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

Masahiro Sato and Tôru Sakai

Synchrotron Radiation Research Center, Japan Atomic Energy Agency, Sayo, Hyogo 679-5148, Japan and CREST Japan Science and Technology Agency, Kawaguchi, Saitama 332-0012, Japan (Received 21 November 2006; published 10 January 2007)

The low-energy physics of three-leg frustrated antiferromagnetic spin-S tubes in the vicinity of the upper critical field are 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 systems¹ 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 spinliquid states. Actually, frustrated systems have provided several peculiar concepts and phenomena so far: resonatingvalence-bond picture, noncollinear orders, symmetryunrelated 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.² 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 $S_i^{\alpha}S_j^{\beta}S_k^{\gamma}S_l^{\delta} \rightarrow \langle S_i^{\alpha}S_j^{\beta} \rangle S_k^{\gamma}S_l^{\delta} + S_i^{\alpha}S_j^{\beta} \langle S_k^{\gamma}S_l^{\delta} \rangle - \langle S_i^{\alpha}S_j^{\beta} \rangle \langle S_k^{\gamma}S_l^{\delta} \rangle$. 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 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

$$\mathcal{H} = \sum_{l=1}^{5} \sum_{j} \left[J \vec{S}_{l,j} \cdot \vec{S}_{l,j+1} + J_{\perp} \vec{S}_{l,j} \cdot \vec{S}_{l+1,j} - H S_{l,j}^{z} \right], \quad (1)$$

where $S_{l,j}$ 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 inter-

esting long-range-ordered states: for a certain high-magneticfield area, a vector chirality $\langle \mathcal{V}_{l,j}^z \rangle = \langle (\vec{S}_{l,j} \times \vec{S}_{l+1,j})^z \rangle$ 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 $S_{l,j}^{\alpha} \leftrightarrow S_{l+1,j}^{\alpha}$, by which $\mathcal{V}_{l,j}^{\alpha}$ 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 twocomponent TLL emerges and all the symmetries are preserved in the strong-rung-coupling regime. Recently a spin tube material [(CuCl₂tachH)₃Cl]Cl₂ (Ref. 3) has been synthesized, and its magnetic properties could be described by a three-leg frustrated spin-tube model.⁴⁻⁶ This also promotes the motivation of studying the spin tube (1).

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} $\geq 0.5J^{.7}$ In addition, a semiquantitative ground-state phase diagram in the J_{\perp} -H plane ($J_{\perp} > 0$), which only shows gapless and gapful regimes, is constructed in Ref. 8; there exists an intermediate magnetization plateau with $M = \langle S_{l,j}^z \rangle = 1/6$. In the case of S=integer and H=0, the system is predicted to be always gapful and to conserve all symmetries.⁹

Before analyzing the quantum spin tube (1), to discuss its classical version is instructive. The classical ground state is



FIG. 1. (Color online) Classical ground state of the spin tube (1).

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FIG. 2. (Color online) Magnon bands in Eq. (2).

an umbrella structure as in Fig. 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 $\langle \mathcal{V}_{l,j}^z \rangle = \frac{\sqrt{3}}{2} \left(1 - \frac{H^2}{S^2(4J+3J_\perp)^2} \right)$. 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 $\Delta S^z = -1$ is exactly calculated as

$$\epsilon_K(k) = H - 2S(J + J_\perp) + 2SJ\cos k + 2SJ_\perp\cos K, \quad (2)$$

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 $\epsilon_{\pm 2\pi/3}$ are always degenerate due to the rung-parity symmetry, the transformation of which induces $K \rightarrow -K$. As we explain in Fig. 2, when *H* becomes lower than the upper (lower) critical value $H_c^u = 4SJ + 3SJ_{\perp}$ ($H_c^l = 3SJ_{\perp}$), magnons of the lowest bands begin to condense (are fully condensed). Moreover, as $H < H_c' = 4SJ$, magnons in the remaining band ϵ_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

$$S_{l,j}^{z} = S - n_{l,j}, \quad S_{l,j}^{-} = b_{l,j}^{\dagger} \sqrt{2S - n_{l,j}},$$
 (3)

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

$$b_{l,j} = \frac{1}{\sqrt{3}} \sum_{K=0,\pm 2\pi/3} e^{iKl} \tilde{b}_{K,j},$$
(4)

