Phases of a Triangular-Lattice Antiferromagnet Near Saturation

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Phys. Rev. Lett. **113**, 087204 — Published 21 August 2014

DOI: 10.1103/PhysRevLett.113.087204

Phases of triangular lattice antiferromagnet near saturation

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We consider 2D Heisenberg antiferromagnets on a triangular lattice with spatially anisotropic interactions in a high magnetic field close to the saturation. We show that this system possess rich phase diagram in field/anisotropy plane due to competition between classical and quantum orders: an incommensurate non-coplanar spiral state, which is favored classically, and a commensurate co-planar state, which is stabilized by quantum fluctuations. We show that the transformation between these two states is highly non-trivial and involves two intermediate phases – the phase with co-planar incommensurate spin order and the one with non-coplanar double-$\mathbf{Q}$ spiral order. The transition between the two co-planar states is of commensurate-incommensurate type, not accompanied by softening of spin-wave excitations. We show that a different sequence of transitions holds in triangular antiferromagnets with exchange anisotropy, such as $Ba_3CoSb_2O_9$.

PACS numbers: 75.10.Jm

Introduction. The field of frustrated quantum magnetism witnessed a remarkable revival of interest in the last few years due to rapid progress in synthesis of new materials and in understanding previously unknown states of matter. The two main lines of research in the field are searches for spin-liquid phases and for new ordered phases with highly non-trivial spin structures [1]. For the latter, the most promising system is a 2D Heisenberg antiferromagnet on a triangular lattice in a finite magnetic field, as this system is known to possess an "accidental" classical degeneracy: every classical spin configuration with a triad of neighboring spins satisfying $\mathbf{S}_\mathbf{r} + \mathbf{S}_{\mathbf{r}+\boldsymbol{\delta}_1} + \mathbf{S}_{\mathbf{r}+\boldsymbol{\delta}_2} = \mathbf{h}/(3J)$, where J is the exchange interaction, belongs to the ground state manifold.

An infinite degeneracy, however, holds only for an ideal Heisenberg system with isotropic nearest-neighbor interaction. Real systems have either spatial anisotropy of exchange interactions, as in Cs_2CuCl_4 [2, 3] and Cs_2CuBr_4 [4–6] for which the interaction J on horizontal bonds is larger than J' on diagonal bonds (see insert in Fig. 1), or exchange anisotropy in spin space, as in $Ba_3CoSb_2O_9$, for which $J_z < J_\perp = J$ (an easy plane anisotropy) [7–10]. An anisotropy of either type breaks accidental degeneracy already at a classical level and for fields $\mathbf{h} = h\hat{z}$ slightly below the saturation field $h_{\rm sat}$ selects a non-coplanar cone state with

$$\langle \mathbf{S}_\mathbf{r}\rangle = (S-\rho)\hat{z} + \sqrt{2S\rho}(\cos[\mathbf{Q}\cdot\mathbf{r}+\varphi]\hat{x} + \sin[\mathbf{Q}\cdot\mathbf{r}+\varphi]\hat{y}), \tag{1}$$

where $\rho \sim S(h_{\rm sat} - h)/h_{\rm sat}$ is the density of magnons (the *condensate fraction*) which determines the magnetization $M = S - \rho$. Here $\varphi \in (0, 2\pi)$ is the phase of the $U(1)$ condensate and $\mathbf{Q} = (Q, 0)$ is the ordering wave vector. It is incommensurate with $Q = Q_{\rm i} = 2\cos^{-1}(-J'/2J)$ in the spatially anisotropic case $J' \neq J$ and commensurate with $Q = Q_0 = 4\pi/3$ for the easy-plane anisotropy (in the last case, the values of $\mathbf{Q}_0\cdot\mathbf{r} = 2\pi\nu/3 \pmod{2\pi}$, with $\nu = \pm1, 0$). The choice of $+\mathbf{Q}$ or $-\mathbf{Q}$ in (1) selects chirality of the cone state and specifies broken Z_2 symmetry.

