Magnetic Excitations of Spin Nematic State in Frustrated Ferromagnetic Chain

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By exploiting density-matrix renormalization group techniques, we investigate the dynamical spin structure factor of a spin-1/2 Heisenberg chain with ferromagnetic nearest-neighbor and antiferromagnetic next-nearest-neighbor exchange interactions in an applied magnetic field. In a field-induced spin nematic regime, we find gapless longitudinal and gapped transverse spin excitation spectra, in accordance with quasi-long-ranged longitudinal and short-ranged transverse spin correlations, respectively. The gapless point coincides with the dominant longitudinal spin correlation, whereas the gap position exhibits a characteristic field dependence contradicting the dominant transverse spin correlation.

Concerning the experimental realization of the SN state in the frustrated ferromagnetic chain, LiCuVO$_4$ has been studied frequently as a prototypical material. In a field-induced phase, neutron diffraction experiments have found a collinear spin-modulated structure, which is consistent with theoretical results for the longitudinal SDW correlation in the SN/SDW phase. These observations are indeed suggestive of the development of the quadrupole correlation. However, since the direct observation of the non-magnetic SN state is difficult, the order parameter is still not identified yet.

To collect evidence that the SN state occurs in reality, it is important to find any specific indications in what we observe by using various microscopic probes. In this context, recent theoretical studies have pointed out that the NMR relaxation rate shows characteristic temperature and field dependencies in the SN/SDW phase. NMR experiments for LiCuVO$_4$ have reported consistent results with theoretical predictions, signaling the formation of the bound magnon pairs. The properties of the low-energy spin excitation spectra have also been discussed.

In this paper, to clarify the property of the SN/SDW phase from the viewpoint of the spin dynamics, we investigate spin excitation spectra in a wide range of momentum and energy by numerical methods. We clearly find that the longitudinal spin excitation spectrum is gapless, while the transverse one is gapped, as naively expected from the behavior of the spin correlations. We discuss the field dependence of the spectral weight transfer in relation with the dominant spin correlation.

A striking feature is that the momentum position of the gap deviates from that of the dominant transverse spin correlation as the system approaches the saturation.

Let us consider the model (1) in an $N$-site chain, and take $J_2 = 1$ as the energy unit. We investigate the spin excitation dynamics at zero temperature by exploiting density-matrix renormalization group (DMRG) techniques. We employ the finite-system algorithm in open boundary conditions. We compute the dynamical spin structure factor, defined by

$$S^q(q, \omega) = \frac{1}{\pi} \text{Im} \left( \langle \psi_G | S^q_0 | \psi_G \rangle \right) \frac{1}{\omega + E_G - H + i \eta} S^q_0 | \psi_G \rangle,$$

where $| \psi_G \rangle$ is the ground state with eigenenergy $E_G$. Note that $| \psi_G \rangle$ is given by the lowest-energy state in the subspace of a given magnetization $m = M/N$, where $M = \sum_i S^z_i$. For the
calculation of $S^\alpha(q,\omega)$ at $m$, we set the magnetic field to be the midpoint of the magnetization plateau of $M$ in the $N$-site system. $\eta$ is a small broadening factor, and we set $\eta = 0.1$ unless otherwise specified. Our particular interest is to clarify the anisotropy between longitudinal $S^z(q,\omega)$ and transverse $S^\alpha(q,\omega)$ due to the formation of the two-magnon bound states in the field-induced SN/SDW phase.

For the analysis of the spin excitation spectrum, we use a dynamical DMRG method, targeting the ground state $|\psi_0\rangle$, an excited state $S^\alpha|\psi_\eta\rangle$, and the so-called correction vector $[\omega + E_0 - H + i\eta]^{-1}S^\alpha|\psi_\eta\rangle$. Here, we note that the truncation error rapidly increases as the number of target states increases. Therefore, to obtain $S^\alpha(q,\omega)$ with keeping high accuracy, we calculate $S^\alpha(q,\omega)$ and $S^\alpha(q,\omega)$ separately, and then use the relation $S^\alpha(q,\omega) = [S^+(q,\omega) + S^-(q,\omega)]/4$, instead of directly calculating $S^\alpha(q,\omega)$. Note also that we calculate the spectrum at $q$ and $\omega$ after one DMRG run with fixed $q$ and $\omega$, so that we need to perform a great number of DMRG runs to obtain a full spectrum.

