{M} \arctan((\Lambda_j - \Lambda_l)/2)$, once a set of $\{I_j\}$ is given. Here, $I_j$ ($j=1-M$) are called Bethe quantum numbers, which are integers (half-odd integers) within $|I_j| \leq (L-M-1)/2$ for odd (even) $L-M$ as in Fig. 1 (a-e) for solutions of real $\{\Lambda_j\}$. Distributions of $\{I_j\}$ are somewhat analogous to momentum distributions of spinless fermions. As in Fig. 1 (b), a hole (particle) created inside (outside) the $\{I_j\}$ of the ground state is called spinon (antispinon) and denoted by $\psi^*$ ($\psi$) [11, 12].

It is also known that there are solutions with complex rapidities [3]. Later, we will consider solutions with an $n$-string ($n \geq 2$), i.e. a set of $n$ rapidities expressed as $\Lambda_j = \Lambda + (n+1-2j) + \eta_j$ for $j=1-n$, where $\Lambda$ is real, and $\eta_j = O(e^{-\epsilon_0})$ with $c > 0$ [16, 22, 23, 24]. We take real $\Lambda_j$ for $j \geq n+1$, and denote the $n$-string solutions by $S_n$. These solutions are specified by two sets of $\{I_j\}$ [22]: One is for the real rapidities, and the other is for the $n$-string, which we denote by $\{I_j^r\}$ and $\{I_j^s\}$, respectively. For $S_n$ ($n \geq 2$), $\{I_j^r\} = \{I_j^s\} = |I^s_1| \leq L/2 - M$ as in Fig. 1 (f-2). We denote $\psi$ and $\psi^*$ of $\{I_j^s\}$ by $\psi_s$ and $\psi^*_s$, respectively, and regard $\{I_j^s\}$ without $\psi_s$ or $\psi^*_s$ also as a part of those with $\psi_s\psi^*_s$.

We investigate behaviors of dynamical structure factors defined as $S^{\alpha\beta}(k,\omega) = \sum_\Omega M^{\alpha\beta}(k, \epsilon_\Omega) \delta(\omega - \epsilon_\Omega)$ for $\alpha=-, +$ and $z$. Here, $M^{\alpha\beta}(k, \epsilon_k)$ is the transition rate between the ground state $|\text{G.S.}\rangle$ in a magnetic field and an excited state $|k, \epsilon_k\rangle$ with excitation energy $\epsilon_k$ and momentum $k$, defined as $M^{\alpha\beta}(k, \epsilon_k) = \langle k, \epsilon_k | S^\alpha_s | \text{G.S.}\rangle \langle \text{G.S.} | S^\beta_s | k, \epsilon_k \rangle$. We calculated $M^{\alpha\beta}(k, \epsilon_k)$ of the Heisenberg chain, following Refs. [13, 10, 17, 18, 23, 24] where $M^{\alpha\beta}(k, \epsilon_k)$ is expressed in a determinant form whose matrix elements are...
FIG. 1: Distributions of Bethe quantum numbers \{I_j\}. Filled symbols denote \{I_j\} of the ground state of \(M=8\) and typical excited states in dynamical structure factors for \(0 \leq k \leq \pi\) in \(L=24\). Blue open square and red solid diamond denote psion \((\psi)\) and antipsion \((\psi^*)\), which are located at points on thick and thin lines, respectively. (a) Ground state. (b) \(\psi\psi^*\) for \(S^{zz}(k,\omega)\). (c) \(2\psi\) for \(S^{+-}(k,\omega)\). (d) \(2\psi^*\) for the continuum near \(k=\pi\) in \(S^{+-}(k,\omega)\). (e) \(1\psi^*\) for the low-energy mode near \(k=0\) in \(S^{+-}(k,\omega)\). (f-1) \(1/4\) of \{I_j\} for 2-string solutions in \(S^{+-}(k,\omega)\). (f-2) \(I_1\) for the n-string of \(S_n\) \((n \geq 2)\) in \(S^{+-}(k,\omega)\).

expressed in terms of rapidities \[24\]. As for string solutions with an \(n\)-string \((n \geq 2)\), we calculated \(M^{\alpha\alpha}(k,\epsilon_k)\) after transforming the matrices so that singularities in determinants can be cancelled as in Refs. \[16,23\]. In \(S^{+-}(k=0,\omega)\), we took into account the resonant mode of the field-induced magnetization \[9,10,18,19,20\]. In this Letter, we show results on dynamical structure factors in magnetic fields in \(L=320\) for \(0 \leq k \leq \pi\), noting \(S^{\alpha\alpha}(k,\omega)=S^{\alpha\alpha}(-k,\omega), \alpha=-,+,\) and \(z\).