Quantum fluctuations are also known to lift accidental degeneracy, and do so already in the *isotropic* system. However, they select different ordered state, which is the co-planar,

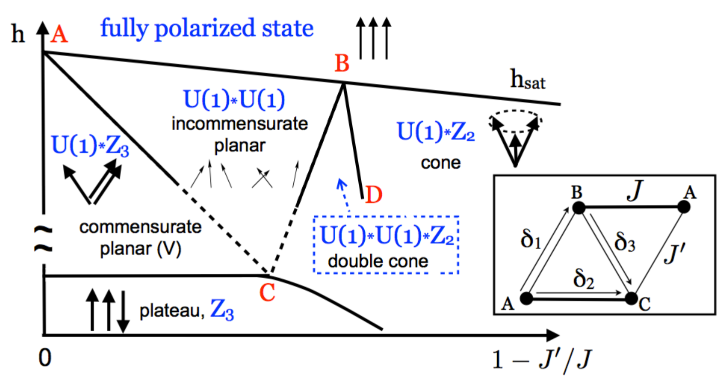

FIG. 1. Phase diagram of the spatially anisotropic triangular lattice antiferromagnet with large S near saturation field, as a function of spatial anisotropy of the interactions. The phases at small and large anisotropy are commensurate co-planar V-phase, whose order parameter manifold is $U(1) \times Z_3$, and incommensurate non-coplanar chiral cone phase, which lives in $U(1) \times Z_2$ manifold. In between, there are two incommensurate phases: a co-planar phase, with $U(1) \times U(1)$ symmetry, and a non-coplanar double cone phase, which is characterized by $U(1) \times U(1) \times Z_2$ manifold. Line AC denotes the CI transition from the V phase to the incommensurate planar phase. The insert shows the geometry of the lattice: exchange is J on horizontal bonds (bold) and J' on diagonal bonds (thin).

commensurate state with two parallel spins in every triad, often called the V state (Fig. 1) [11–13].

This order is described by

$$\langle \mathbf{S}_\mathbf{r}\rangle = (S - 2\rho\cos^2[\mathbf{Q}\cdot\mathbf{r}+\theta])\hat{z} + \sqrt{4S\rho}\cos[\mathbf{Q}\cdot\mathbf{r}+\theta] \times(\cos\varphi\hat{x} + \sin\varphi\hat{y}), \tag{2}$$

where $\mathbf{Q} = \mathbf{Q}_0$, $\rho = \rho_{\mathbf{Q}_0} + \rho_{-\mathbf{Q}_0}$ is the sum of two equal contributions from condensates with wave vectors $\pm\mathbf{Q}_0 = (\pm Q_0, 0)$, φ is a common phase of the two condensates (broken $U(1)$), and θ is their *relative* phase. The values of θ in the commensurate V phase are constrained to $\theta = \pi\ell/3$, where $\ell = 0, 1, 2$ describe three distinct degenerate spin configurations (broken Z_3 symmetry).

The issue we consider in this paper is how the system evolves at $h \leq h_{\rm sat}$ from the co-planar V state, selected by quantum fluctuations, to the non-coplanar cone state, selected by classical fluctuations, as the anisotropy increases. We show that this evolution involves commensurate-incommensurate transition (CIT) and, in the case of $J - J'$ model, an intermediate double cone phase.

The phase diagrams. To begin, it is instructive to compare order parameter manifolds in the two phases. The order parameter manifold in the V phase is $U(1) \times Z_3$ and that in the cone phase is $U(1) \times Z_2$. The symmetry breaking patterns in the two phases are not compatible, hence one should expect either first-order transition(s) or an intermediate phase(s). We show that in $J - J'$ model the evolution occurs via two intermediate phases, see Fig. 1. As $\delta J = J - J'$ increases, the V phase first undergoes a CIT at $\delta J_{c1} \sim (J/\sqrt{S})(h_{\rm sat} - h)/h_{\rm sat}$ (line AC in Fig. 1). The new phase remains co-planar, like in (2), but the phase θ becomes incommensurate and coordinate-dependent, extending broken Z_3 to $U(1)$. The incommensurate co-planar $U(1) \times U(1)$ state exists up to a second critical $\delta J_{c2} \sim J/\sqrt{S}$, where the system breaks the Z_2 symmetry between the condensates at $\pm Q$ (line BC in Fig. 1). At larger δJ the two condensates still develop, but one of them shifts to a new wave vector $\bar{\mathbf{Q}}$ and its magnitude gets smaller. The resulting state is a non-coplanar double cone state with order parameter manifold $U(1) \times U(1) \times Z_2$. Finally, at the third critical anisotropy $\delta J_{c3} = \delta J_{c2}[1 + O((h_{\rm sat} - h)/h_{\rm sat})]$ the magnitude of the condensate at $\bar{\mathbf{Q}}$ vanishes and the double cone transforms into a single cone (line BD in Fig. 1).

