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Excitation spectrum of a spin-$\frac{1}{2}$ chain with competing interactions

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The excitation spectrum is studied around the degenerate point at the Ising limit in the one-dimensional anisotropic Heisenberg antiferromagnets with next-nearest-neighbor interactions. Solitons are found as propagation modes in the Néel state and in the (2,2) antiphase state, while triplet-dimer excitations are found propagating in the dimer state. These excitations determine the shape of dynamical spin-correlation functions, which are calculated by an exact diagonalization for finite chains. The phase diagram is determined from the condition that the energy gap vanishes in the excitation spectrum.

Recently, one-dimensional quantum spin systems with competing interactions have attracted much attention as a model of statistical mechanics and the many-body problem, since they include two phenomena of current interest, frustration and quantum fluctuation. A typical example of such systems is the antiferromagnetic Heisenberg model with the next-nearest-neighbor interaction:

$$H = H_{ZZ} + H_{XY}\,, \tag{1}$$

where

$$H_{ZZ} = \sum_{i=1}^{N} (J_1 S_i^z S_{i+1}^z + J_2 S_i^z S_{i+2}^z)\,, \tag{2}$$

$$H_{XY} = \sum_{i=1}^{N} [J_1^{\perp}(S_i^x S_{i+1}^x + S_i^y S_{i+1}^y) + J_2^{\perp}(S_i^x S_{i+2}^x + S_i^y S_{i+2}^y)]\,, \tag{3}$$

with J_1, J_2, $J_1^{\perp}$, $J_2^{\perp} > 0$. $S_i^{\alpha}(\alpha = x,y,z)$ is the spin-$\frac{1}{2}$ operator at the ith site. Within the nearest-neighbor interaction, the ground state is the "spin fluid" state or the Néel state. The next-nearest-neighbor interaction complicates the situation, giving rise to the "dimer" state where neighboring spins are forming singlet pairs. This state, which is known to be the exact ground state under the condition $J_2^{\perp}/J_1^{\perp} = J_2/J_1 = 0.5$,[1,2] results apparently from the effect of the combination of the frustration and the quantum fluctuation. The renormalization-group technique has been successfully applied to this model, leading to the determination of the phase diagram among the spin fluid state, the dimer state, and the Néel state.[3-5]

At a glance, one may think that the quantum fluctuation in an Ising-like region would merely be suppressed and nothing interesting would happen. This observation, however, is not true in the presence of frustration; the degenerate point ($J_1 = 2J_2$, $J_1^{\perp} = J_2^{\perp} = 0$) appears where the ground state is infinitely degenerate due to frustration,[6-8] and quantum fluctuation could be important around this point. Figure 1 shows the ground-state phase diagram in the Ising-like region near the degenerate point, whose determination will be discussed later, under the condition $J_2^{\perp}/J_1^{\perp} = J_2/J_1$. On the other hand, Emery and Noguera[5] have recently studied the corresponding phase diagram under the condition $J_2^{\perp} = 0$, which is very similar to our phase diagram in the region near the degenerate point. We emphasize that, as seen from the appearance of the dimer state as the ground state, the effect of frustration and quantum fluctuation play an important role even in this rather limited region.

As a first step to attack the dynamical properties of the present system, we explore in this paper an Ising-like region near the degenerate point, expecting that the combination of frustration and quantum fluctuation may give rise to interesting effects; we study what low-lying excitations are and how they manifest in dynamical spin-correlation functions.

The situation near the degenerate point enables us to use a perturbation theory with respect to $J_1^{\perp}/|J_1 - 2J_2|$ [in the Néel and (2,2) antiphase states] and to