We calculated the spectral weight in \(L=320\), using up to \(2\psi^*2\psi^*\) excitations, and evaluated the contributions to sum rules \[23\]. The results are shown by open symbols in Fig. 2 (a) and (b). The spectral weight of up to \(2\psi^*2\psi^*\) excitations in \(S^{+-}(k,\omega)\) satisfies more than 90\% of the sum rule in \(L=320\) as shown by open diamonds in Fig. 2 (b). The spectral weight in \(S^{+-}(k,\omega)\) severely decreases in the small \(S^z\) regime as shown by open circles in Fig. 2 (a). Figure 2 (e) shows \(S^{+-}(k,\omega)\) in the small \(S^z\) regime. Although \(S^{+-}(k,\omega)\) should be continuously connected to that in zero field (Fig. 2 (d)) as the magnetic field decreases, most of the spectral weight in \(S^{+-}(k,\omega)\) is missing. In particular, the large intensity near the des Cloizeaux-Pearson mode (dCP) \[26\] (the lower edge of the continuum in zero field) is almost lost. This implies that there are other important excitations for \(S^{+-}(k,\omega)\).

To explain the origin of the missing spectral weight, we consider \(S_2\) and \(S_3\) defined above. Among many states in \(S_2\) and \(S_3\), we found that \(S_2\) with \(\psi\psi^*\) and \(S_3\) with \(2\psi^*\) have large weights in \(S^{+-}(k,\omega)\). By taking into account these string solutions, \(S^{+-}(k,\omega)\) in the small \(S^z\) regime almost recovers the missing weight near the dCP mode and that of the continuum near \(k=\pi\) as shown in Fig. 2 (f). The intensity near the dCP mode is mainly due to the \(S_2\), and that of the continuum near \(k=\pi\) is mainly due to the \(S_3\). The contributions from these string solutions increase as the magnetic field decreases as shown by solid symbols in Fig. 2 (a). By taking them into account, more than 80\% of the total spectral weight in \(S^{+-}(k,\omega)\) is explained in a wide range of \(S^z\) in \(L=320\) \[27\].

As for \(n\)-string solutions \((n \geq 2)\) in \(S^{zz}(k,\omega)\), we found that a large spectral weight is carried by \(S_2\) with \(2\psi^*\) in low fields. As shown by solid squares in Fig. 2 (b), the decrease of \(S^{zz}(k,\omega)\) of up to \(2\psi^*2\psi^*\) excitations in the small \(S^z\) regime (open squares in Fig. 2 (b)) is almost compensated for by the \(S_2\), and more than 90\% of the total spectral weight in \(S^{zz}(k,\omega)\) is explained in a wide range of \(S^z\) in \(L=320\) \[27\]. These string solutions are relevant to the continuum near \(k=\pi\) in low fields.

Figure 3 (a) shows behaviors of dynamical structure factors in magnetic fields in \(L=320\), where we took into account excitations of up to \(2\psi^*2\psi^*\) and the above-described string solutions. Hereafter, we use these excitations to calculate dynamical structure factors. In \(S^{+-}(k,\omega)\), there are three sets of dominant continua as shown in the top row of Fig. 3 (a). The high-energy continuum \((\omega/J>2)\), which is mainly due to \(S_2\), goes up to higher energies as the magnetic field increases, separated from the low-energy continua. Although this continuum has been observed in the high-field regime by numeri-
ically dominant in $S$ and $(from above)$ at $S^2/L=1/8$, $1/4$ and $3/8$ (from the left) in $L=320$. The data are broadened in a Lorentzian form with FWHM=0.08. (b) Same as (a) but those of dynamically dominant excitations in $L=2240$ with the product ansatz.