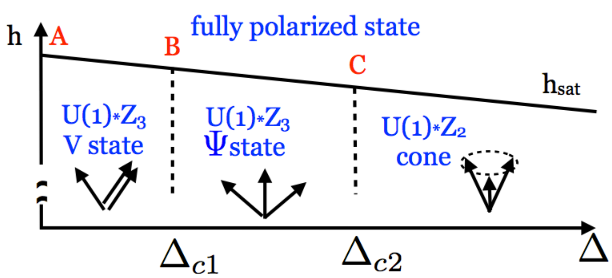

FIG. 2. The phase diagram of the XXZ model in a magnetic field near a saturation value, $\Delta = (J - J_z)/J$. The cone and V states are the same as in Fig. 1, but the transformation from one phase to the other with increasing spin exchange anisotropy proceeds differently from the case of spatial exchange anisotropy and involves one intermediate co-planar commensurate phase with Ψ-like spin pattern.

In systems with easy-plane anisotropy $\Delta = (J - J_z)/J > 0$, the the ordering wave vector remains commensurate, $Q = Q_0 = \pm 4\pi/3$, for all $\Delta > 0$, and the evolution from quantum-preferred V state to classically-preferred cone state proceeds differently, via two first-order phase transitions (see Fig. 2). The V state with $\theta = \ell\pi/3$ survives up to some critical $\Delta_{c1} \sim 1/S$, where another commensurate co-planar order develops, for which $\theta = (2\ell + 1)\pi/6$. The corresponding spin pattern resembles Greek letter Ψ and we label this state a Ψ phase. The Ψ phase survives up to $\Delta_{c2} \geq \Delta_{c1}$, beyond which the spin configuration turns into the commensurate cone state.

We now discuss the model and the calculations which lead to phase diagrams in Figs. 1 and 2.

The model. The isotropic Heisenberg antiferromagnet on a triangular lattice is described by the Hamiltonian

$$\mathcal{H}_0 = \frac{1}{2} J \sum_{\mathbf{r},\boldsymbol{\delta}} \mathbf{S}_{\mathbf{r}} \cdot \mathbf{S}_{\mathbf{r}+\boldsymbol{\delta}} - \sum_{\mathbf{r}} h S^z_{\mathbf{r}}, \tag{3}$$

where $\boldsymbol{\delta}$ are nearest-neighbor vectors of the triangular lattice. The two perturbations we consider are

$$\delta\mathcal{H}_{\rm anis} = (J' - J) \sum_{\mathbf{r}} \mathbf{S}_{\mathbf{r}} \cdot (\mathbf{S}_{\mathbf{r}+\boldsymbol{\delta}_1} + \mathbf{S}_{\mathbf{r}+\boldsymbol{\delta}_3}), \tag{4}$$

$$\delta\mathcal{H}_{\rm xxz} = \frac{1}{2}(J_z - J) \sum_{\mathbf{r},\pm\boldsymbol{\delta}_{1,2,3}} S^z_{\mathbf{r}} S^z_{\mathbf{r}+\boldsymbol{\delta}}. \tag{5}$$

where $\langle \mathbf{r}, \mathbf{r} + \boldsymbol{\delta}_{1,3} \rangle$ are diagonal bonds.