Let us start with a brief discussion on the spectrum at zero field. In Fig. 1(a), we present the intensity plot of $S^\alpha(q,\omega)$ at $J_1 = -1$, $J_2 = 1$ (energy unit throughout the paper), $m = 0$, and $h = 0$. Note that $S^\alpha(q,\omega) = S^\alpha(q,\omega)$ at $h = 0$ due to the SU(2) spin rotation symmetry. We find a sinusoidal dispersion that gives the lower boundary of a continuum. The sinusoidal dispersion represents the spinon excitation, described by the des Cloizeaux-Pearson mode, since the system decouples into two antiferromagnetic chains if $J_1 \to 0$. We see a large amount of spectral weight at a lowest-energy peak $(q_0,\omega_0) = (\pi/2, 0.04)$. Note here that the spectrum is asymmetric with respect to $q_0$, as pointed out by the previous studies. That is, the spectral weight mainly lies near the lower boundary of the continuum for $q < q_0$, whereas it is distributed to a high energy region for $q > q_0$. On the other hand, $\omega_0$ coincides with a spin excitation energy,

$$\Delta(N, M) = [E_0(N, M + 1) + E_0(N, M - 1) - 2E_0(N, M)]/2,$$

where $E_0(N, M)$ is the lowest energy of the $N$-site system in the subspace of $M$ at $h = 0$. We find that $\Delta(N,0)$ shifts to lower energy as $N$ increases, and it is extrapolated to almost zero in the limit of $N \to \infty$. Note that an exponentially small gap has been predicted by renormalization group theory, but it is hard to detect such a small gap numerically.

Now, let us look into the longitudinal and transverse spin excitation spectra in the SN/SDW phase. In Figs. 1(b) and 1(c), we show $S^z(q,\omega)$ and $S^\alpha(q,\omega)$, respectively, at $J_1 = -1$, $J_2 = 1$, $m = 0.125$, and $h = 0.649$. For $S^z(q,\omega)$, a lowest-energy peak is at $(q_0,\omega_0) = (0.375\pi, 0.00)$. That is, $q_0$ moves toward small momentum from the position at zero field. $\omega_0$ is nearly zero, indicating a gapless mode for the longitudinal spin excitation. We see that a certain amount of spectral weight is transferred to the origin due to a finite uniform magnetization, which is also indicative of a gapless mode. These gapless points are clearly visible due to the large intensity. Moreover, $S^\alpha(q,\omega)$ seems to be gapless at $q = \pi - q_0$ and $q = \pi$, although we do not observe significant intensity near the possible gapless points in $q > \pi/2$.

In contrast, for $S^\alpha(q,\omega)$, we observe a lowest-energy peak at $(q_0,\omega_0) = (31\pi/64, 0.15)$. That is, $q_0$ remains near $\pi/2$ even in the SN/SDW phase. On the other hand, $\omega_0$ appears to be a finite energy. In fact, $\omega_0$ agrees with the spin excitation energy $\Delta(N, M)$, and it is extrapolated to a finite value in the limit of $N \to \infty$ with $m = M/N$ fixed, indicating a gapped mode for the transverse spin excitation. Note that $S^\alpha(q,\omega)$ consists of $S^-(q,\omega)$ and $S^+(q,\omega)$. As shown in Fig. 2, both spectra have the lowest-energy peak at the same position, while the overall structure is different between them. We find that $S^\alpha(q,\omega)$ is highly dispersive in a wide range of momentum and energy, and $S^+(q,\omega)$ is less dispersive and it is mainly concentrated in a small region near the lowest-energy peak.

Here, let us discuss how the lowest-energy peak position depends on $m$ in the whole range of $m$. In Fig. 3(a), we plot $q_0$ of $S^\alpha(q,\omega)$ as a function of $m$ for several values of $J_1$. Note that we also find a sharp peak at the origin for finite $m$, but
we focus on the field-induced shift of the peak position which originally locates near $\pi/2$ at zero field. At small $m$, where the system is in the VC phase, $q_0$ shows little dependence on $m$. At large $m$, where the system is in the SN/SDW phase, $q_0$ follows the relation $q_0 = (1/2 - m)\pi$ regardless of $J_1$, which supports the bosonization result.\(^{24}\) Note that $q_0$ is represented by the density of bound magnons $1/2 - m$.

