Spin-Wave Instabilities and Non-Collinear Magnetic Phases of a Geometrically-Frustrated Triangular-Lattice Antiferromagnet

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This paper examines the relation between the spin-wave instabilities of collinear magnetic phases and the resulting non-collinear phases for a geometrically-frustrated triangular-lattice antiferromagnet in the high spin limit. Using a combination of phenomenological and Monte-Carlo techniques, we demonstrate that the instability wave-vector with the strongest intensity in the collinear phase determines the wave-vector of a cycloid or the dominant elastic peak of a more complex non-collinear phase. Our results are related to the observed multi-ferroic phase of Al-doped CuFeO2.

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It is well-known that the transition between different magnetic ground states may be signaled by the softening of a spin-wave (SW) mode. In the simplest case of a conventional square-lattice antiferromagnet, the softening of a SW mode at wave-vectors $(\pi, 0)$ and $(0, \pi)$ signals the spin flop and canting of the magnetic moments at a critical field. In the manganites [1], the SW instabilities of the ferromagnetic state have been used to construct the phase diagram for the antiferromagnetic (AF) phases that appear with Sr doping. The softening of a SW excitation at $(\pi, 0)$ signals the instability of the Néel state and the canting of the spins in a spin-1/2 union-jack lattice [2]. But the relation between the SW instabilities of a collinear phase and a resulting non-collinear phase is less clear when multiple SW instabilities occur simultaneously or when the non-collinear phase has a complex magnetic structure with several elastic peaks. This paper explores the relation between the SW instabilities of the collinear 4 and 8-sublattice (SL) phases of a geometrically-frustrated triangular-lattice antiferromagnet (TLA) and the non-collinear phases that appear with decreasing anisotropy $D$. When multiple SW instabilities of the collinear phase occur at once, the instability wave-vector with the largest intensity determines the dominant ordering wave-vector of the resulting non-collinear phase. One of the predicted non-collinear phases may be related to the multi-ferroic phase that appears in CuFeO2 with Al doping [3].

Frustrated TLAs with AF nearest-neighbor exchange $J_1 < 0$ exhibit a remarkable number of competing ground states [3]. With interactions $J_i$ up to third nearest neighbors (denoted in Fig 1) and assuming Ising spins along the $z$ direction, Takagi and Mekata [5] obtained a phase diagram with ferromagnetic (FM), 2-SL, 3-SL, 4-SL, and 8-SL phases. A portion of that phase diagram is sketched in Fig. 1. For the geometrically-frustrated TLA CuFeO2 in fields below 7 T, the ground state is the 4-SL phase [4, 7] and the black dot in Fig. 1 denotes the estimated ratio of exchange parameters $J_3/|J_1| \approx -0.44$.

FIG. 1: (Color online) A portion of the TLA phase diagram for large $D$ and $J_1 < 0$. The dashed (red) curve divides the 4-SL phase into 4-SL I and 4-SL II regions. For the 4- and 8-SL phases, red circles denote up spins and blue circles denote down spins; the solid (gray) line defines the unit cell. The dashed (blue) line obeys the relation $J_3 = 1.3J_2$, the black circle is the estimated location of the exchange parameters for CuFeO2, and the white circle lies on the boundary between the 4-SL I and 4-SL II regions.

$J_3/|J_1| \approx -0.57$ [8].

As demonstrated by the small SW gap of about 0.9 meV [3, 8] on either side of the ordering wave-vector $q = \pi x$, the spin fluctuations of CuFeO2 are much softer than would be expected for Ising spins. With Heisenberg spins, the collinear magnetic phases of a TLA become locally unstable below a critical anisotropy $D_c$ that depends on the exchange parameters $J_i$. The observed softening of the SW modes in CuFeO$_{2-x}$Al$_2$O$_2$ with Al doping [3] can be reproduced by lowering $D$ towards $D_c$ [10] in the 4-SL I region of Fig. 1. For Al concentrations above about $x_c \approx 0.016$, the magnetic ground state of CuFe$_{1-x}$Al$_x$O$_2$ becomes non-collinear and dis-
plays multi-ferroic properties [11, 12, 13].

We have determined the ground-state magnetic phases of the TLA using a combination of Monte-Carlo (MC) simulations and phenomenological techniques. The TLA Hamiltonian is

\[ H = -\frac{1}{2} \sum_{i \neq j} J_{ij} S_i \cdot S_j - D \sum_i S_{iz}^2, \]

(1)

where \( J_{ij} \) includes first, second, and third-neighbor interactions (shown in Fig. 1). The nearest-neighbor distance has been set to 1. The SW frequencies of the collinear phases are obtained by performing a Holstein-Primakoff 1/S expansion about the classical limit. Above the critical anisotropy \( D_c \), a collinear phase is locally stable if the SW frequencies \( \omega(k) \) are positive and real for every momentum \( k \).