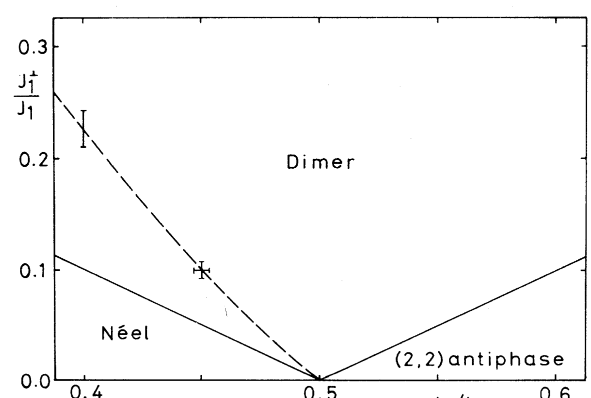

FIG. 1. The ground-state phase diagram on the $J_1^{\perp}/J_1$ vs J_2/J_1 plane under the condition $J_2^{\perp}/J_1^{\perp} = J_2/J_1$. The solid lines denote the phase boundaries determined by the condition that the minimum in the energy band of solitons touches on zero. The dashed line with error bars is the phase boundary determined from an exact diagonalization of finite chains, where higher-order effects are taken into account.

$|J_1-2J_2|/J_1^\perp$ (in the dimer state), thereby leading to a clear picture of excited states. The perturbational analysis explains well the behavior of the dynamical correlation function, which we numerically calculate in a finite chain of $N=12$, following Ishimura and Shiba's idea[9]

$$S_{zz}(Q,\omega)=(1/2\pi)\int_{-\infty}^{\infty} dt e^{-i\omega t}\langle S_{-Q}^z(0)S_Q^z(t)\rangle , \quad (4)$$

with

$$S_Q^z=N^{-1/2}\sum_j \exp(iQj)S_j^z .$$

We also study higher-order effects on the energy gap in the excitation spectrum, using an exact numerical diagonalization method on finite chains (up to $N=20$) under periodic boundary conditions to extrapolate the results to $N\to\infty$.[10]

In the following, we discuss excited states separately in three typical cases. In our units, $\hbar=k_B=1$.

(1) *Néel state with small $J_1^\perp$ and $J_2^\perp$.* We consider a soliton excitation (domain wall), which connects two degenerate ground states[11]

$$\psi_{2j+1}^{a(\beta)}=\uparrow\downarrow\ldots\uparrow\downarrow\uparrow\downarrow\alpha_{2j+1}(\beta_{2j+1})\uparrow\downarrow\uparrow\downarrow\ldots , \quad (5)$$

where α_{2j+1} and β_{2j+1} denote up-spin and down-spin states at the $2j+1$th site. The $\psi_{2j+1}^{a(\beta)}$ has an excitation energy $\varepsilon_0\equiv(\frac{1}{2})|J_1-2J_2|$ with respect to H_{ZZ}, if we neglect an extra energy due to a misfit at the boundary of the chain. For avoiding the misfit and being compatible with periodic boundary conditions, an even number of solitons should be excited. The spin-wave excitations, having high excitation energy of an order J_1, are irrelevant to the present case. The H_{XY} makes the soliton propagate in a chain; the $J_1^\perp$ term connects $\psi_{2j+1}^{a(\beta)}$ to $\psi_{2j-1}^{a(\beta)}$ and $\psi_{2j+3}^{a(\beta)}$ with the amplitude $J_1^\perp/2$, while the $J_2^\perp$ term connects it only to the states with high excitation energy of order J_1 and J_2. Neglecting such high-energy states, we get the wave function and the energy as

$$\psi(k)^{\alpha(\beta)}=(2/N)^{1/2}\sum_j \psi_{2j+1}^{a(\beta)}\exp[ik(2j+1)] , \quad (6)$$

$$\varepsilon(k)^{\alpha(\beta)}=\varepsilon_0+J_1^\perp\cos(2k) . \quad (7)$$

The propagation of solitons can be seen in $S_{zz}(Q,\omega)$. Figure 2(a) shows $S_{zz}(Q,\omega)$ of an $N=12$ chain; the width and the intensity of the central peak increase with increasing Q, and a squarelike shoulder develops, just like Villain's prediction in the model with only the nearest-neighbor interaction,[9,11–13] owing to propagation of solitons.