Fig. 1 (f-1), which can be regarded as a part of the above-dominant excitations in $L$ near $k=\pi$ mainly comes from $S$.

From many excitations considered above, we extract dynamically dominant excitations of $O(L^2)$ states which characterize the behaviors of Fig. 3 (a). Then, the product ansatz $^{11}$: $S^{\text{dom}}_{\text{dom}}(k, \omega)=M^{\text{dom}}_{\text{dom}}(k, \omega)D(k, \omega)$ with $D(k, \epsilon_k^L)=2/(\epsilon_k^L-\epsilon_k^L)$ can be applied. Here, in energy regions where there is more than one sequence of states, we took the one with the largest spectral weight. Hereafter, we denote $S^{\text{dom}}_{\text{dom}}(k, \omega)$ of the dynamically dominant excitations as $S^{\text{dom}}_{\text{dom}}(k, \omega)$ for $\alpha=-, +$ and $z$. Their contributions to sum rules in $L=2240$ are shown in Fig. 2 (c).

The top row of Fig. 3 (b) shows $S^{\text{dom}}_{\text{dom}}(k, \omega)$. The low-energy continuum near $\omega/J=1.7$ is due to $S^2$ with $1\psi_\alpha$ as in Fig. 1 (f-1), which can be regarded as a part of the abovementioned $S^2$ with $\psi_\alpha\psi_\alpha^*$. The low-energy continuum near $k=\pi$ comes from $2\psi^*$ excitations where two $\psi^*$s are located on opposite sides of the filled region of $\{I_f\}$ from each other as in Fig. 1 (d). The low-energy mode near $k=0$ is due to excitations with one $\psi^*$ in the right empty region of $\{I_f\}$ for $0\leq k\leq \pi$ as in Fig. 1 (e). This mode has been mentioned in the literature. $^{9,10,13,15,20}$

The dynamically dominant excitations for $S^{-+}(k, \omega)$ and $S^{zz}(k, \omega)$ are known as $2\psi$ and $\psi^*\psi^*$ excitations as in Fig. 1 (c) and (b), respectively. $^{9,10,11}$ The results are shown in the second and third rows of Fig. 3 (b).

Noting that $2\psi$, $\psi^*\psi^*$ and $2\psi^*$ excitations are dynamically dominant in $S^{-+}(k, \omega)$, $S^{zz}(k, \omega)$ and $S^{+-}(k, \omega)$ and that these excitations have $S^z=+1$, 0 and $-1$, respectively, we can naturally assign $S^z=+1/2$ and $-1/2$ to $\psi$ and $\psi^*$, respectively. Also, noting that excitations of $S^2$ with $\psi_\alpha\psi_\alpha^*$ and those with $2\psi_\alpha$ have large weights in $S^{-+}(k, \omega)$ and $S^{zz}(k, \omega)$ and that these excitations have $S^z=-1$ and 0, respectively, we can naturally interpret the quasiparticle representing the 2-string (Fig. 1 (f-2)) as a bound state of two $\psi^*$s which carries $S^z=-1$. This assignment is also applicable to 4-spinon states of $S^{-+}(k, \omega)$ in zero field which have four spinons and a 2-string. The $1\psi^*$ mode near $k=0$ in $S^{+\bar{z}}(k, \omega)$ can also be regarded as the most dominant part of a $2\psi^*$ continuum where both $\psi^*$s are located in the right empty region of $\{I_f\}$ (Fig. 1 (e) as compared with (d)) for $0\leq k\leq \pi$. But, since the spectral weight in this continuum is very small except the $1\psi^*$ mode, the $\psi^*$ in this mode practically behaves as a quasiparticle carrying $S^z=-\bar{z}$, which reduces to a magnon carrying $S^z=-1$ above the saturation field.

