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Spin-current order in anisotropic triangular antiferromagnets

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We analyze instabilities of the collinear up-up-down state of a two-dimensional quantum spin-S spatially anisotropic triangular lattice antiferromagnet in a magnetic field. We find, within large-S approximation, that near the end point of the plateau, the collinear state becomes unstable due to condensation of two-magnon bound pairs rather than single magnons. The two-magnon instability leads to a novel 2D vector chiral phase with alternating spin currents but no magnetic order in the direction transverse to the field. This phase breaks a discrete $Z_2$ symmetry but preserves a continuous $U(1)$ one of rotations about the field axis. It possesses orbital antiferromagnetism and displays a magnetoelectric effect.

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Introduction. The field of frustrated quantum magnetism has witnessed a remarkable revival of interest in recent years due to rapid progress in the fabrication and characterization of new materials and a multitude of theoretical ideas about competing orders and new quantum states of matter [1]. Studies of two-dimensional (2D) quantum triangular lattice antiferromagnets with spatially anisotropic exchange, such as Cs\(_2\)CuCl\(_4\) and Cs\(_2\)CuBr\(_4\), are of particular interest because of their surprisingly rich phase diagrams in a magnetic field [2, 3] which includes novel quantum states which have no classical analogs and display a wealth of properties which are highly sought after for applications. The large number of different phases involved, which reaches 9 in the case of Cs\(_2\)CuBr\(_4\)[3], reveals a highly complex interplay between quantum fluctuations and anisotropy of the interactions.

One of the best understood phases of a frustrated spin system in a magnetic field is a collinear state with a fixed, field-independent magnetization equal to exactly 1/3 of the saturation value. In this state, known as the up-up-down (UUD), two spins in each triangle point up and one points down. This quantum state preserves continuous $U(1)$ symmetry of rotations about the field direction and has finite gaps in all spin excitations [4]. The UUD state is similar to plateau states in quantum Hall effect, although, unlike them, it spontaneously breaks lattice translational symmetry. An extension of the UUD state with unbroken translational symmetry has been proposed theoretically[5, 6] but not yet found experimentally.

In a classical isotropic 2D Heisenberg systems with nearest exchange $J$, the UUD phase is the ground state for just one value of the external field $h = 3J$ (1/3 of the saturation field $h_{sat} = 9J$). At all other fields spins order in a non-collinear fashion. In an anisotropic lattice with exchanges $J$ and $J'$ (see Fig. 1), a non-collinear order wins for all fields, so that classically UUD phase is never a ground state. For quantum systems, the situation is different as quantum fluctuations favor a collinear spin structure and compete with classical fluctuations [4, 7, 8]. In the isotropic case, quantum fluctuations stabilize the UUD phase with gapped spin-wave excitations in a finite interval of $h$ with the width of order $1/S$. In an anisotropic case, the width of the UUD phase is determined by the competition between $1/S$, which measures the strength of quantum fluctuations, and the degree of antisotropy of exchange interactions $(1 - J'/J)$ (Ref. [8]). The dimensionless parameter, which determines the UUD width relative to its value in the isotropic case, is $\delta = (40/3)S(1 - J'/J)^2$ (we use the same numerical factor as in [8]). The UUD phase persists up a finite anisotropy $\delta_{cr} = 4$, see Fig. 1. The boundaries of the UUD phase have been determined from the local stability analysis [8] as the values of $h$ at which spin-wave dispersion softens. Of the two low-energy spin-wave branches, one softens at the lower boundary of the UUD phase and another at the upper boundary. Near the critical $J'/J$, both spin-wave instabilities occur at finite momenta, and each leads to a chiral, non-coplanar state (often called a distorted umbrella), in which $\langle S_{r}\rangle$ has finite components along both directions perpendicular to the field [8, 9] (see Fig. 1).