(2) *(2,2) antiphase state with small $J_1^\perp$ and $J_2^\perp$.* We consider two kinds of soliton excitations,

$$\psi_{4j+3}^a=\uparrow\downarrow\downarrow\uparrow\ldots\uparrow\downarrow\alpha_{4j+3}\uparrow\downarrow\downarrow\uparrow\ldots , \quad (8)$$

$$\psi_{2j+1/2}^b=\uparrow\downarrow\downarrow\uparrow\ldots\uparrow\downarrow|\uparrow\downarrow\downarrow\uparrow\uparrow\downarrow\ldots , \quad (9)$$

where $\psi_{2j+1/2}^b$ has a domain wall between the $2j$th and $2j+1$th spin. [The | is inserted in Eq. (9) to show the presence of the domain wall.] The ψ_{4j+3}^a and $\psi_{2j+1/2}^b$ have excitation energies $\varepsilon_0^a\equiv(\frac{1}{4})|J_1-2J_2|$ and $\varepsilon_0^b\equiv(\frac{1}{2})|J_1-2J_2|$ with respect to H_{ZZ}, if we neglect extra energies due to a misfit at the boundary of the chain. At least four ψ^a-type solitons or two ψ^b-type solitons should be excited to avoid the misfit under periodic boundary conditions. The operation of the $J_1^\perp$ term of H_{XY} on ψ_{4j+3}^a creates only a state like

$$\phi=\uparrow\downarrow\downarrow\uparrow\ldots\uparrow\downarrow|\uparrow\downarrow|\uparrow\downarrow\alpha_{4j+7}\uparrow\downarrow\ldots ,$$

which has an excitation energy $(\frac{5}{4})|J_1-2J_2|$. Thus, the ψ_{4j+3}^a is localized within the first order of $J_1^\perp$. The operation of the $J_1^\perp$ term on $\psi_{2j+1/2}^b$ makes it connected to $\psi_{2j-2+1/2}^b$ and $\psi_{2j+2+1/2}^b$ with the amplitude $J_1^\perp/2$, thereby making the excitation propagate in a chain. The wave function and the energy are given by

$$\psi^b(k)=(2/N)^{1/2}\sum_j \psi_{2j+1/2}^b\exp[ik(2j+1/2)] , \quad (10)$$

$$\varepsilon^b(k)=\varepsilon_0^b+J_1^\perp\cos(2k) . \quad (11)$$

The $J_2^\perp$ term in H_{XY} changes the situation, making the localized excitation propagate by connecting ψ_{4j+3}^a to ψ_{4j+7}^a and ψ_{4j-1}^a with the amplitude $J_2^\perp/2$. The wave function and the energy are thus given within the first order of $J_2^\perp$ by

$$\psi^a(k)=(4/N)^{1/2}\sum_j \psi_{4j+3}^a\exp[ik(4j+3)] , \quad (12)$$

$$\varepsilon^a(k)=\varepsilon_0^a+J_2^\perp\cos(4k) . \quad (13)$$

The operation of the $J_2^\perp$ term on $\psi_{2j+1/2}^b$ creates the state,

$$\phi'=\uparrow\downarrow\downarrow\uparrow\ldots\uparrow\downarrow|\uparrow\uparrow\downarrow\downarrow|\uparrow\downarrow\downarrow\uparrow\ldots ,$$

and a further operation on ϕ' creates

$$\phi''=\uparrow\downarrow\downarrow\uparrow\ldots\uparrow\downarrow|\uparrow\uparrow\downarrow\downarrow\uparrow\uparrow\downarrow\downarrow|\uparrow\downarrow\downarrow\uparrow\ldots ,$$

where ϕ' and ϕ'' have the excitation energy $\varepsilon_0^b(\equiv 2\varepsilon_0^a)$. These modes are interpreted as two ψ^a-type solitons propagating.

These excitations may dominate the behavior of $S_{zz}(Q,\omega)$. The central components of $\omega\sim 0$ are strong for $J_2^\perp=0$ [Fig. 2(b)]. This may be due to a large contribution of the localized ψ^a-type solitons. In the presence of $J_2^\perp$, however, we can clearly see a squarelike shoulder develop due to the propagation of solitons, in agreement with the above analysis.