In Fig. 3(b), we show the $m$ dependence of $q_0$ of $S^-(q, \omega)$. In the VC phase at small $m$, we notice that $q_0$ agrees with the incommensurability of the transverse spin correlation, as will be shown in Fig. 4(d). Note here that it has been revealed that the incommensurate wave number in the VC phase is strongly quantum renormalized toward $\pi/2$ compared with the pitch angle $\cos^{-1}(-J_1/4J_2)$ of the helical order in the classical spin case.\(^{6}\) Indeed, we observe that $q_0 = \pi/2$ at $J_1 = -0.5$ and $-1$, although the corresponding classical pitch angle is 0.46$\pi$ and 0.42$\pi$, respectively. For small $|J_1|$, we see that $q_0$ remains at $\pi/2$ below a threshold value of $m$ even in the SN/SDW phase at large $m$. However, $q_0$ goes away from $\pi/2$ as $m$ approaches the saturation, and the deviation from $\pi/2$ is more pronounced for larger $|J_1|$. On the other hand, the exact energy dispersion of the one-magnon excited state in the fully polarized state is

$$
ei(q) = J_1(\cos q - 1) + J_2(2\cos 2q - 1) + h,$$

and its minimum is at $q = \cos^{-1}(-J_1/4J_2)$, which coincides with the classical pitch angle.\(^{4}\) The present numerical results at the saturation totally agree with this exact description. We mention that the bosonization analysis shows that the bottom of the one-magnon band is found at $\pi/2$,\(^{24}\) but it is valid only for the weak-coupling regime $|J_1| \ll J_2$ and inapplicable in the limit of $m \to 1/2$. We thus confirm that the bottom of the one-magnon band moves from the quantum renormalized incommensurate wave number to the classical pitch angle as $m$ increases from zero to the saturation. This feature would be useful to determine exchange couplings of real materials.

To gain an insight into the properties of the field-induced spectral weight transfer and the dominant spin correlation, we examine the spin structure factor,

$$S^a(q) = \langle \psi_G | S_q^a S_q^a | \psi_G \rangle,$$

with attention to sum rules on the integrated intensity of the dynamical spin structure factor. To obtain $S^a(q)$, we perform ground-state DMRG calculations independent of dynamical DMRG runs for $S^a(q, \omega)$, since we can obtain accurate results with relatively small computational cost. The sum rule for the energy-integrated intensity reads

$$I^a(q) \equiv \int \frac{d\omega}{2\pi} S^a(q, \omega) = S^a(q),$$

leading to the spin structure factor. As for the total intensity $I^a \equiv \sum q S^a(q)$, we have

$$I^a = I^c = N/4, \quad I^s = N/2 \mp M.$$

In Fig. 4(a), we show $S^z(q)$ for several values of $m$ at $J_1 = -1$. There is a clear peak at $q = \pi/2$ for $m = 0$, and it shifts toward small momentum with increasing $m$, as indicated by vertical arrows. The peak position of $S^z(q)$ representing the SDW correlation is in agreement with the lowest-energy peak position $q_0$ of $S^z(q, \omega)$ in Fig. 3(a). We also find that $S^z(q)$ grows at $q = 0$ as $m$ increases. In other momentum parts, $S^z(q)$ is suppressed so as to keep the total intensity $I^z = N/4$. Accordingly, $S^z(q, \omega)$ exhibits the spectral weight transfer, as seen in Fig. 1(b).

