Vector chirality and inhomogeneous magnetization in frustrated spin tubes in high magnetic fields

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The low-energy physics of three-leg frustrated antiferromagnetic spin-\(S\) tubes in the vicinity of the upper critical field is studied. Utilizing the effective field theory based on the spin-wave approximation, we argue that in the intermediate-interchain-coupling regime, the ground state exhibits a vector chiral order or an inhomogeneous magnetization for the interchain (rung) direction and the low-energy excitations are described by a one-component Tomonaga-Luttinger liquid (TLL). In both chiral and inhomogeneous phases, the \(Z_2\) parity symmetry along the rung direction is spontaneously broken. It is also predicted that a two-component TLL appears and all the symmetries are restored in the strong-rung-coupling case.

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I. INTRODUCTION

Frustrated spin systems have been continuously explored for more than five decades. Frustration is considered as an important keyword to generate exotic, unconventional magnetic orders, disorders and excitations including even spin-liquid states. Actually, frustrated systems have provided several peculiar concepts and phenomena so far: resonating-valence-bond picture, noncollinear orders, symmetry-unrelated degeneracy, order-by-disorder mechanism, etc.

In recent years, frustrated magnets containing four-spin exchanges as well as standard two-spin ones have been intensively studied. In such magnets, fascinating magnetic orders (nematic, chiral, dimer orders, etc.), which order parameter is defined by products of spin operators, are shown to be present. In a sense, these new orders are a natural consequence of the four-spin exchange because for such an interaction, it is possible to perform a mean-field approximation, \(S_i^αS_j^βS_k^γS_l^δ\rightarrow (S_i^αS_j^βS_k^γ)S_l^δ + S_i^αS_j^β(S_k^γS_l^δ) - (S_i^αS_k^γS_j^β)S_l^δ - (S_i^αS_j^βS_l^δ)S_k^γ\). Furthermore, it is well known that effects of four-spin exchanges are fairly small in a large number of real magnets. Thus, to discover intriguing magnetic orders within spin systems containing only two-spin exchanges could more stimulate many experimentalists and would be theoretically a more challenging issue.

In one dimension, as representatives of geometrically frustrated spin systems with only two-spin exchanges, one can consider zigzag spin chains and three-leg antiferromagnetic (AF) spin tubes, i.e., ladders with a periodic boundary condition (PBC) along the interchain (rung) direction. In this paper, we study the latter model in a magnetic field. The Hamiltonian is written as

\[
H = \sum_{i=1}^{3} \sum_{j} \left[ J_{l} \vec{S}_{i,j} \cdot \vec{S}_{i,j+1} + J_{\perp} \vec{S}_{i,j} \cdot \vec{S}_{i,j+1,j} - HS_{i,j} \right],
\]

where \(\vec{S}_{i,j}\) is spin-\(S\) operator on site \(j\) of the \(l\)th chain \((l = 1, 2, 3)\), \(J > 0\) \((J_{\perp} > 0)\) is the intrachain (interchain) coupling, and the PBC \(\vec{S}_{4,j} = \vec{S}_{1,j}\) is imposed. Focusing on the vicinity of the upper critical field and applying an effective field theory approach, we show the possibility of two interesting long-range-ordered states: for a certain high-magnetic-field area, a vector chirality \(\langle V_{i,j}^z \rangle = \langle (\vec{S}_{i,j} \times \vec{S}_{i,j+1,j})^z \rangle\) or an inhomogeneous magnetization along the rung direction occurs in a one-component Tomonaga-Luttinger-liquid (TLL) state. In the chiral phase, the \(Z_2\) rung-parity symmetry \(S_{i,j}^α \leftrightarrow S_{i,j+1,j}^α\), by which \(V_{i,j}\) changes its sign, is spontaneously broken, while the inhomogeneous magnetization in another phase breaks the one-site translational symmetry for the rung as well as the rung-parity one. We also predict that a two-component TLL emerges and all the symmetries are preserved in the strong-rung-coupling regime. Recently a spin tube material \([[CuCl_2]tachH]_3Cl]_2\) (Ref. 3) has been synthesized and its magnetic properties could be described by a three-leg frustrated spin-tube model. This also promotes the motivation of studying the spin tube model.