(3) *Dimer state.* Under the condition that $J_2^\perp/J_1^\perp=J_2/J_1=0.5$, the ground state is exactly expressed as $\psi_g=[1,2][3,4][5,6]\ldots$, where

$$[2j+1,2j+2]=(1/\sqrt{2})(\alpha_{2j+1}\beta_{2j+2}-\beta_{2j+1}\alpha_{2j+2}) ,$$

with the ground-state energy $E_g/(N/2)=-J_1/4-J_1^\perp/2$.

Now we consider a low-lying excited state near this condition,

$$\psi_{2j+1}^c=[1,2][3,4]\ldots T_{2j+1}[2j+3,2j+4]\ldots , \quad (14)$$

with

$$T_{2j+1}=(1/\sqrt{2})(\alpha_{2j+1}\beta_{2j+2}+\beta_{2j+1}\alpha_{2j+2}) .$$

The operation of the Hamiltonian on this state gives

$$(H-E_g)\psi_{2j+1}^c=J_1^\perp\psi_{2j+1}^c+(\tfrac{1}{4})(-J_1+2J_2)(\psi_{2j-1}^c+\psi_{2j+3}^c) , \quad (15)$$

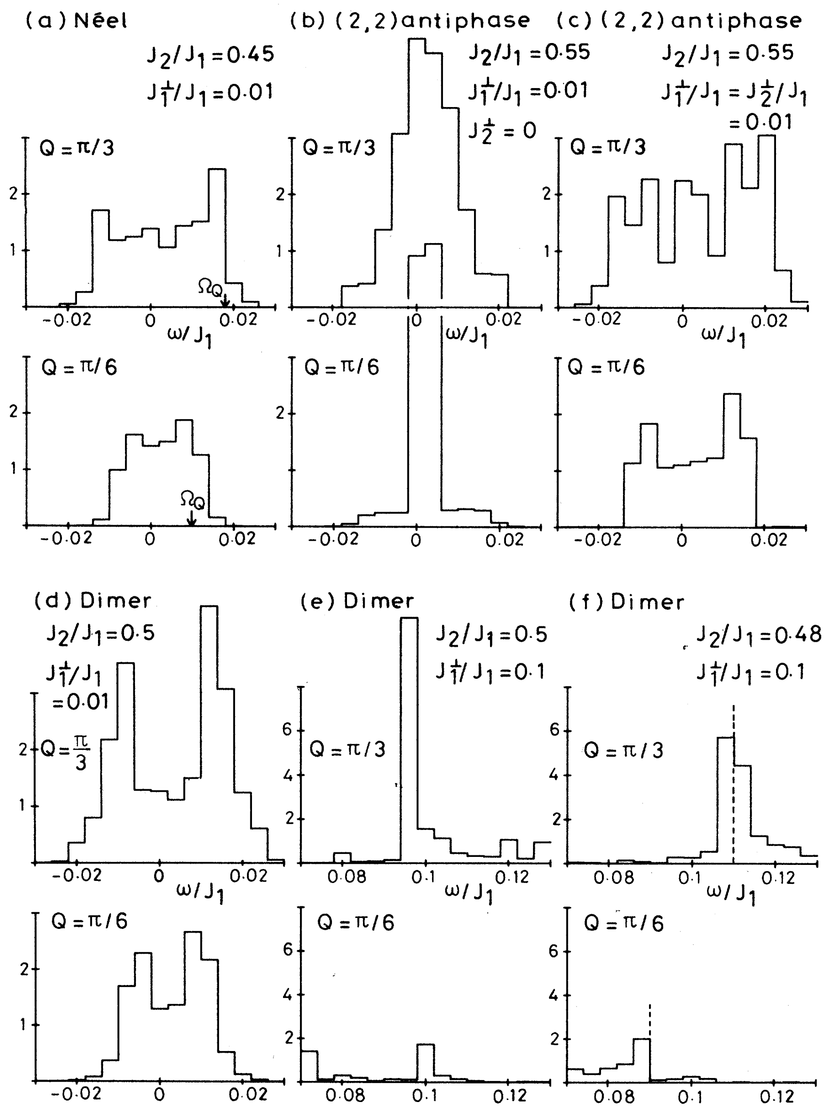

FIG. 2. $S_{zz}(Q,\omega)$ for an $N=12$ chain with T (temperature) $=0.04J_1$. (a) Néel state; (b) and (c) (2,2) antiphase state; (d)–(f) dimer state. Except for (b) and (c), $J_2^\perp/J_1^\perp=J_2/J_1$. The temperature is sufficiently high for solitons and triplet-dimer excitations to be thermally excited in (a)–(d), while it is low in comparison with the excitation energy of the triplet-dimer excitations in (e) and (f). The arrows in (a) represent the position of the Villain mode given by $\Omega_Q=2J_1^\perp|\sin Q|$. Broken lines in (f) represent the energy of the triplet-dimer excitation given by Eq. (17).

where neglected is the state

$$\phi'=[1,2][3,4]\ldots\uparrow\uparrow\downarrow\downarrow[2j+3,2j+4]\ldots$$

because of its high excitation energy. The diagonalization of Eq. (15) yields the wave function and the energy of the triplet-dimer excitation,

$$\psi^c(k)=(2/N)^{1/2}\sum_j \psi^c_{2j+1}\exp[ik(2j+1)]\,, \qquad (16)$$

$$\varepsilon^c(k)=J_1^\perp+(\tfrac{1}{2})(-J_1+2J_2)\cos(2k)\,. \qquad (17)$$

A soliton excitation[2]

$$\phi=[1,2]\ldots[2j-1,2j]\alpha_{2j+1}[2j+2,2j+3]\ldots$$

