Spontaneous chiral symmetry breaking on a fully polarized frustrated magnet at finite temperature

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Frustration can introduce more-than-two minima in a spin dispersion relation even in a fully polarized magnet under high magnetic field. We generally discuss, on the fully polarized phase, the possibility of the chiral symmetry breaking where the number of magnons pumped by finite temperature deviates to one side of minima. We study this phase by constructing the Ginzburg-Landau energy which is controlled by the external magnetic field and interactions between magnons near the dispersion minima. This chirality breaking phase accompanies, not a magnetization perpendicular to the external field, but the vector chirality $S_m \times S_a$. We also discuss the possibility of this anomaly in the magnetization curve is still controversial both theoretically [16] and experimentally [17]. We shall also discuss the possibility that magnons at finite temperature may have the SU(2) rotational symmetry in the spin space in the Hamiltonian, and the magnetization accompanies the symmetry breaking. In addition, various types of symmetry breaking phases in magnets have been found. One is the spin nematic phase, where not the magnetization but the long-range order breaks. It is well known that, the Heisenberg ferromagnet has the SU(2) rotational symmetry in the spin space in the Hamiltonian, and the magnetization accompanies the symmetry breaking. In this letter, we discuss a new type of symmetry breaking, namely, the breaking of chiral symmetry in a fully polarized frustrated magnet.

All spins align in any Heisenberg frustrated magnets by applying sufficiently high external field. In the fully polarized phase, the spin-flip excitation (magnon) can have a nontrivial dispersion relation due to frustration. For example, 1-dimensional (1D) $J_1$-$J_2$ spin chain under high magnetic field has two minima in the dispersion relation [3-6]. Besides, there are several systems realizing two dispersion minima, e.g., the saturated phase in the antiferromagnetic triangular lattice, and the $J_1$-$J_2$ models in the square lattice, the cubic lattice, the bcc lattice and so on. These two minima are related to each other by the chiral (inversion) symmetry of the lattice. We shall discuss the possibility that magnons at finite temperature, which has finite density, deviate to one side of minima: the possibility of the spontaneous chiral symmetry breaking. For the concrete discussion, we shall construct the effective Hamiltonian at low temperature and using the mean-field theory.

There are several compounds which may be viewed as 1D $J_1$-$J_2$ chains with interchain couplings [7]. LiCuVO$_4$ is one of appealing compounds which may realize this system [8-14]: near the saturation field, the magnetization curve shows a signature of a nontrivial phase [12]. The trial-wavefunction treatment suggests that the anomalous behavior near the saturation field is the evidence of a spin-nematic phase [15]. However, microscopic understanding of this anomaly in the magnetization curve is still controversial both theoretically [15] and experimentally [17]. We shall also discuss the possibility that this anomaly implies the chiral symmetry breaking phase.

Effective Hamiltonian on a saturated phase- Let us consider a spin-1/2 Heisenberg model with generic interactions in a magnetic field:

$$\mathcal{H} = \sum_{\langle i,j \rangle} J_{ij} S_i \cdot S_j + H \sum_j S_j^z. \quad (1)$$

For convenience, we adopt the hardcore boson representation of spin operator:

$$S_i^z = -1/2 + a_i^+ a_i, \quad S_i^+ = a_i^+, \quad S_i^- = a_i \quad (2)$$

We obtain the following boson Hamiltonian

$$H = \sum_{q} (\epsilon(q) - \mu) a_q^+ a_q + \frac{1}{2N} \sum_{q,k,k'} V(q) a_{k+q}^+ a_{k'}^+ -q a_k a_{k'}^+, \quad (3)$$

$$\epsilon(q) = \frac{1}{2} \sum_{j} J_{ij} \cos(\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)), \quad (4)$$

$$\omega(q) = \epsilon(q) - \epsilon_{\min}, \quad \mu = H_c - H, \quad H_c = \epsilon(0) - \epsilon_{\min}, \quad V(q) = 2(\epsilon(q) + U),$$