Existing results of the model \([1]\) are summarized here. In the \(S = \frac{1}{2}\) case, the zero-field ground states are gapped and doubly degenerate with spontaneously breaking the one-site translational symmetry along the chain, at least when \(J_{\perp} \gtrsim 0.5J_{l}\). In addition, a semi-quantitative ground-state phase diagram in the \(J_{\perp}-H\) plane \((J_{\perp} > 0)\), which only shows gapless and gapful regimes, is constructed in Ref. 5; there exists an intermediate magnetization plateau with \(M = \langle S_{i,j}^z \rangle = 1/6\). In the case of \(S = \) integer and \(H = 0\), the system is predicted to be always gapful and to conserve all symmetries.\(^2\)

Before analyzing the quantum spin tube \([1]\), to discuss its classical version is instructive. The classical ground state is an umbrella structure as in Fig. \(\cite{1}\). In this state, symmetries of the \(U(1)\) spin rotation around the spin \(z\) axis, one-site translations, and parity transformations along both the chain and the rung directions are all broken. Consequently, the system exhibits a finite vector...
chirality \( \langle V_{i,j} \rangle = \frac{\sqrt{3}}{2} (1 - \frac{H^2}{2S(4J + 3J_\perp)^2}) \). From this result, the vector chiral order is expected to exist even in the quantum version. However, since generally quantum fluctuation is quite strong in one dimension and tends to destroy any ordering, it is nontrivial whether or not the chiral order remains and broken symmetries are restored in the model (1).

II. EFFECTIVE THEORY

Here we construct the effective theory for the quantum spin tube (1) in a high magnetic field. Let us begin with the fully polarized state with \( M = S \). For the state, the energy dispersion of one magnon with \( \Delta S^2 = -1 \) is exactly calculated as

\[
\epsilon_K(k) = H - 2S(J + J_\perp) + 2SJ \cos k + 2SJ_\perp \cos K, \tag{2}
\]

where \( K = 0, \pm \frac{2\pi}{3} \) is the wave number for the rung and that for the chain, \( k \), is in \( |k| < \pi \). The lowest bands \( \epsilon_{\pm,0} \) are always degenerate due to the rung-parity symmetry, transformation of which induces \( K \rightarrow -K \).

As we explain in Fig. 2 when \( H \) becomes lower than the upper (lower) critical value \( H_c^u = 4SJ + 3SJ_\perp \) (\( H_c^d = 3SJ_\perp \)), magnons of the lowest bands become condensed (are fully condensed). Moreover, as \( H < H_c^u = 4SJ \), magnons in the remaining band \( \epsilon_0 \) are also condensed.

Supposing that multimagnon bound states are absent or their excitation energies are higher than those of one magnon states (this is highly expected in antiferromagnetic systems and we have numerically verified it near the saturation), we may describe the low-energy physics around \( H \sim H_c^u \) using one-magnon excitations. A suitable method for such a description is spin-wave theory (1/S expansion). It makes spins bosonize as

\[
S_{i,j}^z = S - n_{i,j}, \quad S_{i,j}^- = b_{i,j}^\dagger \sqrt{2S - n_{i,j}}, \tag{3}
\]

where \( b_{i,j} \) is the magnon annihilation operator, and \( n_{i,j} = b_{i,j}^\dagger b_{i,j} \) denotes the magnon number. Substituting Eq. (3) in the model (1) and introducing the Fourier transformation of \( b_{i,j} \) for the rung as

\[
b_{i,j} = \frac{1}{\sqrt{3}} \sum_{K=0,\pm2\pi/3} e^{iKl} b_{K,j}, \tag{4}
\]

we obtain the bosonic spin-wave Hamiltonian. As expected, the bilinear part of \( b_{K,j} \) reproduces the free-spin-wave dispersion \( \epsilon_K(k) \). In order to study the low-energy and long-distance properties of the spin tube, we further introduce continuous boson fields \( \Psi_{\pm} \) as follows:

\[
\bar{b}_{0,j} \rightarrow (-1)^j \sqrt{a_0} \Psi_{0}(x), \quad \bar{b}_{\pm,2\pi-j} \rightarrow (-1)^j \sqrt{a_0} \Psi_{\pm}(x), \tag{5}
\]