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

$$\tilde{b}_{0,j} \to (-1)^j \sqrt{a_0} \Psi_0(x), \quad \tilde{b}_{\pm 2\pi/3,j} \to (-1)^j \sqrt{a_0} \Psi_{\pm}(x),$$
 (5)

where a_0 is the lattice spacing and $x=ja_0$. 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:

$$\begin{aligned} \mathcal{H}_{\rm eff} &= \int dx \sum_{q=0,+,-} \left[\frac{1}{2m_q} \partial_x \Psi_q^{\dagger} \partial_x \Psi_q - \mu_q \rho_q \right] + g_0 \rho_0^2 \\ &+ g_1 (\rho_+ + \rho_-)^2 + f_0 \rho_0 (\rho_+ + \rho_-) + f_1 \rho_+ \rho_- \\ &+ \lambda_0 (\Psi_0^2 \Psi_+^{\dagger} \Psi_-^{\dagger} + \text{H.c.}) + \lambda_1 (\Psi_+^2 \Psi_0^{\dagger} \Psi_-^{\dagger} + \Psi_-^2 \Psi_0^{\dagger} \Psi_+^{\dagger} \\ &+ \text{H.c.}) + \cdots, \end{aligned}$$
(6)

where $\rho_q = \Psi_q^{\dagger} \Psi_q$ is the magnon-density field. (This Hamiltonian can also be derived via the path-integral approach.¹⁰) The first two terms correspond to the free-spin-wave part, and if the chemical potential μ_q is positive, the magnon Ψ_q is condensed.¹¹ We set $\mu_0 = 4SJ - H$ and $\mu_{\pm} = \mu = S(4J + 3J_{\perp})$ -H so that H_c^u and H_c' are fixed. Other parameters in Eq. (6) are evaluated as $1/m_q = 2SJa_0^2$ ($m_q = m$), $g_0 = 2Ja_0/3$, $g_1 = (4J + 3J_{\perp})a_0/6$, $f_0 = 8Ja_0/3$, $f_1 = 4Ja_0/3$, $\lambda_0 = (8J - 3J_{\perp})a_0/6$, 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 lowestmagnon-condensed case, where $\mu > 0$, $\mu_0 < 0$, and $\max[H_c^l, H_c^\prime] < H < H_c^u$. For this case, the low-energy physics must be governed by two condensed fields Ψ_{\pm} . The effective theory is derived by integrating out the massive magnon Ψ_0 via the cumulant expansion in terms of the free-spin-wave part of Ψ_0 in the partition function. The main effect of the Ψ_0 sector is that an *attractive* interaction between ρ_+ and $\rho_$ originates from the second cumulant of the λ_0 term. As a result, the coupling constant f_1 is changed as

$$f_1 \to \tilde{f}_1 = f_1 - C \frac{\lambda_0^2 a_0^{-2}}{\sqrt{m|\mu_0|^3}},$$
 (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 $Ja_0\tau$ are smaller (larger) than the correlation length $(m|\mu_0|)^{-1/2}$ $(\tau: \text{ imaginary time})$ and assumed that $(m|\mu_0|)^{-1/2}$ is at most $O(a_0)$.¹² For the resultant Hamiltonian $\mathcal{H}'_{\text{eff}}[\Psi_{\pm}]$, the Haldane's harmonic-fluid approach (i.e., bosonization) (Refs. 13–15) could be applicable. Using the bosonization formulas $\rho_{\pm}(x) \approx \{\bar{\rho}_{\pm} + \partial_x \phi_{\pm}/\pi\} \sum_{n=-\infty}^{\infty} e^{i2n(\phi_{\pm} - \pi \bar{\rho}_{\pm} x)}$ and $\Psi_{\pm}^{\dagger} \sim \{\bar{\rho}_{\pm} + \partial_x \phi_{\pm}/\pi\}^{1/2} \sum_{n=-\infty}^{\infty} e^{i2n(\phi_{\pm} - \pi \bar{\rho}_{\pm} x)} e^{-i\theta_{\pm}}$, where $\bar{\rho}_{\pm} = \langle \rho_{\pm} \rangle$, we obtain a bosonized Hamiltonian of the phase fields $(\phi_{\pm}, \theta_{\pm})$. Introducing the new fields $\phi_{s,a} = (\phi_{+} \pm \phi_{-})/\sqrt{2}$ and $\theta_{s,a} = (\theta_{+} \pm \theta_{-})/\sqrt{2}$ further, we can represent the phase-field Hamiltonian as

$$\mathcal{H}_{[\phi,\theta]} = \int dx \sum_{q=s,a} \frac{v_q}{2\pi} [K_q (\partial_x \theta_q)^2 + K_q^{-1} (\partial_x \phi_q)^2] + g_\phi \cos(2\sqrt{2}\phi_a) + g_\theta \cos(3\sqrt{2}\theta_a) + \cdots, \quad (8)$$