where $\epsilon_{\min}$ is the minimum of $\epsilon(q)$ and $U(\rightarrow \infty)$ is the hardcore potential respecting $S = 1/2$ at each site. The external field $H$ controls the energy of the magnon, which can be viewed as the chemical potential. For $H > H_c (\mu < 0)$, the magnon excitation is gapped, and the stable ferromagnetic phase is expected. If the magnetic field reduces below $H_c$, the gap of magnon closes and the single-magnon Bose-Einstein condensation (BEC) may occur, which leads to $\langle S_i^- \rangle = \langle a_i \rangle \neq 0$. In this letter, we concentrate on this gapped fully polarized phase for $\mu < 0$.

As commented before, the dispersion relation may have two minima due to frustration. For example, the Hamiltonian of the $J_1$-$J_2$ 1D chain is given by:

$$\mathcal{H} = \sum_i J_1 S_i \cdot S_{i+1} + J_2 S_i \cdot S_{i+2} + H \sum_i S_i^z, \quad (5)$$

where $J_1 < 0$, $J_2 > 0$. For $|J_1/4J_2| < 1$, the dispersion minima are at $q = \pm Q_1$, where $Q_1 = \arccos(-J_1/4J_2)$ as shown in Fig. [1]. As discussed later, the effect of interchain coupling

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may be easily included by extending this Hamiltonian. In the following discussion, for simplicity, we assume that the dispersion minima are located at \( q = \pm Q \) in a 3-dimensional magnet.

At sufficiently low temperature, the underlying physics may be understood by magnons near the dispersion minima. The important physical quantity is the interaction between magnons near the minima, which is given by the ladder diagram of Fig. 2:

\[
\Gamma(K; p, p') = V(p' - p) + V(-p' - p) - \frac{1}{2} \int \frac{d^d p''}{(2\pi)^d} \left( \omega(K/2 + p'' + p' - p) + \omega(K/2 - p' - p) - i0^+ \right),
\]

where \( K \) and \( p \) are respectively the center-of-mass momentum of the two magnons, and the relative momentum. The interaction \( \Gamma_1 \) between the magnons at the same minimum is given by \( \Gamma_1 = \Gamma(2Q; 0, 0)/2 \). The interaction \( \Gamma_2 \) between magnons at different minima is given by \( \Gamma_2 = \Gamma(0; Q, Q) \). \( \Gamma_1 \) and \( \Gamma_2 \) can be exactly solved \([4, 18, 19]\).

\[
\Gamma(\Delta, K; p, p') = \Gamma(K; p, p') + \Gamma(K; p, p')
\]

where the mass term is obtained by expanding \( \omega(k) \) near the minima up to the quadratic term (we assume \( m \) is the same for two minima). We also approximated that the interaction between magnons near minima are given by \( \Gamma_{1,2} \). We drop the higher order interaction term, which may not be important when the density of magnon is small. We concentrate on the case that \( \Gamma_1 > 0 \); otherwise, the higher order interaction terms (represented by dots) are needed to suppress the density of magnons.

Even in a fully polarized phase for \( \mu < 0 \), there are finite density of magnons at finite temperature. Naively we expect that, if the interaction \( \Gamma \) between \( u \) and \( v \) bosons is repulsive and sufficiently large relative to \( \Gamma_1 \), magnons favor decline to one side of minima in the energetic viewpoint, which leads to the chiral symmetry breaking.

Before going into the details, let us comment on the relevance between our scenario and the previous studies. The concept of the deviation of two-component bosons itself is not specific to our case. For example, in our setting, slightly below the saturation field for \( \mu > 0 \), magnon BEC may occur. If \( 0 < \Gamma_1 < \Gamma_2 \), there appears the difference of the densities of condensed magnons for \( k = \pm Q \), which accompanies the chirality breaking (cone phase) \([4]\). In addition, the frustrated 1D spin tube at zero temperature under high magnetic field can be described by many-component bosonic model, and the deviation of the densities can also lead to the chirality breaking \([20]\). The recent numerical simulation confirms that the spontaneous population imbalance of two-component bosons actually occurs in the 1D system at zero temperature \([21]\).