where \( a_0 \) is the lattice spacing, and \( x = j a_0 \). Using these and taking into account the magnon interaction terms up to the lowest order of the 1/S expansion, we arrive in the following effective Hamiltonian,

\[
\mathcal{H}_{\text{eff}} = \int dx \sum_{q=0,\pm \pi} \left[ \frac{1}{2m_q} \partial_x \Psi_{\pm}(x) - \mu_{q} \Psi_{\pm}(x) \right]
+ g_0 \rho_0^\dagger \rho_0 + g_1 (\rho_+ + \rho_-)^2 + f_0 \rho_0 (\rho_+ + \rho_-) + f_1 (\rho_+ + \rho_-)
+ \lambda_0 (\Psi_{0}^\dagger \Psi_0^\dagger \Psi_{\pm}^\dagger + \text{h.c.})
+ \lambda_1 (\Psi_{0}^\dagger \Psi_0^\dagger \Psi_{\pm}^\dagger + \text{h.c.}) + \cdots, \tag{6}
\]

where \( \rho_q = \Psi_q \Psi_q^\dagger \) is the magnon-density field. (This Hamiltonian can also be derived via the path-integral approach (2)). The first two terms correspond to the free-spin-wave part, and if the chemical potential \( \mu_q \) is positive, the magnon \( \Psi_q \) is condensed. We set \( m_q = 4SJ - H \) and \( \mu_\pm = \mu = S(4J + 3J_\perp) - H \) so that \( H_c^u = 4SJ \) and \( H_c^d \) are fixed. Other parameters in Eq. (6) are evaluated as \( 1/m_q = 2SJ_0^2 \) (\( m_q = m \)), \( g_0 = 2J_0 a_0/3 \), \( g_1 = (4J + 3J_\perp) a_0/6 \), \( f_0 = 8J_0 a_0/3 \), \( f_1 = 4J_0 a_0/3 \), \( \lambda_0 = (8J - 3J_\perp) a_0/6 \), and \( \lambda_1 = (16J - 3J_\perp) a_0/12 \). These values would be somewhat changed due to high-energy modes, the curvature of the dispersion, higher-order interactions, and the hard-core property of magnons neglected in the spin-wave theory.

III. LOWEST-BAND-MAGNON CONDENSED STATE

Based on the effective theory (6), we investigate the spin tube near saturation. In this section, we consider the lowest-magnon-condensed case, where \( \mu_0 > 0, \mu_\pm < 0 \), and \( \max[H_c^u, H_c^d] < H < H_c^u \). For this case, the low-energy physics must be governed by two condensed fields \( \Psi_{\pm} \). The effective theory is derived by integrating out the massive magnon \( \Psi_{0} \) via the cumulant expansion in terms of the free-spin-wave part of \( \Psi_{0} \) in the partition
function. The main effect of the $\Psi_0$ sector is that an attractive interaction between $\rho_+ \pi$ and $\rho_- \pi$ originates from the second cumulant of the $\lambda_0$ term. As a result, the coupling constant $f_1$ is changed as

$$ f_1 \rightarrow \tilde{f}_1 = f_1 - C \frac{\chi^2 \phi_0^2}{\sqrt{|m|\mu_0^3}} \quad (7) $$