To confirm the relevance to real materials, we compare the present results with available experimental data on quasi-one-dimensional materials which can be effectively regarded as spin-1/2 antiferromagnetic Heisenberg chains. In inelastic neutron scattering experiments, a quantity proportional to $S^{\text{av}}(k, \omega)$ is observed. Here, we define $S^{\text{av}}(k, \omega)\equiv[S^{-+}(k, \omega)+S^{+\bar{z}}(k, \omega)+4S^{zz}(k, \omega)]/6$. The fourth row of Fig. 3 shows the results of $S^{\text{av}}(k, \omega)$ and $S^{\text{av}}_{\text{dom}}(k, \omega)$, where $S^{\text{av}}_{\text{dom}}(k, \omega)$ denotes $S^{\text{av}}(k, \omega)$ of the dynamically dominant excitations.

In Fig. 4 (a), we compare the present results with experimental results on CuCl$_2$-2N(C$_5$D$_5$)$_2$ (CPC) $^{28}$. We broadened the numerical results of $S^{\text{av}}(k, \omega)$ in a gaus-
The dynamical, dominant excitations of these solutions continuously connect the dCP mode in the low-energy regime, which leads to a natural interpretation of $\psi$ and $\psi^*$ as quasiparticles in magnetic fields carrying $S^z=\pm 1/2$ and $-1/2$, respectively. Comparisons with available experimental results reasonably support the relevance to real materials. For the identification of dynamically dominant excitations, further experiments in higher fields at high energies are desired. Behaviors shown in this Letter are expected to be more or less true for general spin-1/2 XXZ chains in magnetic fields.

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[1] H. Bethe, Z. Phys. 71, 205 (1931).
[2] L. D. Faddeev et al., Phys. Lett. A 85, 375 (1981).
[3] F. D. M. Haldane, Phys. Rev. Lett. 66, 1592 (1991).
[4] D. A. Tennant et al., Phys. Rev. B 52, 13668 (1995).
[5] D. C. Dender et al., Phys. Rev. B 53, 2583 (1996).
[6] P. R. Hammar et al., Phys. Rev. B 59, 1098 (1999).
[7] N. Ishimaru, and H. Shiba, Prog. Theor. Phys. 57, 1862 (1977); ibid. 64, 479 (1980).
[8] M. B. Stone et al., Phys. Rev. Lett. 91, 037205 (2003).
[9] G. Müller et al., Phys. Rev. B 24, 1429 (1981).
[10] M. Karbach et al., Phys. Rev. B 66, 054405 (2002).
[11] M. Karbach et al., Phys. Rev. B 62, 14871 (2000).
[12] A. Fledderjohn et al., Phys. Rev. B 54, 7168 (1996).
[13] J. -S. Caux et al., Phys. Rev. Lett. 95, 077201 (2005).
[14] R. G. Pereira et al., Phys. Rev. Lett. 96, 257202 (2006).
[15] J. Sato et al., J. Phys. Soc. Jpn. 73, 3008 (2004).
[16] J. -S. Caux et al., J. Stat. Mech. P09003 (2005).
[17] D. Biegel et al., J. Phys. A: Math. Gen. 36 5361 (2003).
[18] D. Biegel et al., Europhys. Lett. 59, 882 (2002).
[19] S. Nishimoto et al., Int. J. Mod. Phys. B 21, 2262 (2007).
[20] K. Lefmann et al., Phys. Rev. B 54, 6340 (1996).
[21] R. B. Griffiths, Phys. Rev. 133, A768 (1964).
[22] M. Takahashi, Prog. Theor. Phys. 46, 401 (1971).
[23] R. Hagemans et al., AIP Conf. Proc. 846, 245 (2006).
[24] N. Kitanine et al., Nucl. Phys. B 544, 647 (1999).
[25] The total spectral weights of $S^{\pm 0}(k,\omega)$ and $L/4-[(S^z)^2]/L$, respectively [12, 16].
[26] J. des Cloizeaux et al., Phys. Rev. 128, 2131 (1962).
[27] Judging from small size-dependence in $L<320$ for Fig. 2 (a,b), we believe the physics will hold for larger systems.
[28] I. U. Heilmann et al., Phys. Rev. B 18, 3530 (1978).
[29] J. A. Chakhalian et al., Phys. Rev. Lett. 91, 027202 (2003).
[30] M. Kohno (unpublished).