We consider a quasi-classical limit $S \gg 1$, when quantum fluctuations are small in $1/S$ and quantum and classical tendencies compete at small anisotropy $\delta J/J \sim 1/\sqrt{S}$ and/or $\Delta/J \sim 1/S$. In this limit, the calculations in the vicinity of the saturation field can be done using a well-established dilute Bose gas expansion and are controlled by simultaneous smallness of $1/S$ and of $(h_{\rm sat} - h)/h_{\rm sat}$ [12, 14–16]. We argue that our results are applicable for all values of S, down to $S = 1/2$, because (i) quantum selection of the V state holds even for $S = 1/2$ [15], and (ii) numerical analysis of $S = 1/2$ systems [15, 18] identified the same phases near saturation field as found here.

We set quantization axis along the field direction and express spin operators $\mathbf{S}_{\mathbf{r}}$ in terms of Holstein-Primakoff bosons a, a^+ as $S^-_{\mathbf{r}} = [2S - a^+_{\mathbf{r}} a_{\mathbf{r}}]^{1/2} a^+_{\mathbf{r}}$, $S^z_{\mathbf{r}} = S - a^+_{\mathbf{r}} a_{\mathbf{r}}$. Substituting this transformation into $\mathcal{H}_{\rm anis/xxz}$ and expanding the square root one obtains the spin-wave Hamiltonian $\mathcal{H} = \mathcal{E}_{\rm cl} + \sum_{j=2}^{\infty} \mathcal{H}^{(j)}$, where $\mathcal{E}_{\rm cl}$ stands for the classical ground state energy, and $\mathcal{H}^{(j)}$ are of j-th order in operators a, a^+. For our purposes, terms up to $j = 6$ have to be retained in the expansion (see the Supplement [19] for technical details). The quadratic part of the spin-wave Hamiltonian reads

$$\mathcal{H}^{(2)} = \sum_{\mathbf{k}} (\omega_{\mathbf{k}} - \mu) a^+_{\mathbf{k}} a_{\mathbf{k}} \tag{6}$$

where $\omega_k = S(J_{\mathbf{k}} - J_{\mathbf{Q}})$ is the spin-wave dispersion, measured relative to its minimum at the saturation field $h_{\rm sat}$, and $\mu = (h_{\rm sat} - h)/h_{\rm sat}$ plays the role of chemical potential. For $J - J'$ model, $J_{\mathbf{k}} = \sum_{\pm\delta_j} J_{\delta_j}(e^{i\mathbf{k}\cdot\delta_j} - 1)$, where $J_{\delta_{1,3}} = J'$ and $J_{\delta_2} = J$. Here $\mathbf{Q} = \mathbf{Q}_{\rm i} = (Q_{\rm i}, 0)$ with $Q_{\rm i} = 2\cos^{-1}(-J'/2J)$. For XXZ model, $J_{\mathbf{k}} = \sum_{\pm\delta_j}(Je^{i\mathbf{k}\cdot\delta_j} - J_z)$ and $\mathbf{Q} = \mathbf{Q}_0 = (4\pi/3, 0)$. In both cases, lowering of a magnetic field below $h_{\rm sat}$ makes $(\omega_{\mathbf{k}} - \mu)$ negative at $\mathbf{k} \approx \pm\mathbf{Q}$, and drives the Bose-Einstein condensation (BEC) of magnons. To account for BEC, we introduce two condensates, $\langle a_{\mathbf{Q}} \rangle = \sqrt{N}\psi_1$ and $\langle a_{-\mathbf{Q}} \rangle = \sqrt{N}\psi_2$, where

$\psi_{1,2}$ are complex order parameters. In real space,

$$\langle a_{\mathbf{r}}\rangle = \frac{1}{\sqrt{N}}\sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}}\langle a_{\pm\mathbf{k}}\rangle = \psi_1 e^{i\mathbf{Q}\cdot\mathbf{r}} + \psi_2 e^{-i\mathbf{Q}\cdot\mathbf{r}}. \quad (7)$$

The ground state energy, per site, of the *uniform* condensed ground state is expanded in powers of $\psi_{1,2}$ as

$$E_0/N = -\mu(|\psi_1|^2+|\psi_2|^2) + \frac{1}{2}\Gamma_1(|\psi_1|^4+|\psi_2|^4) + \Gamma_2|\psi_1|^2|\psi_2|^2 + \Gamma_3((\bar{\psi}_1\psi_2)^3 + \text{h.c.})... \quad (8)$$

where $\bar{\psi}_j$ denotes complex conjugated of ψ_j, dots stand for higher order terms, and we omitted a constant term. We verified [19] that higher orders in ψ_j do not modify our analysis.