where $C$ is a positive dimensionless constant of $O(1)$. Here, we have approximated the Matsubara Green’s function $\langle T_\tau \Psi_0(x, \tau) \Psi_0^\dagger(0,0) \rangle$ as $1/a_0$ (zero) when $|x|$ and $J a_0 \tau$ are smaller (larger) than the correlation length $\langle m \mu_0 \rangle^{-1/2}$ [$\tau$: imaginary time], and assumed that $\langle m \mu_0 \rangle^{-1/2}$ is at most $O(a_0)$. For the resultant Hamiltonian $H'_\text{eff}[\psi_\pm]$, the Haldane’s harmonic-fluid approach (i.e., bosonization) (Refs. [13][14][15]) could be applicable. Using the bosonization formulas $\rho_\pm(x) \approx (\tilde{\rho}_\pm + \partial_\mu \phi_\pm/\pi)\sum_{n=-\infty}^{\infty} e^{i2n(x - \pi \rho_\pm x)}$ and $\Psi_\pm \sim (\tilde{\rho}_\pm + \partial_\mu \phi_\pm/\pi)\sum_{n=-\infty}^{\infty} e^{i2n(x - \pi \rho_\pm x)}e^{-i n^2 \pi^2}$, where $\tilde{\rho}_\pm = \langle \rho_\pm \rangle$, we obtain a bosonized Hamiltonian of the phase fields $\phi_\pm, \theta_\pm$. Introducing the new fields $\phi_{a,s} = (\phi_\pm + \phi_\pm)/\sqrt{2}$ and $\theta_{a,s} = (\theta_+ \pm \theta_\mp)/\sqrt{2}$ further, we can represent the phase-field Hamiltonian as

$$ H_{\phi, \theta} = \int dx \sum_{s=q,a} \frac{v_s}{2\pi} \left[ K_q (\partial_\theta \phi_q)^2 + K_{a}^{-1} (\partial_\phi \theta_q)^2 \right] 
+ g_\phi \cos(2\sqrt{2}\phi_a) + g_\theta \cos(3\sqrt{2}\theta_a) + \cdots, \quad (8) $$

where we have assumed $\tilde{\rho}_+ = \tilde{\rho}_- = \tilde{\rho}$ (see below) and dropped terms with spatially oscillating factors $e^{i2n\tilde{\rho}x}$. The $g_\phi$ and $g_\theta$ terms for example originate from $\rho_+ \pi$ and the third cumulant, respectively. Unfortunately, the values of $g_\phi, \theta$ cannot be evaluated quantitatively within the present approach.

In the phase-field picture, a spin rotation around the $S^z$ axis $S_{i,j}^+ \rightarrow e^{i\gamma} S_{i,j}^+$, the one-site translation along the chain $S_{i,j}^0 \rightarrow S_{i,j+1}^0$, and along the rung $S_{i,j} \rightarrow S_{i+1,j}^0$, and the site-parity transformation along the chain $S_{i,j}^0 \rightarrow S_{i,j+1}^0$ are, respectively, expressed as $\theta_\pm \rightarrow \theta_\pm + \gamma$, $\phi_\pm(x, \theta_\pm(x)) \rightarrow (\phi_\pm(x + a) - \pi \rho_\pm a, \phi_\pm(x + a_0) - \pi \rho_\pm a_0, \theta_\pm(x + a_0)), \theta_\pm \rightarrow \theta_\pm \pm 2\pi/3; \text{ and } \phi_\pm(x, \theta_\pm(x)) \rightarrow (\tilde{\rho}_\pm(x, \theta_\pm(x)) \rightarrow (\tilde{\rho}_\pm(x) \pm \theta_\pm(x))$.

Further, the rung-parity transformation $S_{i,j}^0 \rightarrow S_{\tilde{\imath},j}^0$ may be realized by $\tilde{\rho}_+ = \tilde{\rho}_-$ and $\phi_\pm, \theta_\pm \rightarrow \phi_\mp, \theta_\mp$. Owing to these symmetries, in all vertex operators without oscillating factors, only $\cos[2n(\phi_+ + \phi_-)]$ and $\cos[3n(\theta_+ - \theta_-)]$ are allowed to exist in Eq. (8). The most relevant $n = 1$ terms indeed appear in Eq. (8).

The bosonization approach for $H'_{\text{eff}}$ evaluates the velocity $v_\phi$ as $v_\phi \approx (\tilde{f}_1 \tilde{\rho}/m)^{1/2}$. Therefore, if $f_1 > 0$, then $v_\phi$ becomes imaginary and it means that the bosonization is invalid. To understand the physical meaning of this instability, [16] we should consider the magnon-density part in $H'_{\text{eff}}$ and then define the following Ginzburg-Landau (GL) potential:

$$ F = g_1 (\rho_+ + \rho_-)^2 + \tilde{f}_1 \rho_+ \rho_- - \mu (\rho_+ + \rho_-). \quad (9) $$