The analysis of the same model for $S = 1/2$, however, found very different states surrounding the UUD plateau near...
We consider a system of localized spins to analyze a possibility of a bound state. Quantum fluctuations win over classical fluctuations and stabilize the system near the UUD phase, and we rescale the “gap” equation for the two-magnon order parameter and show that it is purely imaginary. Such order parameter breaks a discrete $Z_2$ symmetry and gives rise to a bond-nematic state with non-zero vector and scalar chiralities within a single triangle of spins: $\langle S_A \cdot S_B \times S_C \rangle \neq 0$ and $\langle S_A \times S_B \times S_C \rangle \neq 0$ (vector and scalar chiralities are proportional to each other since the total magnetization $M = \langle S_z \rangle$ is finite). Such a state supports circulating spin currents (Fig. 2) and we label it a spin-current state (SC). We present the modified large-$S$ phase diagram of the model in Fig. 1.

Experimental signatures of a SC state are rather peculiar. First, it exhibits a magneto-electric effect because both spin current and electric field are odd under spatial reflections and couple linearly [12]. As a result, spin-wave excitations of the SC state depend linearly on $E$. Second, orbiting spin currents generate charge currents, which in turn produce staggered magnetic moments, which can be measured by NMR and μSR [13].

The model. We consider a system of localized spins on an anisotropic triangular lattice with Heisenberg nearest-neighbor interactions $J$ and $J'$, subject to an external field $h = 2\mu_B H_z$:

$$\mathcal{H} = \sum_\mathbf{r} \left( J S_\mathbf{r} S_{\mathbf{r}+\mathbf{a}_1} + J' \sum_{j=\mathbf{1,2}} S_\mathbf{r} S_{\mathbf{r}+\mathbf{a}_j} - \tilde{h} S_\mathbf{r}^z \right),$$

where $\mathbf{a}_{1,2} = a(1/2, \pm \sqrt{3}/2)$ connects spins on neighboring chains, and $a$ is the lattice constant. For convenience, we rescale $h = hS$ and use $h$ for the field. The saturation field, above which the magnetization $M$ reaches maximum possible value $M_{\text{sat}} = S$, is given by $h_{\text{sat}} = (2J + J')/2J$. We are interested in the behavior of the system near $h_{\text{sat}}/3$, where quantum fluctuations win over classical fluctuations and stabilize UUD phase in a finite range of fields. In the isotropic case, $J' = J$, the UUD phase exists in a field range between $h_{c1} = (h_{\text{sat}}/3)(1-0.5/2S)$ and $h_{c2} = (h_{\text{sat}}/3)(1+1.3/2S)$. In the anisotropic case, $J' < J$, the width of the UUD phase decreases and eventually vanishes at $\delta_{\text{cr}} = 4$, which defines $J'_{\text{cr}} = J(1-\sqrt{3}/10S)$.

The excitation spectrum of the UUD phase at $\delta \leq 4$ can be straightforwardly obtained by using a three-sublattice representation for two spin-up and one spin-down sublattices and introducing [8, 11] three sets of Holstein-Primakoff bosons, $a, b$, and $c$. One of the three spin-wave branches describes the precession of the total magnetization, has energy of the order $h_{\text{sat}}/3$, and is irrelevant to our analysis. The other two branches, denoted $d_{1,2,k}$ below, describe low-energy excitations. Explicitly,

$$\mathcal{H}_{\text{und}}^{(2)} = S \sum_{\mathbf{k}} \left( \omega_1 d_{1,\mathbf{k}}^\dagger d_{1,\mathbf{k}} + \omega_2 d_{2,\mathbf{k}}^\dagger d_{2,\mathbf{k}} \right),$$

where at small $k$

$$\omega_{1,2}(\mathbf{k}) = \pm \left( h - h_0 - \frac{1}{5S} J - \frac{3}{4} J' k^2 \right) + \frac{3J}{20S} Z_\mathbf{k},$$