Whether the state at $\mu = 0+$ is co-planar or chiral is decided by the sign of $\Gamma_1 - \Gamma_2$ [12]. For $\Gamma_1 < \Gamma_2$, it is energetically favorable to break Z_2 symmetry between condensates and choose $\psi_1 \neq 0, \psi_2 = 0$ or vice versa. Parameterizing the condensate as $\psi_1 = \sqrt{\rho}e^{i\varphi}$, where $\rho = \mu/\Gamma_1$, and using Eq.(7), we obtain the cone configuration, Eq.(1). The order parameter manifold of this state is $U(1)\times Z_2$.

When $\Gamma_1 > \Gamma_2$, it is energetically favorable to preserve Z_2 symmetry and develop both condensates with equal magnitude $\rho = \mu/(\Gamma_1+\Gamma_2)$, i.e., set $\psi_1 = \sqrt{\rho}e^{i\theta_1}, \psi_2 = \sqrt{\rho}e^{i\theta_2}$. This corresponds to co-planar state with the common phase $\varphi = (\theta_1+\theta_2)/2$ and the relative phase $\theta = (\theta_1-\theta_2)/2$. The order parameter in this state is given by Eq. (2) with $\mathbf{Q}$ equal to either $\mathbf{Q}_{\rm i}$ ($J-J'$ model) or $\mathbf{Q}_0$ (XXZ model). For $\mathbf{Q} = \mathbf{Q}_{\rm i}$, the state is incommensurate co-planar configuration in Fig. 1. The order parameter manifold of this state is $U(1)\times U(1)$, where one $U(1)$ is associated with φ and the other with θ. For $\mathbf{Q} = \mathbf{Q}_0$, the co-planar order is commensurate. In this case, the symmetry is further reduced by Γ_3 term, which is allowed because $e^{i3\mathbf{Q}_0\cdot\mathbf{r}} = 1$ for all sites $\mathbf{r}$ of the lattice. This term locks the relative phase of the condensates θ to three values, reducing the broken symmetry to $U(1)\times Z_3$. For $\Gamma_3 < 0$, $\theta = \pi\ell/3$, where $\ell = 0, 1, 2$. For $\Gamma_3 > 0$, $\theta = (2\ell+1)\pi/6$. These are V and Ψ states in Figs. 1 and 2.

Accidental degeneracy of the isotropic model (3) in the classical limit shows up via $\Gamma_1^{(0)} = \Gamma_2^{(0)} = 9J$ and $\Gamma_3^{(0)} = 0$, where the superscript '0' indicates that these expressions are of zeroth order in $1/S$. We now analyze the situation in the presence of anisotropy and quantum fluctuations: first for $J-J'$ model with $J \neq J'$, and then for XXZ one with $J_z \neq J$.

Phases of the $J-J'$ model. We computed $\Gamma_{1,2}^{(0)}$ for classical spins, but in the presence of the the spatial anisotropy and found that it tilts the balance in favor of the cone phase: $\Delta\Gamma^{(0)} = \Gamma_2^{(0)} - \Gamma_1^{(0)} = J(1-J'/J)^2(2+J'/J)^2 > 0$. Quantum $1/S$ corrections, on the other hand, favor the co-planar state: $\Delta\Gamma^{(1)} < 0$. We obtained [19]

$$\Delta\Gamma^{(1)} = \frac{1}{16S}\sum_{\mathbf{k}\in\mathrm{BZ}}\Big(\frac{(J_0+5J_{\mathbf{k}})^2}{J_0-J_{\mathbf{k}}} - \frac{(J_0-4J_{\mathbf{Q}+\mathbf{k}})^2}{J_{\mathbf{Q}+\mathbf{k}}-J_{\mathbf{Q}}}\Big) + \frac{3J}{8S} \approx -\frac{1.6J}{S}. \quad (9)$$