is irrelevant to the present case because of its high excitation energy of an order J_1.

As for $S_{zz}(Q,\omega)$, strong peaks appear at $\omega=\pm J_1^\perp$, and the intensity increases with increasing Q [Fig. 2(d)]. The peaks are interpreted as a result of the transition caused by S_Q^z from a singlet dimer $[2j+1,2j+2]$ to T_{2j+1} (and vice versa at high temperatures). The dispersion relation of the triplet-dimer excitations is clearly seen in the shift of the peak position with changing Q, for $|J_1-2J_2|\neq 0$ [Figs. 2(e) and 2(f)].

The phase boundary may be determined by the condition that the minimum in the energy band of solitons touches on zero: $\varepsilon_0=J_1^\perp$ for $J_2/J_1<0.5$, and $\varepsilon_0^b=J_1^\perp$ ($\varepsilon_0^a=J_2^\perp$) for $J_2/J_1>0.5$. This condition may be modified by higher-order effects. To study these effects, first we numerically calculate the energy gap Δ_N between the lowest energy in triplet states $(M \equiv \langle \sum_i S_i^z \rangle = 1)$ and the ground-state $(M=0)$ energy for finite chains of up to $N=20$, and then estimate its limiting $(N \to \infty)$ value Δ_∞ by making a least-squares fit of the results for $N=14$, 16, 18, and 20 to a quadratic function of $1/N$, that is, $\Delta_N = \Delta_\infty + c/N + c'/N^2$ with numerical constants c and c'. We determine the phase boundary from the condition that Δ_∞ vanishes. Figure 3 shows Δ_∞ as well as Δ_N for $J_2/J_1 < 0.5$ under the condition $J_2^\perp/J_1^\perp = J_2/J_1$, as a function of $J_1^\perp$. The nonlinear correction to Δ_∞ works in the direction of maintaining Δ_∞, thereby leading to stabilizing the Néel state. (See also Fig. 1.) For $J_2/J_1 > 0.5$, however, we have not been able to estimate reliable values of Δ_∞, not only because finite-size results only for $N=8$, 12, 16, and 20 are available for extrapolations but because the N dependence of these results is not monotonic.[10]