where we have assumed $\bar{\rho}_{+} = \bar{\rho}_{-} = \bar{\rho}$ (see below) and dropped terms with spatially oscillating factors $e^{i2n\pi\bar{\rho}x}$. The g_{ϕ} and g_{θ} terms for example originate from $\rho_{+}\rho_{-}$ 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 $S^{+}_{l,j}$ $\rightarrow e^{i\gamma}S^{+}_{l,j}$, the one-site translation along the chain $S^{\alpha}_{l,j} \rightarrow S^{\alpha}_{l,j+1}$, that along the rung $S^{\alpha}_{l,j} \rightarrow S^{\alpha}_{l+1,j}$, and the site-parity transformation along the chain $S^{\alpha}_{l,j} \rightarrow S^{\alpha}_{l,-j}$ are, respectively, expressed as

$$\theta_{\pm} \to \theta_{\pm} + \gamma,$$

$$(\phi_{\pm}(x), \theta_{\pm}(x)) \to (\phi_{\pm}(x + a_0) - \pi \overline{\rho}_{\pm} a_0, \theta_{\pm}(x + a_0) - \pi)$$

$$\theta_{\pm} \to \theta_{\pm} \pm 2\pi/3,$$

and

$$(\phi_{\pm}(x), \theta_{\pm}(x)) \to (-\phi_{\pm}(-x), \theta_{\pm}(-x)).$$

Furthermore, the rung-parity transformation $S_{1,j}^{\alpha} \leftrightarrow S_{3,j}^{\alpha}$ may be realized by $\bar{\rho}_{+} = \bar{\rho}_{-}$ 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 $\mathcal{H}'_{\text{eff}}$ evaluates the velocity v_a as $v_a \approx (-\tilde{f}_1 \bar{\rho}/m)^{1/2}$. Therefore, if $\tilde{f}_1 > 0$, then v_a 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 $\mathcal{H}'_{\text{eff}}$ and then define the following Ginzburg-Landau (GL) potential:

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

It is clear that as $f_1 > 0$, the potential is minimized by imposing $\rho_+ \neq \rho_-$. Moreover, it is found that

$$\rho_{+} - \rho_{-} \propto \tilde{b}_{2\pi/3,j}^{\dagger} \tilde{b}_{2\pi/3,j} - \tilde{b}_{-2\pi/3,j}^{\dagger} \tilde{b}_{-2\pi/3,j} \sim \sum_{l=1}^{3} \mathcal{V}_{l,j}^{z}.$$
 (10)

We thus conclude that for $\tilde{f}_1 > 0$, a finite long-range vector chiral order $\langle \mathcal{V}_{l,j}^z \rangle$ 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., $\lambda_0 \sim -O(J_{\perp})$], $\tilde{f}_1 < 0$ generally holds,^{12,17} while for J_{\perp} ~O(J) (i.e., $\lambda_0 \sim 0$), when *H* becomes closer to H_c^u , \tilde{f}_1 increases and tends to be positive. Consequently, the chiral phase is present in an intermediate-rung-coupling regime. Supposing that $\rho_+ > \rho_-$ holds in the chiral phase, we can speculate that the Ψ_- mode constructs a massive spectrum, whereas the Ψ_+ part provides a TLL state.¹⁰ 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. If $H \sim H_c^u$, the TLL parameter would be close to the universal value 1. The correlation function of the chirality might exhibit a power decay: $\langle \mathcal{V}_{l,j}^z \mathcal{V}_{l,0}^z \rangle \approx \langle \mathcal{V}_{l,j}^z \rangle^2 - \text{const}/j^2 + \cdots$ at $j \to \infty$.¹⁰

Let us now discuss the case of $f_1 < 0$, where $\bar{\rho}_+ = \bar{\rho}_-$ is restored and the bosonization is available. The ϕ_s sector in Eq. (8) yields a TLL, which is strongly stabilized by symmetries, while the low-energy physics of the ϕ_a sector depends on whether $\cos(2\sqrt{2}\phi_a)$ and $\cos(3\sqrt{2}\theta_a)$ are relevant or not: the scaling dimensions of these two are $2K_a$ and $9/(2K_a)$, respectively. The Hamiltonian $\mathcal{H}'_{\text{eff}}$ leads to $K_a \propto (-\bar{\rho}/\tilde{f}_1)^{1/2}$. Therefore, when $\overline{f_1} \sim 0$ and $\overline{\rho}$ is large enough, K_a is always much larger than 1. In this case, $\cos(3\sqrt{2}\theta_a)$ and $\cos(2\sqrt{2}\phi_a)$ are, respectively, highly relevant and irrelevant, and then the ϕ_a sector obtains a massive spectrum. If $g_{\theta} > 0$ (<0), the phase field θ_a is pinned on lines $\theta_a = \sqrt{2(2n)}$ $(\sqrt{2n\pi/3})$ in the $\theta_+ - \theta_-$ plane. Among these lines, only six lines intersect the physically relevant "Brillouin" zone, $-\pi < \theta_{+} \le \pi$ and $-\pi \le \theta_{-} < \pi$. This result implies that the ground states possess the sixfold degeneracy. To investigate the physical meaning of locking θ_a and the ground-state degeneracy, let us focus on the magnetization per site. The bosonization represents it as