MC simulations were used to find the non-collinear magnetic phases of the TLA. The simulations were started at a high-enough temperature to rule out metastable states. To mimic the process of thermal annealing, the system was slowly cooled to a final temperature (in units of \( |J_1|/S^2 \)) ranging from \( 4 \times 10^{-3} \) to \( 1 \times 10^{-4} \). Lowering the final temperature further did not significantly change the resulting non-collinear phase. Using lattices of varying sizes with periodic boundary conditions, we found that there was no substantial change for lattices greater than \( 16 \times 16 \).

In Fig. 1 the 4-SL phase is separated into regions I and II by the curve \( J_1/J_2 - 2 = J_2/J_1 \). In region 4-SL II, the instability wave-vectors are given by \( \mathbf{k} = (\pi/3)\mathbf{x} \), independent of the exchange parameters; in region 4-SL I, the instability wave-vectors depend on the exchange parameters [14]. With the exchange parameters corresponding to the black circle in region 4-SL I, we determined the stable magnetic phases as a function of \( D \).

As shown in Fig. 2, the 4-SL phase is stable down to \( D/|J_1| \approx 0.27 \), below which MC simulations obtain the complex non-collinear (CNC) phase shown on the top of Fig. 2. Since the same up or down spin frequently occurs at sites \( \mathbf{R} = n\mathbf{x} + n\sqrt{3}\mathbf{y} \) and \( \mathbf{R'} = \mathbf{R} + x/2 + \sqrt{3}y/2 \) or \( \mathbf{R''} = \mathbf{R} - x/2 + \sqrt{3}y/2 \), the CNC phase retains some of the same correlations present in the 4-SL phase. Translationally invariant in the \( y \) direction, the CNC phase has a period in the \( x \) direction between 2 and 3 lattice constants. Because the MC simulations were performed on a finite lattice, the energy of the CNC phase is overestimated and we cannot say whether this phase is commensurate or incommensurate.

Below a second threshold value of \( D/|J_1| \approx 0.08 \), a cycloid like the one sketched in the bottom panel of Fig. 2 has a lower energy than the CNC phase. As discussed below, the wave-vector of the cycloid is independent of \( D \). If the CNC phase were neglected, then the cycloid would achieve a lower energy than the 4-SL phase below \( D/|J_1| \approx 0.2 \), still above the critical value \( D_c/|J_1| \approx 0.15 \) for the local stability of the 4-SL phase.

To gain a better understanding of the phases stabilized within the TLA, we have evaluated the magnetic phases along the line with \( J_3/J_2 = 1.3 \) drawn through the black dot in Fig. 1. Five stable phases are presented as a function of \( |J_1|/D \) and \( |J_2|/D \) in Fig. 3. The 4-SL phase is stable along a strip through the diagonal of this plot. Although not indicated by this figure, the 4-SL region disappears above \( |J_1|/D \approx 40 \). Close to the origin or for large \( D \), a collinear 8-SL region is indicated in Fig. 1. Two cycloids are also obtained: in the upper left, cycloid II with wave-vector \( 4\pi x/3 \); in the lower right, cycloid I with the variable wave-vector indicated in the figure [15]. Finally, a CNC phase appears just below the 4-SL phase and disappears above \( |J_1|/D \approx 20 \). Regions of local stability for the collinear phases are indicated in Fig. 3 by the dashed black lines [14]. The results in Fig. 2 can be obtained from Fig. 3 by drawing a line from the origin with slope \( 2.28 \) (gray) so that \( J_2/|J_1| = -0.44 \), which passes from the 4-SL phase through the CNC phase into cycloid I.

The classical energies of each of these phases can be written as \( E/S^2 = A_1 J_1 + A_2 J_2 + A_3 J_3 - A_D D \). The coefficients for each phase are given in Table I. Only a non-collinear phase with \( 0.5 < A_D < 1 \) can intercede between a collinear phase with \( A_D = 1 \) and a cycloid with \( A_D = 0.5 \). For the CNC phase with \( A_D \approx 0.71 \), the error bars indicate the range of parameters obtained from MC simulations near \( |J_1|/D = 5.7 \) and \( |J_2|/D = 2.5 \). This phase is characterized by rather weak next-neighbor correlations with small \( |A_2| \). The CNC phase space in Fig. 3 is obtained by using the results of Table I to evaluate the energies of the MC spin configurations as
functions of $|J_1|/D$ and $|J_2|/D$. Hence, the CNC region may be underestimated.

The ordering wave-vector $\mathbf{q} = q\mathbf{x}$ of cycloid I is evaluated by minimizing $E$ with respect to $q$. So $q$ depends only on the ratios $J_2/J_1$ and $J_3/J_2$, as indicated by the diagonal lines in Fig. 3. Cycloid II with $q = 4\pi/3$ corresponds to the 120° Néel state found in a classical TLA with $D = 0$ and nearest and next-nearest neighbor interactions [10] when $|J_2/J_1| < 1/8$. A slightly distorted Néel state is stable for nonzero $D$ over a range of exchange parameters with $|J_3/J_2| > 1/2$, so that the diagonal line in Fig. 1 passes through the 4-SL and 8-SL phases. MC simulations were used to confirm the stability of cycloids I and II in Fig. 3.