and $h_0 = J + 2J'$. The excitation $d_{1,\mathbf{k}}$ softens at the lower boundary of the UUD phase, at $h = h_{c1}(\delta) = h_{\text{end}} - 9J/(40S) \sqrt{(4-\delta)/3}$, where $h_{\text{end}} = h_0(1 + 20/(120S))$. The softening happens at a finite momenta $\pm k_1 = (\pm k_1, 0)$, where $k_1 \approx (3/(10S))^{1/2}(1 + \sqrt{(4-\delta)/12})$. The excitation $d_{2,\mathbf{k}}$ softens at the upper boundary $h = h_{c2}(\delta) = h_{\text{end}} + 27J/(40S) \sqrt{(4-\delta)/3}$, at momenta $\pm k_2 = (\pm k_2, 0)$, where $k_2 = (3/(10S))^{1/2}(1 - \sqrt{(4-\delta)/12})$. The spin-wave softening at either $h_{c1}(\delta)$ or $h_{c2}(\delta)$ signals condensation of one-magnon excitations. A Ginzburg-Landau-type analysis shows [8] that condensation spontaneously breaks $Z_2$ symmetry between degenerate minima at $\pm k_1$ and $\pm k_2$. As a result, one-magnon condensation gives rise to an incommensurate spiral order with spontaneously broken $O(2) \times Z_2$ symmetry and a finite non-coplanar-long range order $\langle S_\mathbf{r}^z \rangle \neq 0$.

At the end-point of the plateau $\delta = 4$, $h_{c1} = h_{c2} = h_{\text{end}}$, both spin-wave branches touch zero simultaneously at $\pm k_0 = (\pm k_0, 0)$, where $k_0 = \sqrt{3}/10S$. The presence of four soft modes leads to a variety of possible non-coplanar chiral orders with non-zero $\langle S_\mathbf{r}^z \rangle$. However, we show below that instead the system undergoes a pre-emptive pairing instability into a state with no transverse order, $\langle S_\mathbf{r}^z \rangle = 0$, but nonetheless with a finite chirality $\langle 2(\mathbf{S}_\mathbf{r} \times \mathbf{S}_\mathbf{r}') \rangle \neq 0$.

Magnon pairing. To analyze a possibility of a bound state of two magnons, we need to include magnon-magnon interaction. The derivation of the interaction Hamiltonian is lengthy but straightforward: one has to express two-magnon interaction Hamiltonian $\mathcal{H}_{\text{und}}^{(4)}$ originally written in terms of $a_\mathbf{k}, b_\mathbf{k}$, and $c_\mathbf{k}$ bosons, in terms of the low-energy eigen-modes $d_{1,\mathbf{k}}$ and $d_{2,\mathbf{k}}$ from Eq. (2). The full transformation is given in [11]. Near momenta $\pm k_0$, which are mostly relevant to the
pairing problem, this transformation simplifies to

\[ a_k = \frac{f(k)}{\sqrt{2}}(e^{i|k|d_{1,k} - e^{-i|k|d_{1,-k}}}, \]

\[ b_k = -\frac{f(k)}{\sqrt{2}}(e^{-i|k|d_{1,k} + e^{i|k|d_{1,-k}}}, \]

\[ c_k = f(k)(d_{2,k} - e^{2i|k|d_{1,k}}, \]

where \( f(k) = \sqrt{\text{Im}[(k_x \pm k_0)^2 + k_y^2 + (1 - \delta/4)k_0^2]}^{-1/4} \) and \( s_k = \pi \text{ sign}(k_x/k_y)/4. \)

Consider first \( \delta < 4 \), when only one boson becomes soft at either \( h_{c1} \) or \( h_{c2} \), while other remains massive and can be neglected. For concreteness, consider the vicinity of \( h_{c1} \), where \( d_1 \) excitation softens. The magnon-magnon pairing interaction involving only \( d_1 \) bosons is

\[ \mathcal{H}_{d1}^{(4)} = \frac{8(J + 2J')}{(4 - \delta)} N^3 \sum_{p,q} d_{1,k+p}^d d_{1,-k-q}^d d_{1,k+q} d_{1,-k-q} \]