$$\langle S_{l,j}^{z} \rangle \approx M - \frac{2}{3} \bar{\rho} a_0 \left\langle \cos\left(\sqrt{2}\,\theta_a + \frac{4}{3}\,\pi l\right) \right\rangle + \cdots$$
 (11)

One can see that the second term in Eq. (11) causes a downdown-up magnetization structure in the case of $g_{\theta} > 0$, while for $g_{\theta} < 0$ an up-up-down structure occurs: for instance, if θ_a is locked to zero for $g_{\theta} < 0$, $\langle S_{1,i}^z \rangle = \langle S_{2,i}^z \rangle = M + \delta$ and $\langle S_{3,i}^z \rangle$ $=M-2\delta[\delta \propto \langle \cos(\sqrt{2}\theta_a) \rangle]$. We thus conclude that an inhomogeneous magnetization for the rung is induced by pinning θ_a . Obviously, the parity and translational symmetries for the rung direction are spontaneously broken in this state. Three of the sixfold-degenerated states are indeed explained by this inhomogeneous distribution. The meaning of the remaining two-fold degeneracy is unknown.¹⁸ 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 $n \ge 2$ are also relevant.¹⁹ From the predictions of the chiral order for $\tilde{f}_1 > 0$ and the inhomogeneous phase under the condition $\tilde{f}_1 < 0$ and $|\tilde{f}_1| \sim 0$, the boundary $\tilde{f}_1=0$ is expected to be a first-order transition.

When $-\tilde{f}_1/\bar{\rho}$ increases so that $K_a < 9/4$, $\cos(3\sqrt{2}\theta_a)$ becomes irrelevant and the low-energy physics of the ϕ_a sector is described by a Gaussian model. This transition must be of a Beresinskii-Kosterlitz-Thouless (BKT) type.²⁰ After the

transition, the system is in a two-component TLL phase with all symmetries enjoying. If $-\tilde{f}_1/\bar{\rho}$ 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 gas²¹ 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 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.²²

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 $\mu > 0$, $\mu_0 > 0$, $H_c^l < H < H_c'$, 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. (9), let us introduce the GL potential for the present case as follows:

$$\mathcal{G} = g_0 \rho_0^2 + g_1 (\rho_+ + \rho_-)^2 + f_0 \rho_0 (\rho_+ + \rho_-) + f_1 \rho_+ \rho_- - \mu_0 \rho_0$$

- $\mu (\rho_+ + \rho_-).$ (12)

To find the stable magnon-density profile (ρ_0, ρ_+, ρ_-) , the Hessian matrix $H_{i,j} = \left[\frac{\partial^2 G}{\partial \rho_i \partial \rho_j}\right]$ is useful. At the local minimum point $(\rho_0, \overline{\rho}, \overline{\rho})$ satisfying $\partial \mathcal{G} / \partial \rho_j = 0$, the eigenvalues of $H_{i,j}$ 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 $C_2 < 0$. 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.^{23,24} At the boundary between these two regime, one might observe a weak singularity such as a magnetization cusp.

V. SUMMARY AND DISCUSSIONS

We have studied the three-leg frustrated spin tube (1) near the upper critical field. It has been predicted that the vector chiral order or the inhomogeneously magnetized order



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

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,^{7,8} 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 be common to all the cases with arbitrary *S*, as far as $S \leq O(1)$. 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 + ∞ 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 $\partial_x \phi_s$ and $\rho_+ + \rho_-$, but it does not directly interact ϕ_a and $\rho_+ - \rho_-$. A specific-heat measurement would be efficient in the detection.

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- ¹⁶When in Ref. 9 we discussed the possibility of the twocomponent TLL in frustrated integer-spin tubes near the lower critical field, this type instability induced by the density-density interaction was not taken into account.

- ¹⁷Note that in the extremely strong-rung-coupling area, the spinwave approach becomes less reliable because some coupling constants are much larger than the spin-wave bandwidth.
- ¹⁸The remaining degeneracy might be related with a nonlocal symmetry breaking.
- ¹⁹The modification of the magnetization would strongly depend on coupling constants of $\cos(3n\sqrt{2}\theta_a)$. However, we cannot quantitatively calculate them within the present approach.
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