Combining classical and quantum contributions, we find that

$$\Delta\Gamma = \Delta\Gamma^{(0)} + \Delta\Gamma^{(1)} = \frac{9(\delta J)^2}{J} - \frac{1.6J}{S} \quad (10)$$

where, we remind, $\delta J \equiv J - J'$. We see that $\Delta\Gamma < 0$ for $\delta J < \delta J_c = 0.42J/\sqrt{S}$, and $\Delta\Gamma > 0$ for larger δJ. The condition $\Delta\Gamma = 0$ selects the point B in Fig. 1 [17].

Split transitions near $\delta J_{\rm c}$. At $\mu = 0+$, the transition between incommensurate planar and cone phases is of first order with no hysteresis. We now analyze how this transition occurs at a finite positive $\mu \neq 0$. We start in the cone state to the right of point B in Fig. 1 and move to smaller δJ. Suppose that the condensate in the cone state has momentum $+\mathbf{Q}_{\rm i}$. Then Goldstone spin-wave mode is at $\mathbf{k} = \mathbf{Q}_{\rm i}$, while excitations near $\mathbf{k} = -\mathbf{Q}_{\rm i}$ have a finite gap. We computed the excitation spectrum $\omega_{\mathbf{k}}^{(1)}$ with quantum $1/S$ corrections and found [19] that near $\mathbf{k} \approx -\mathbf{Q}_{\rm i}$

$$\omega_{\mathbf{k}}^{(1)} \approx \frac{3J}{4}\left[(k_x+\bar{Q}_{\rm i})^2 + k_y^2 + \epsilon_{\min}\right], \quad (11)$$

$$\epsilon_{\min} = \frac{12\mu}{h_{\rm sat}J^2}\left[(\delta J)^2 - (\delta J_{\rm c})^2\left(1+\frac{\mu}{h_{\rm sat}}\right)\right], \quad (12)$$

where $\bar{Q}_{\rm i} = Q_{\rm i} + (4\pi/3 - Q_{\rm i})(3\mu/h_{\rm sat}) \approx Q_{\rm i} + 1.45\mu/(h_{\rm sat}\sqrt{S})$. The cone state becomes unstable at $\epsilon_{\min} = 0$, i.e., at $\delta J_{c3} \approx \delta J_{\rm c}(1+\mu/(2h_{\rm sat}))$, and gives rise to magnon condensation with momentum $(-\bar{Q}_{\rm i}, 0)$, which is *different* from $-\mathbf{Q}_{\rm i}$. The condensation of magnons with $(-\bar{Q}_{\rm i}, 0)$ then gives rise to a secondary cone order, with momentum not related by symmetry to that of the primary cone order. The resulting spin configuration is a double cone with $U(1)\times U(1)\times Z_2$ order parameter manifold. The primary condensate sets the transverse component of $\langle \mathbf{S}_{\mathbf{r}}^{\perp}\rangle = \langle S_{\mathbf{r}}^x + iS_{\mathbf{r}}^y\rangle$ to be $\exp[i\mathbf{Q}_{\rm i}\cdot\mathbf{r} + i\theta_1]$ and the second condensate adds $\exp[-i\bar{\mathbf{Q}}_{\rm i}\cdot\mathbf{r} + i\theta_2]$.

At smaller $\delta J \leq \delta J_{c3}$ the position of the minimum in $\omega_k^{(1)}$ in (11) evolves and drifts towards $-\mathbf{Q}_{\rm i}$. Once it reaches $-\mathbf{Q}_{\rm i}$, at $\delta J = \delta J_{c2}$, the two cone configurations interfere constructively and give rise to an incommensurate co-planar state. Critical δJ_{c2} can be estimated by requiring that $\omega_k^{(1)} = 0$ at $\mathbf{k} = -\mathbf{Q}_{\rm i}$. This yields $\delta J_{c2} = \delta J_{c3}(1 - O(\mu/h_{\rm sat})) < \delta J_{c3}$. Therefore the transformation from a cone to an incommensurate co-planar state at at a finite μ occurs via two transitions at δJ_{c2} and δJ_{c3} and involves an intermediate double cone phase (Fig. 1).