In summary, we have studied low-lying excitations near the degenerate point, and have found that the soliton excitations [in the Néel and (2,2) antiphase states] and the triplet-dimer excitations (in the dimer state) could propagate in the chain, dominating the behavior of $S_{zz}(Q,\omega)$. The ground-state phase diagram is determined by the condition that the energy gap vanishes in the excitation spectrum. We have imposed the condition $J_2^\perp/J_1^\perp = J_2/J_1$ to stabilize the dimer state; the amplitude coming from the $J_2^\perp$ term has an effect to cancel the amplitude of breaking the singlet dimer associated with the $J_1^\perp$ term. (The cancellation is perfect at $J_2^\perp/J_1^\perp = 0.5$.) As concerns the comparison of our present predictions with experimental results, there is, unfortunately, no actual Ising-like spin-$\frac{1}{2}$ system with considerable next-nearest-neighbor interactions. We hope that a new material can be synthesized for experiments in the near future; a zigzag chain of $FeMgBO_4$ has already been synthesized with considerable next-nearest-neighbor interactions, although the relevant spin is $\frac{5}{2}$ and the interactions between Fe^{3+} ions are cut off randomly by Mg^{2+} ions.[14]

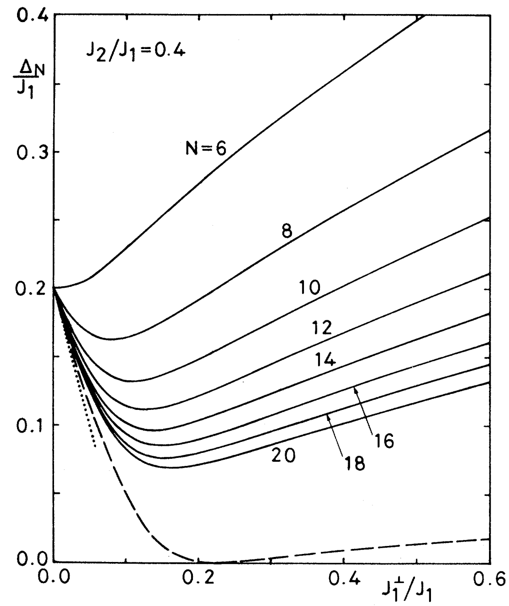

FIG. 3. The energy gap Δ_N as a function of $J_1^\perp$ for $J_2/J_1 (= J_2^\perp/J_1^\perp) = 0.4$. The solid lines show the finite-size results for $N=6, 8, \ldots, 20$, and the dashed line represents the limiting result Δ_∞. The dotted line is the lowest bound of the continuum of two solitons, on the basis of the linear theory [Eq. (7)].

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[1]C. K. Majumdar, J. Phys. C 3, 911 (1969).
[2]B. S. Shastry and B. Sutherland, Phys. Rev. Lett. **47**, 964 (1981).
[3]F. D. M. Haldane, Phys. Rev. B **25**, 4925 (1982).
[4]K. Kuboki and H. Fukuyama, J. Phys. Soc. Jpn. **56**, 3126 (1987).
[5]V. J. Emery and C. Noguera, Phys. Rev. Lett. **60**, 631 (1988).
[6]J. Stephenson, Can. J. Phys. **48**, 1724 (1970).
[7]M. Nagase, T. Nakamura, and J. Igarashi, Prog. Theor. Phys. **56**, 1396 (1976).
[8]It was pointed out that low-lying excitations are expressed as *block excitations* where spins are reversed from the Néel state in blocks with two spins or more, near the degenerate point in the Ising limit. This type of excited state corresponds to the states with many solitons in the present case; J. Igarashi, Prog. Theor. Phys. **57**, 1209 (1977).
[9]N. Ishimura and H. Shiba, Prog. Theor. Phys. **63**, 743 (1980).
[10]T. Tonegawa and I. Harada, J. Phys. Soc. Jpn. **56**, 2153 (1987).
[11]J. Villain, Physica B **79**, 1 (1975).
[12]H. Yoshizawa and K. Hirakawa, J. Phys. Soc. Jpn. **46**, 448 (1979); K. Hirakawa and H. Yoshizawa, *ibid.* **46**, 455 (1979).
[13]H. Yoshizawa, K. Hirakawa, S. K. Satija, and G. Shirane, Phys. Rev. B **23**, 2298 (1981).
[14]A. Wiedenmann, P. Burlet, H. Scheuer, and P. Convert, Solid State Commun. **38**, 129 (1981).