Next, we study the possibility of our scenario by using the mean-field approximation. In advance, we note that the chirality breaking may be the same university class as the Ising transition. Hence, even if it seems to appear in any dimensions within the mean-field treatment, it may occur only in 2D- or 3D magnetic systems at finite temperature.

**Mean field treatment** - At finite temperature, the density of magnons is finite: \( n_u = \langle 1/N \rangle \sum_k \langle u_k^\dagger u_k \rangle \neq 0 \), \( n_v = \langle 1/N \rangle \sum_k \langle v_k^\dagger v_k \rangle \neq 0 \). If we neglect the term \( ((1/N) \sum_k u_k^\dagger u_k - n_u)^2 \) and \( ((1/N) \sum_k v_k^\dagger v_k - n_v)^2 \), we obtain the mean-field Hamiltonian:

\[
H_{\text{mean}} = \sum_k (\epsilon_k^u u_k^\dagger u_k + \epsilon_k^v v_k^\dagger v_k) - \Gamma_1 N(n_u^2 + n_v^2) - \Gamma_2 N n_u n_v,
\]

where

\[
\epsilon_k^u = \sum_{i=x,y,z} \frac{k_i^2}{2m_i} - \mu + \Gamma_t n_t + \Gamma_s n_s,
\]

\[
\epsilon_k^v = \sum_{i=x,y,z} \frac{k_i^2}{2m_i} - \mu + \Gamma_t n_t - \Gamma_s n_s,
\]

\[
\Gamma_t = \Gamma_1 + \frac{\Gamma_2}{2}, \quad \Gamma_s = \Gamma_1 - \frac{\Gamma_2}{2},
\]

\( n_s \) can be viewed as the order parameter of the chirality breaking phase. Then, the free energy at finite temperature is given
bosons can become different: \( n_u \neq n_v (n_s \neq 0) \).

\[
\frac{F}{N} = \frac{1}{N} \sum_{k} \frac{1}{\beta} \left( \log(1 - e^{-\beta \epsilon_k}) + \log(1 - e^{-\beta \epsilon_{-k}}) \right) - \frac{\Gamma_t}{2} n_t^2 - \frac{\Gamma_s}{2} n_s^2.
\]

(10)

\[
\frac{\partial F}{\partial n_t} = 0 \quad \text{and} \quad \frac{\partial F}{\partial n_s} = 0 \quad \text{produces the self-consistent equations}
\]

\[
n_t = \frac{1}{N} \sum_{k} (g_b(\epsilon_k^u) + g_b(\epsilon_k^v)) \quad \text{and} \quad n_s = \frac{1}{N} \sum_{k} (g_b(\epsilon_k^u) - g_b(\epsilon_k^v)),
\]

(11a)

(11b)

where \( g_b(\epsilon) = 1/(e^{\beta \epsilon} - 1) \). Note that, if \( \Gamma_s > 0 \) and \( n_s > 0 \) \((n_s < 0)\), the right-hand side of (11b) becomes negative (positive): there is no solution of \( n_s \neq 0 \) for \( \Gamma_s > 0 \) and the chirality is never broken. Hence, in the following discussion we always assume \( \Gamma_s < 0 \). Later, we shall see the concrete model of \( n_s \neq 0 \). The schematic figure of this chirality breaking is shown in Fig. 3.

When this phase appears, the z-component of the vector chirality \( \langle S_3 \times S_3 \rangle \) has a finite value and can be viewed as the order parameter. On the other hand, the magnetization does not display nontrivial behaviors in this phase: \( \langle S_3^x \rangle = \langle S_3^y \rangle = 0 \). Hence, the order parameter of this phase is the same as the chiral phase at zero temperature found in the pure-1D \( J_1-J_2 \) chain [22].

Before seeing the concrete example, let us discuss the general aspect of the second-order-phase transition by using the Landau expansion.