Instability of the V phase. We now return to Eq. (8) and consider the transition between the V phase and the incommensurate co-planar phase. At $\mu = 0+$, this transition holds at infinitesimally small δJ (point A in Fig. 1). We show that at a finite μ, the V phase survives up to a finite $\delta J_{c1} \sim (J/\sqrt{S})(\mu/h_{\rm sat})$. The argument is that in the V phase $\mathbf{Q} = \mathbf{Q}_0$ is commensurate and Γ_3 term in Eq. (8) is allowed. We recall that at $\delta J = 0$ and for classical spins $\Gamma_3 = 0$. We computed the classical contribution to Γ_3 at $\delta J > 0$ and the contribution due to quantum fluctuations at

$\delta J = 0$. We found [19] that the classical contribution vanishes, but the quantum contribution is finite to order $1/S^2$ and makes Γ_3 negative:

$$\Gamma_3 = \Gamma_3^{(2)} = \frac{3}{32S^2}\sum_{\mathbf{k}\in\mathrm{BZ}}\Big(\frac{(5J_{\mathbf{k}}+J_0)(5J_{\mathbf{Q}+\mathbf{k}}+J_0)J_{\mathbf{Q}-\mathbf{k}}}{(J_0-J_{\mathbf{k}})(J_0-J_{\mathbf{Q}+\mathbf{k}})} - -\frac{(5J_{\mathbf{k}}+J_0)(J_{\mathbf{k}}+J_0)}{2(J_0-J_{\mathbf{k}})}\Big) + \frac{3J_0}{64S^2} \approx -\frac{0.69J}{S^2} \qquad (13)$$

Because $\Gamma_3 < 0$, the V phase has extra negative energy compared to incommensurate phases, and one needs a finite δJ to overcome this energy difference.

We now argue that the transition at δJ_{c1} is of CIT kind. To see this, we allow for spatially non-uniform configurations of the condensate $\psi_{1,2}(\mathbf{r})$. This adds spatial gradient terms to (4): the isotropic Hamiltonian $\mathcal{H}_0$ produces conventional quadratic in gradient contribution $\propto \rho(\partial_x\theta)^2$, while $\delta\mathcal{H}_{\rm anis}$ (4) adds a *linear* gradient term $\propto \rho S\delta J\partial_x(\theta_1-\theta_2)$. Combining these two classical contributions with the quantum Γ_3 term in (8), we obtain the energy density for the relative phase $\theta = (\theta_1-\theta_2)/2$:

$$\mathcal{E}_\theta = \frac{3JS^2\mu}{4h_{\rm sat}}(\partial_x\theta)^2 + \frac{\sqrt{3}\delta JS^2\mu}{h_{\rm sat}}\partial_x\theta + S\frac{(\Gamma_3S^2)}{4}\frac{\mu^3}{h_{\rm sat}^3}\cos[6\theta] \qquad (14)$$

Eq. (14) is of standard sine-Gordon form, which allows us to borrow the results from [15]: the equilibrium value of θ shifts from the commensurate $\theta = \pi\ell/3$ in the V phase to an incommensurate value when the coefficient of the linear gradient term in (14) exceeds the geometric mean of the coefficients of two other terms in (14). Using Eq. (14) we find that CIT occurs at $\delta J_{c1} = 1.17(J/\sqrt{S})(\mu/h_{\rm sat}) = 0.13\mu/S^{3/2}$ (line AC in Fig. 1). At $\delta J > \delta J_{c1}$, θ acquires linear dependence on x, $\theta = \tilde{Q}x$. In this situation, the spin configuration becomes incommensurate but remains co-planar (Fig. 1).