This interaction is obviously strongly repulsive and does not give rise to a bound state. The same holds for \( d_2 \) mode near \( h_{c2} \). As a result, one-magnon condensations at \( d_1 \) and \( d_2 \) bosons, both of which are gapless at \( \pm k_0 \). The \( d_1 - d_2 \) interaction with zero total momentum has two relevant terms: one describes "normal" \( 2 \to 2 \) process with simultaneous creation and annihilation of \( d_1 \) and \( d_2 \) bosons, the other describes "anomalous" \( 4 \to 0 \) and \( 0 \to 4 \) processes with simultaneous creation or annihilation of \( d_1 \) and \( d_2 \) bosons. We find that the strongest pairing interaction involves momentum transfer \( \pm 2k_0 \) for each of the bosons involved.

The corresponding interaction reads

\[ \mathcal{H}_{d1d2} = \frac{3}{N} \sum_{p,q} \Phi(p, q) \left( d_{1,k_0+p}^d d_{2,k_0-p}^d d_{1,-k_0+q} d_{2,-k_0-q} - d_{1,k_0+p}^d d_{2,-k_0-p}^d d_{1,-k_0+q} d_{2,-k_0-q} \right) + \text{h.c.} \]

where \( p \) and \( q \) are much smaller than \( k_0 \), and the vertex

\[ \Phi(p, q) = -(J + 2J') f^2(p) f^2(q) \rightarrow -(J + 2J') \frac{k_0^2}{|p||q|} \]

where \( f(p) \) was introduced after Eq. (5), and the limit stands for \( \delta \to 4 \). The pairing interaction with small momentum transfer, \( \Phi(p, q) d_{1,k_0+p}^d d_{2,-k_0-p}^d d_{1,k_0+q} d_{2,-k_0-q} \), has a much smaller \( \Phi(p, q) \) which remains finite in the limit \( p, q \to 0 \). Such interaction is then irrelevant for our analysis.

Now observe that the sign of \( 2 \to 2 \) term is negative, while the one of \( 4 \to 0 \) term is positive. The negative sign of the \( 2 \to 2 \) term implies that the "normal" interaction between \( d_1 \) and \( d_2 \) bosons is attractive and favors a pairing with

\[ F_{k_0}(p) = \langle d_{1,k_0+p} d_{2,-k_0-p} \rangle = \tilde{\Upsilon}/|p| = F_{-k_0}(p). \]
Although our analysis of two-particle instabilities has described a novel two-magnon interaction. Indeed, the interaction Hamiltonian (7) can be written as

\[ H_{\text{DM}}^{(2)} = -\frac{9J}{N} \mathcal{H}_{k_0}^{\text{DM}} \mathcal{H}_{-k_0}^{\text{DM}}, \]

where [11]

\[ \mathcal{H}_{\pm k_0}^{\text{DM}} = \frac{1}{6S} \sum_r \hat{\epsilon} \cdot S_r \times (S_{r+a_1} + S_{r+a_2}) \]

\[ = i \sum_{k \in \pm k_0} f_k^2 \left( d_{1,k}^d d_{2,-k} - d_{1,k}^d d_{2,-k}^d \right). \] (12)

As a result, the development of a non-zero \( \gamma \) can be viewed as the appearance of Dzyaloshinskii-Moriya (DM) interaction. Indeed, the interaction Hamiltonian (7) can be written as

\[ H_{k_0}^{\text{DM}} = \frac{1}{6S} \sum_r \hat{\epsilon} \cdot S_r \times (S_{r+a_1} + S_{r+a_2}) \]

\[ = i \sum_{k \in \pm k_0} f_k^2 \left( d_{1,k}^d d_{2,-k} - d_{1,k}^d d_{2,-k}^d \right). \]

This observation helps to understand magneto-electric effect in the SC state: because \( \mathcal{H}_{\pm k_0}^{\text{DM}} \) couples linearly to an electric field \( E \), i.e.,

\[ D = D_0 + D_1 E + \ldots \] as a result, spin-wave excitations of the SC phase depend linearly on \( E \).