**Landau free energy with respect to \( n_s \).** We assume that eq. (11a) is satisfied. Then, the Landau free energy for \( n_s \) is given by

\[
L = \frac{d^2 F}{dn_s^2} |_{n_s=0} n_s^2 + O(n_s^4).
\]

(12)

Hence, if \( \frac{d^2 F}{dn_s^2} |_{n_s=0} < 0 \), \( n_s \neq 0 \) and the chirality breaking phase is expected. At \( \frac{d^2 F}{dn_s^2} |_{n_s=0} = 0 \), the second order phase transition may occur. Explicitly,

\[
\frac{dF}{dn_s} = \frac{\partial F}{\partial n_t} \frac{dn_t}{dn_s} + \frac{\partial F}{\partial n_s} \frac{dn_s}{dn_s} = \left( \frac{1}{N} \sum_{k} (g_b(\epsilon_k^u) - g_b(\epsilon_k^v)) \right) \Gamma_s - \Gamma_s n_s,
\]

(13)

where we use \( \frac{\partial F}{\partial n_s} = 0 \). Thus

\[
\frac{d^2 F}{dn_s^2} |_{n_s=0} = \frac{2}{N} \sum_{k} g_b^2(\epsilon_k) \Gamma_s^2 - \Gamma_s
\]

(14)

\[
= \frac{1}{N} \sum_{k} -2 \beta \epsilon_k^2 e^{\beta \epsilon_k} (e^{\beta \epsilon_k} - 1)^2 - \Gamma_s,
\]

where we use \( \frac{dn_t}{dn_s} |_{n_s=0} = 0 \) and \( \epsilon_k = k^2/\beta - \mu + \Gamma_t n_t \).

For the low temperature limit \( \beta \to \infty \), we obtain

\[
\frac{d^2 F}{dn_s^2} |_{n_s=0} \approx - \Gamma_s > 0,
\]

(15)

which means a non-chirality breaking phase. This may be because the effect of interactions is irrelevant in the dilute limit. For the large temperature limit \( \beta \to 0 \),

\[
\frac{d^2 F}{dn_s^2} |_{n_s=0} \approx 2 \beta \Gamma_t n_t (\Gamma_s + 1) > 0,
\]

(16)

\[
n_t \approx \sqrt{\frac{2}{\beta \Gamma_t}},
\]

where we use \( \Gamma_t > |\Gamma_s| \) since we assumed that \( \Gamma_1 > 0 \) and \( \Gamma_s < 0 \) (\( 2 \Gamma_2 > \Gamma_1 > 0 \)). For large and small \( \beta \) limit, \( \frac{d^2 F}{dn_s^2} |_{n_s=0} > 0 \): the chirality breaking phase does not appear. Hence, it can appear only between an appropriate temperature region as illustrated in Fig. 4.

The other limit gives us the information of the qualitative behavior. For the large \( -\mu \) limit, we obtain

\[
\frac{d^2 F}{dn_s^2} |_{n_s=0} \approx - \mu \to - \infty \quad - \Gamma_s > 0.
\]

(17)
Hence, if the chirality breaking phase appears under any external field, this phase disappears by increasing the external field further enough.

Concrete calculation- Finally, let us concretely calculate eq. (11). We use the input parameters which have a relevance to LiCuVO4 as illustrated in Fig. 5. The mass parameter is directly derived as \( m_3 = 3.7, m_3^{(1)} = 6.7 \), and \( m_c^{(1)} = 0.068 \) (meV\(^{-1}\)), where we use \( J_1 = -1.6, J_2 = 3.8, J_3 = 0.4, J_4 = -0.015, J_5 = 0.08 \) (meV) following Ref. 9.

![FIG. 5: The lattice structure and the exchange interactions of LiCuVO4](Image)

FIG. 5: The lattice structure and the exchange interactions of LiCuVO4. Dots represent \( S = 1/2 \) spins.