Phases of $\mathcal{H}_{\rm xxz}$. For the XXZ model with exchange anisotropy, J and J' remain equal, but $J_z < J_\perp = J$ on all bonds. We verified [19] that $\mathbf{Q}$ remains commensurate for all $J_z/J \leq 1$, i.e., $\mathbf{Q} = \mathbf{Q}_0 = (4\pi/3, 0)$. In this situation, we found $\Gamma_2^{(0)} - \Gamma_1^{(0)} = -J_{\mathbf{Q}}(1-J_z/J) = 3J\Delta$. Quantum corrections to Γ_1 and Γ_2 are determined within the same isotropic model (3) and are given by (10). Using this, we immediately find that the ground state of the quantum XXZ model is coplanar for $\Delta \leq \Delta_{c2} = 0.53/S$ and is a cone for $\Delta > \Delta_{c2}$. The transition between co-planar and cone states near Δ_{c2} remains first-order for a finite $\mu > 0$, i.e., no intermediate double spiral state appears. This is the consequence of the fact that $\mathbf{Q} = \mathbf{Q}_0$ remains commensurate. Still, the transformation from the V phase to the cone phase does involve a new intermediate state, which comes about due to the change of sign of Γ_3. Exchange anisotropy Δ gives rise to a positive Γ_3 to order $1/S$: $\Gamma_3^{(1)} = J(1+2J_z/J)(1-J_z/J)/(2S) \approx 3J\Delta/(2S)$ (see [19] for details). At the same time the quantum corrections give rise to *negative* Γ_3 to order $1/S^2$ already at $\Delta = 0$, see (13). Combining the two, we find that

$$\Gamma_3 = \Gamma_3^{(1)} + \Gamma_3^{(2)} = \frac{3J\Delta}{2S} - \frac{0.69J}{S^2}. \qquad (15)$$

changes sign at $\Delta_{c1} = 0.45/S < \Delta_{c2} = 0.53/S$. At smaller $\Delta < \Delta_{c1}$, $\Gamma_3 < 0$, and the spin configuration is the V state (the energy is minimized by setting $\cos 6\theta = 1$, see (8)). However, in the interval $\Delta_{c1} < \Delta < \Delta_{c2}$, $\Gamma_3 > 0$ becomes positive. The energy is now minimized by $\cos 6\theta = -1$, which corresponds to the Ψ state in Fig. 2. The transition is highly unconventional symmetry-wise because the order parameter manifold is $U(1) \times Z_3$ in both phases, but extends to a larger $U(1) \times U(1)$ symmetry at the transition point.

We present the phase diagram of XXZ model in Fig. 2. A very similar phase diagram has been recently obtained in the numerical cluster mean-field analysis of the $S = 1/2$ XXZ model [18].

To summarize, in this paper we considered anisotropic 2D Heisenberg antiferromagnets on a triangular lattice in a high magnetic field close to the saturation. We analyzed the cases of spatially anisotropic interactions, like in Cs_2CuCl_4 and Cs_2CuBr_4 and of exchange anisotropy, as in $Ba_3CoSb_2O_9$. We showed that the phase diagram in field/anisotropy plane is quite rich due to competition between classical non-coplanar and quantum co-planar orders. This competition leads to multiple transitions and highly non-trivial intermediate phases, including a novel double cone state.

The analysis of this paper can be easily extended to quasi-2D layered systems, with inter-layer antiferromagnetic interaction $0 < J'' \ll J$. This additional exchange interaction leads to the staggering of coplanar spin configurations, of either V or Ψ kind, between the adjacent layers, as can easily be seen by treating $\varphi \to \varphi_z$ in Eq.(2) as layer-dependent variable with discrete index z. One then immediately finds that $J''\sum_{\mathbf{r},z}\vec{S}_{\mathbf{r},z}\cdot\vec{S}_{\mathbf{r},z+1}$ is minimized by $\varphi_z = \varphi + \pi z$, in agreement with earlier spin-wave [21] and Monte Carlo [9] studies.

We acknowledge useful conversations with L. Balents and C. Batista. This work is supported by DOE grant DE-FG02-ER46900 (AC) and NSF grant DMR-12-06774 (OAS and WJ).

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