It is clear that as $\tilde{f}_1 > 0$, the potential is minimized by imposing $\rho_+ \neq \rho_-$. Moreover, it is found that

$$ \rho_+ - \rho_- \sim b_{i,j}^\dagger b_{i,j} - b_{i,j}^\dagger b_{i,j} \sim \sum_{l=1}^3 \tilde{V}_{i,j}. \quad (10) $$

We thus conclude that for $f_1 > 0$, a finite long-range vector chiral order $\langle \tilde{V}_{i,j} \rangle$ exists, and the rung-parity symmetry is spontaneously broken. For $J_\perp \ll J$ (i.e., $\mu_0/J \ll 1$) or $J_\perp \gg J$ [i.e., $\lambda_0 \sim -O(J_\perp)$], $f_1 < 0$ generally holds [17][18] while for $J_\perp \sim O(J)$ (i.e., $\lambda_0 \sim 0$), when $H$ becomes closer to $H', \tilde{f}_1$ increases and tends to be positive. Consequently, the chiral phase is present in an intermediate-rung-coupling regime. Supposing that $\rho_+ > \rho_- \pi$ holds in the chiral phase, we can speculate that the $\Psi_\pi$ mode constructs a massive spectrum, whereas the $\Psi_\pi$ part provides a TLL state. Namely, the coexistence of the chiral order and the TLL is predicted.

The presence of the TLL is also supported by the previous study in Ref. [8]. When $H \sim H'$, the TLL parameter would be close to the universal value 1. The correlation function of the chirality might exhibit a power decay: $\langle \tilde{V}_{i,j} \tilde{V}_{i,j} \rangle \sim \tilde{V}_{i,j} \tilde{V}_{i,j} \sim \cdots + \cdots$. This result implies that the ground states possess the sixfold degeneracy. To investigate the physical meaning of locking $\theta_a$ and the ground-state degeneracy, let us focus on the magnetization per site. The bosonization represents it as

$$ \langle S_{i,j}^z \rangle \approx -\frac{2}{3a_0} \left[ \cos \left( \sqrt{2} \theta_a \mp \frac{4}{3} \pi l \right) \right] + \cdots. \quad (11) $$

One can see that the second term in Eq. (11) causes a down-down magnetization structure in the case of $g_0 > 0$, while for $g_0 < 0$ an up-up-down structure occurs: for instance, if $\theta_a$ is locked to zero for $g_0 < 0$, then $\langle S_{i,j}^z \rangle = -\frac{2}{3a_0} \left[ \cos \left( \sqrt{2} \theta_a \pm \frac{4}{3} \pi l \right) \right] + \cdots$.
this inhomogeneous distribution. The meaning of the remaining twofold degeneracy is unknown. Remarkably, the inhomogeneously magnetized phase is not at all expected from the classical tube system (see Fig. 1). We note that this inhomogeneous distribution might slightly be modified if \( \cos(3n\sqrt{2}\theta_a) \) with \( n \geq 2 \) are also relevant. From the predictions of the chiral order for \( f_1 > 0 \) and the inhomogeneous phase under the condition \( f_1 < 0 \) and \( |f_1| \sim 0 \), the boundary \( f_1 = 0 \) is expected to be a first-order transition.

When \(-f_1/\bar{\rho}\) increases so that \( K_a < 9/4, \cos(3\sqrt{2}\theta_a) \) becomes irrelevant and the low-energy physics of the \( \phi_a \) sector is described by a Gaussian model. This transition must be of a Beresinskii-Kosterlitz-Thouless (BKT) type. After the transition, the system is in a two-component TLL phase with all symmetries enjoying. If \(-f_1/\bar{\rho}\) is further increased due to the growth of \( J_\perp \) or the decrease of \( \bar{\rho} \), \( \cos(2\sqrt{2}\phi_a) \) seems to become relevant. However, the exact results for the integrable Bose gas imply that in a one-dimensional Bose system with a short-range repulsive interaction, the TLL parameter is not usually smaller than 1 even when the interaction becomes extremely strong. The two-component TLL is hence expected to continue even when \( J_\perp \gg J \) or \( \bar{\rho} \) is small (see the Endnote [17]). The prediction of the two-component TLL in the strong-rung-coupling regime is in agreement with a previous study applying the strong-rung-coupling approach to the \( S = \frac{1}{2} \) tube.