By solving eq. (6), we obtain \( \Gamma_1 = -1.8, \Gamma_2 = 2.9 \) (meV). Naively, the attractive \( \Gamma_1 \) interaction and the repulsive \( \Gamma_2 \) interaction may favor the chirality breaking. However, we cannot treat the negative \( \Gamma_1 \) within the previously discussed mean-field theory due to the lack of the value of a sextic interaction term. Hence, in the following, we use only the mass term of LiCuVO4, and freely assume various \( \Gamma_{1,2} \) of positive \( \Gamma_1 \). In our-assumed-mass parameter, 2-dimensionality may hold below \( T \approx 5K \), and we focus on the case for \( T < 5K \). For simplicity, in the numerical calculation, the cutoff of \( k \) was taken nearly infinity since the integrand in eq. (11) becomes negligible for large \( k \). If we assume \( \Gamma_1 = 0.5, \Gamma_2 = 0.5 \), the chirality breaking phase does not appear for \( T < 5K \) unless \( \Gamma_2 > 30 \). If we substitute \( \Gamma_1 = 0.5 \) and \( \Gamma_2 = 50 \) by hand, the chirality breaking phase appears for \( T > 1.5K \) at \( \mu = 0 \). At \( T = 4K \), the magnetization curve is shown in Fig. 6, where \( H_c \) is assumed as that of LiCuVO4 given by eq. (4) [23]. The chirality breaking phase appears slightly above the saturation field. Fig. 6 qualitatively captures the properties of the anomalous behavior near the saturation field in LiCuVO4 [23].

Conclusion and discussion- We have suggested the chirality breaking phase in the fully saturated phase with two (or more) dispersion minima in frustrated magnets. If the interaction between magnons at different minima is repulsive and strong enough, the magnons pumped at finite temperature may deviate to one side of minima. We argued that this phase can exist, if some conditions are met, by using the effective Hamiltonian and the mean-field calculation. The order parameter of this phase is the \( z \)-component of the vector chirality \( \langle \mathbf{S}_l \times \mathbf{S}_m \rangle^z \), while the magnetization is usual: \( \langle \mathbf{S}_l^z \rangle = \text{const} \), \( \langle \mathbf{S}_l^x \rangle = \langle \mathbf{S}_l^y \rangle = 0 \). From the Landau expansion, we found that this phase is re-entrant and does not appear at the zero-temperature and high-temperature limit. This may be because, for the zero-temperature limit, the density of magnon is small and the effect of the interaction is negligible, and, for the high-temperature limit, the entropy dominates the physics. We also found that, for large \( -\mu \) limit, the chirality is not broken, since the density of magnons becomes small. These qualitative behaviors imply: (i) this phase can appear for an appropriate temperature region; (ii) this phase favors small \( |\mu| \) (slightly above the saturation field), and increment of \(-\mu\), which lowers the density of magnons, destabilizes this phase. We have also suggested that, the later qualitative behavior (ii) is consistent with the properties of the anomalous behavior of the magnetization curve found in LiCuVO4 [12]. Hence, if the experimentally observed anomalous behavior near saturation field actually implies the existence of a nontrivial phase [12], and if the phase is not a spin-nematic [16], it may be important to experimentally pursue the possibility of the chiral symmetry breaking slightly above the saturation field in LiCuVO4.

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[23] For simplicity, we do not consider the critical field of a bound-
magnon instability, which is known to exist in the parameter of
LiCuVO$_4$ [13].
[24] Even if we take into account the fact that $\Gamma_2$ is affected twice
as much as $\Gamma_1$ by change of the original scattering amplitude
eq. (6), $\Gamma_1 = 0.5$ and $\Gamma_2 = 50$ is not a little different from the
original values of LiCuVO$_4$ ($\Gamma_1 = -1.8$ and $\Gamma_2 = 2.9$), which
cannot be treated due to the theoretical difficulty up to now.
However, this does not exclude the possibility of the chirality
breaking phase in LiCuVO$_4$. Our approach is limited within the
mean-field treatment, which may not be reliable in quasi-1D
systems. In addition, we did not consider the effect of a bound
magnon [13], the density of which may be of the same order
as that of single magnons slightly above the saturation field.
Hence, our estimate may be considered as a phenomenological
and preliminary one.