SC order has been previously explored in 1D spin ladders [14–16] and was suggested for a frustrated Heisenberg model in 2D [17, 18]. There, however, a SC state is a spiral state, in which a continuous \( U(1) \) symmetry is restored by strong quantum fluctuations [18]. In our case spiral states are present in the phase diagram away from the end-point of the UUD phase, while the SC state emerges as a result of a pre-emptive two-magnon instability rather than due to divergent one-magnon fluctuations. Our two-magnon instability (which necessary leads to an imaginary order parameter) is also fundamentally different from two-magnon instabilities with real order parameter which lead to a spin-nematic order, either on a site or on a bond [19–24]. Such order generally occurs in systems with ferromagnetic exchanges at least on some of the bonds, when there is an attractive interaction between magnons. Here, all exchange couplings are antiferromagnetic, and magnon-magnon interaction is repulsive. Our pairing of magnons from different branches is conceptually similar to the inter-pocket pairing in multi-band fermionic systems, such as Fe-based superconductors with only electron pockets [25].

The phase diagram near the end point of UUD state has been recently analyzed in [9] in a self-consistent semiclassical formalism. This method, however, does not allow for the analysis of two-particle instabilities.

**Comparison with SDW state.** Although our analysis uses \( 1/S \) expansion, it is nevertheless instructive to compare symmetry properties of our spin-current state with that of a collinear SDW state observed for \( S = 1/2 \) near the end point of the UUD phase. Like we said, spin-current state is much closer to SDW state than a spiral state (the result of one-magnon condensation) because both spin-current and SDW states preserve \( U(1) \) symmetry of rotations about the field direction. But the two states do differ as SDW state has no chiral order [10]. It may be that \( S = 1/2 \) is simply special and non-chiral SDW state is only present at \( S = 1/2 \). But it also may be that the two-magnon instability, which we found, is only a ‘tip of the iceberg’, and the two-magnon condensation triggers the development of multi-magnon condensates at some \( \delta > \delta_{\text{cr}} \), which in turn changes the properties of the spin-current state. This last possibility is inspired by the observation that SDW state is incommensurate and that the UUD-SDW transition for \( S = 1/2 \) is a commensurate-incommensurate transition [10]. Such transition occurs via a proliferation of solitons – strings of displaced spins which are shifted from their equilibrium UUD pattern. Since changing the direction of a single spin \( S \) requires \( 2S \) magnons, a proliferation of solitons implies condensation of \( 2S \) magnons per every displaced spin. Then, in magnon description, a commensurate-incommensurate transition involves a condensation of an infinite number of magnons. One can imagine, by analogy with coupled superconducting and spin density orders [26], that proliferation of SC domain walls, depicted in Fig. 2, may cause the appearance of an incommensurate modulation of \( \rho(x) \) due to “density-density” type coupling between the magnon density and the density of domain walls. Whether or not this is the case requires going beyond the instability condition (11) and analyzing excitation spectrum and inter-pair interactions within the spin-current phase [27].

**Conclusions.** We have described a novel two-magnon pairing instability of the up-up-down phase of the spatially anisotropic triangular lattice antiferromagnet in a magnetic field. The magnon pairing is of “inter-band” type in that the condensate is made out of bosons from the two different spin-wave branches. This instability pre-empts a single-magnon condensation for arbitrary spin \( S \) and gives rise to a highly unconventional 2D order in which transverse spin components are disordered, yet the ground state has a non-zero vector chirality on every lattice bond and circulating spin currents in every elementary triangle. This state breaks \( Z_2 \) chiral symmetry but preserves \( U(1) \) symmetry of rotations about the field direction. The development of such a phase can be thought of as a spontaneous generation of the Dzyaloshinskii-Moriya interaction. This new state exhibits a magneto-electric effect, which gives rise to a non-trivial linear dependence of spin-wave excitations on the applied electric field \( E \), and also has staggered magnetic moments, which can be measured by NMR and \( \mu \)SR.

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