IV. THREE-BAND-MAGNON CONDENSED STATE

Here, we consider the case where all three kinds of magnons \( \Psi_{+,+,0} \) are condensed. This situation could be realized under the condition of \( \mu > 0, \mu_0 > 0, H^c_l < H < H^c_r \), and \( J_\perp < 4J/3 \). This means that the three-band-magnon condensed state is allowed to exist only in the weak-rung-coupling regime. Like Eq. [2], let us introduce the GL potential for the present case as follows:

\[
G = g_0 \rho_0^2 + g_1 (\rho_+ + \rho_-)^2 + f_0 \rho_0 (\rho_+ + \rho_-) + f_1 \rho_+ \rho_- - \mu_0 \rho_0 - \mu (\rho_+ + \rho_-).
\] (12)

To find the stable magnon-density profile \((\rho_0, \rho_+, \rho_-)\), the Hessian matrix \( H_{ij} = \left[ \frac{\partial^2 G}{\partial \rho_i \partial \rho_j} \right] \) is useful. At the local minimum point \((\rho_0, \bar{\rho}, \bar{\rho})\) satisfying \( \partial G / \partial \rho_i = 0 \), the eigenvalues of \( H_{ij} \) are \(-4J/3, C_1, \) and \( C_2 \) \((-4J/3 < C_1 < 0 \) and \( C_2 > 0 \)). The corresponding eigenvectors are \((\delta \rho_0, \delta \rho_+, \delta \rho_-) \propto (0, 1, -1), (-C_3, 1, 1), \) and \( (C_3, 1, 1), \) where \( C_3 > 0 \). The negative eigenvalue \(-4J/3 \) and its eigenvector indicate that the ground state takes \( \rho_+ = \rho_- \) (\( \neq 0 \)). Moreover, a positive eigenvalue \( C_2 \) implies the existence of the TLL. We therefore predict that the chiral order \((\rho_+ \neq \rho_-)\) and a one-component TLL state still remain when the system moves from the lowest-magnon-condensed regime to all-magnon-condensed one. At

FIG. 3: Schematic ground-state phase diagram of the \( S = \frac{1}{2} \) spin tube. The area away from the saturation is discussed elsewhere (Ref. [24]). See the Endnotes [12] and [17].

V. SUMMARY AND DISCUSSIONS

We have studied the three-leg frustrated spin tube near the upper critical field. It has been predicted that the vector chiral order or the inhomogeneously magnetized order emerges in the magnetic-field-driven TLL phase in the intermediate-rung-coupling regime. It is remarkable that in these two phases, the TLL criticality (massless modes) and the spontaneous breakdown of discrete parity or translational symmetries for the rung direction coexist. We have also shown that when the rung coupling becomes strong enough, the inhomogeneous phase vanishes and instead the two-component TLL occurs with preserving all the symmetries.

Combining our results and the existent ones, we can draw the ground-state phase diagram for the \( S = \frac{1}{2} \) tube as in Fig. 3. The global phase structure near the saturation would common to all the cases with arbitrary \( S \), as far as \( S \lesssim O(1) \). Although in general the spin-wave approach used in this paper is not very reliable for small-\( S \) cases, we believe that it is valid if we consider the region where \( M \) is sufficiently close to the saturation value: in such a region, multimagnon scattering processes are expected to be negligible. When \( J_\perp \) is changed from +0 to +\( \infty \) with \( M \) fixed near the saturation, the following scenario is expected: TLL plus chirality \( \rightarrow \) [first-order transition] \( \rightarrow \) TLL plus inhomogeneous magnetization \( \rightarrow \) [BKT transition] \( \rightarrow \) two-component TLL.

We finally note that the predicted first-order and BKT transitions could not be detected by observing the magnetization \( M \) because \( H \) couples to \( \partial \rho_0 \) and \( \rho_+ + \rho_- \), but it does not directly interact \( \phi_i \) and \( \rho_+ + \rho_- \). A specific-heat measurement would be efficient in the detection.
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