We show $S^z(q)$ at $J_1 = -1$ and $J_1 = -2.5$ in Figs. 4(b) and 4(c), respectively. We find a sharp peak structure in the VC phase at small $m$, while the peak structure becomes broad in the SN/SDW phase at large $m$. This is because the transverse spin correlation exhibits an algebraic decay in the VC phase, and it is short-ranged in the SN/SDW phase. As $m$ increases, the borad peak is levelled off, while the baseline goes up, and eventually we find a completely flat profile, i.e., $S^z(q) = 1/4$, at the saturation. On the other hand, the peak position depends

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**Fig. 3.** (Color online) The lowest-energy peak position $q_0$ of (a) $S^z(q, \omega)$ and (b) $S^-(q, \omega)$ for several values of $J_1$ at $J_2 = 1$ as a function of $m$. Solid symbols denote the VC phase at low fields, and open symbols denote the SN/SDW phase at high fields. Note that $q_0$ shows a stepwise change simply due to finite-size effects. The resolution of the momentum is $2\pi/N$ and we use $N = 128$ in the present calculations. Here, the broadening factor is set to $\eta = 0.02$ to determine the position of the lowest-energy peak precisely.

**Fig. 4.** (Color online) (a) $S^z(q)$ at $J_1 = -1$, (b) $S^z(q)$ at $J_1 = -1$, and (c) $S^z(q)$ at $J_1 = -2.5$ for several values of $m$. (d) Pitch angle $Q$, determined from the maximum point of $S^z(q)$, as a function of $m$. Solid symbols denote the VC phase at low fields, and open symbols denote the SN/SDW phase at high fields. $J_2 = 1$ is fixed. Data are obtained by DMRG calculations with $N = 128$. 

Figs. 3(b) and 4(d). However, in the SN\((q,\omega)\) have a peak at the same position. In the VC phase at small \(m\), \(Q\) agrees with the lowest-energy peak position \(q_0\) of \(S^{-}(q,\omega)\), as denoted by solid symbols in Figs. 3(b) and 4(d). However, in the SN/SDW phase at large \(m\), \(Q\) behaves quite differently from \(q_0\) as \(m\) approaches the saturation. We find that \(Q\) changes toward \(\pi/2\) regardless of \(J_1\), rather than the classical pitch angle \(\cos^{-1}(-J_1/J_2)\) as \(q_0\), indicating that the dominant \(q\) component is not equivalent to the momentum position of the opening gap. This discrepancy is naturally understood in terms of the short-range nature of the transverse spin correlation. It is inadequate to take only the leading asymptotic term that represents the exponential decay of the transverse spin correlation function in order to describe the excitation dynamics correctly. In fact, we have seen that \(S^{+}(q)\) turns into the flat profile at the saturation, meaning that not only the dominant \(q\) component but also other components equally contribute to the spectral weight.

Finally, we present the dependence of the energy gap of the transverse spin excitation in Fig. 5. We extrapolate finite-size data to the thermodynamic limit \(N \rightarrow \infty\) by assuming a linear relation \(\Delta(N,M) = \Delta(m) + a/N\). For \(m \gtrsim 0.1\), we find that \(\Delta(m)\) increases as \(|J_1|\) becomes large for \(J_1 = -0.5\), \(-1\), and \(-1.5\), indicating that the ferromagnetic exchange interaction stabilizes the two-magnon bound state. \(\Delta(m)\) turns to decrease with further increasing \(|J_1|\), shown for \(J_1 = -2\). For small \(m\), \(\Delta(m)\) is rapidly reduced as \(m\) decreases down to the boundary between the SN/SDW and VC phases.

In summary, we have studied the spin excitation dynamics of the frustrated ferromagnetic chain in the magnetic field by numerical methods. In the field-induced SN regime, the spin excitation spectra exhibit highly anisotropic behavior between gapless longitudinal and gapped transverse components. The field dependence of the gapless point of \(S^{-}(q,\omega)\) is consistent with the dominant longitudinal spin correlation. In contrast, the gap position of \(S^{+}(q,\omega)\) shows a unique field dependence that contradicts with the dominant transverse spin correlation. We hope that these features could be examined by inelastic neutron scattering experiments when searching for signatures of the SN/SDW phase. On the other hand, we should observe gapless excitations in the quadrupole channel. It would be an interesting future problem to study the quadrupole excitation dynamics and its possible relevance to observables.\(^{34}\)

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Fig. 5. (Color online) The extrapolated gap \(\Delta(m)\) for several values of \(J_1\) at \(J_2 = 1\) as a function of \(m\). Here we plot data in the SN/SDW phase. Note that the zero-field gap is tiny,\(^{33}\) and the VC phase is supposed to be gapless.

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