With decreasing $D$ or moving away from the origin of Fig. 3 along a diagonal, the 4-SL phase becomes unstable either to cycloid II or to the CNC phase. The white line bisecting region the 4-SL strip in Fig. 3 corresponds to the white point in Fig. 1 at the border between the 4-SL I and 4-SL II regions with $J_2/J_1 = -0.36$. In region 4-SL II or above the white diagonal line, the 4-SL phase has instabilities at the wave-vectors $(\pi \pm \pi/3)x$. The SW intensity at the larger of these two wave-vectors always dominates and the 4-SL phase evolves into cycloid II with wavector $4\pi x/3$.

In region 4-SL I, the 4-SL phase has three unique SW instabilities: one at wave-vector $k_1$ along the $x$ axis, another at $k_2$ rotated by $\pi/3$, and a third at $k_3$ rotated by $-\pi/3$. All three have the same magnitude with $\pi/2 < k_i < \pi$. Other SW instabilities in the 4-SL I-region can be related by a symmetry operation to one of these three. We find that the instability at $k_1$ always has a larger intensity than the “twins” at $k_2$ or $k_3$ or than any of the other wave-vectors related by symmetry. Correspondingly, cycloid I along any diagonal in Fig. 3 has the same wave-vector $\mathbf{q}$ as the dominant instability of the 4-SL phase.

Similar conclusions are reached for the 8-SL phase, which switches to cycloid I along any diagonal in Fig. 3 again. Although the SW instability of the 8-SL phase occurs simultaneously at several wave-vectors, the dominant wave-vector instability of the 8-SL phase coincides with the wave-vector $\mathbf{q}$ of cycloid I along any diagonal in Fig. 3.

However, the CNC phase that intercedes between the

![Figure 3](image-url)

**FIG. 3:** (Color online) Phase diagram for the TLA as a function of $|J_1|/D$ and $|J_2|/D$ with $J_3 = 1.3J_2$ containing five regions: 4-SL (blue), 8-SL (green), CNC (violet), cycloid I (variable green-orange), and cycloid II (maroon). The dashed (white) line separates regions 4-SL I and 4-SL II. The dotted (black) curves denote the metastable boundaries for the 4-SL and 8-SL regions. Cycloid I has wave-vectors $\mathbf{q}$ that range from $0.684\pi$ to $0.923\pi$ in intervals of $0.016\pi$.

![Figure 4](image-url)

**FIG. 4:** (Color online) (a-b) SW frequency and intensity versus wave-vector $k_x$ for the 4-SL phase with $|J_1|/D = 5.5$, $J_3 = 1.3J_2$, where $|J_2|/D$ varies from 1.30 to 2.67. (c) Fourier transform for the $S_z$, $S_y$, and $S_x$ components of the CNC phase with the same exchange parameters as above and $|J_2|/D = 2.5$. 

MC simulations, the dominant wave-vector tic peaks shown in Fig. 4(c). Within the precision of our MC simulations, this distortion is undetectable and the SW gaps for both phases diverge.

The CNC phase may be related to the multi-ferroic phase observed in Al-doped CuFeO$_2$ [2], which was recently investigated by Nakajima et al. [17]. Based on neutron-scattering measurements, those authors concluded that the ground state is a modified cycloid with the same spin on sites $\mathbf{R}$ and $\mathbf{R}'$ (see above). This phase has peaks at wave-vectors on either side of $\pi \mathbf{x}$, in agreement with the neutron measurements. However, a modified cycloid cannot be stabilized by a Hamiltonian with the form of Eq. (1), regardless of the exchange and anisotropy parameters. With an additional phase slip $\delta$ for the spins at sites $\mathbf{R}'$, a pure cycloid with $\delta = 0$ and a single elastic peak always has lower energy than the phase proposed in Ref. [17] with $\delta = -q/2$. This conclusion has been verified by MC simulations.

Like the non-collinear phase proposed earlier [17], the CNC phase also contains FM correlations between sites $\mathbf{R}$ and $\mathbf{R}'$ or $\mathbf{R}''$. So the CNC phase also has elastic peaks on either side of $\pi \mathbf{x}$ at $k_z \approx 0.87\pi$ and $1.13\pi$, as shown in Fig. 3(c). Because the FM correlations are not perfect and vary along the $x$ direction, the CNC phase contains several other elastic peaks that may allow it to be experimentally distinguished from the phase proposed in Ref. [17].

To summarize, we have shown that the dominant wave-vector of a non-collinear phase in a frustrated TLA corresponds to the dominant instability wave-vector of a collinear phase as the anisotropy is lowered and spin fluctuations become softer. The CNC phase sketched in Fig. 2 is a more reasonable candidate for the multi-ferroic phase observed in Al-doped CuFeO$_2$ than the one previously proposed.

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| Phase   | $A_1$   | $A_2$   | $A_3$   | $A_D$  |
|---------|---------|---------|---------|--------|
| 4-SL    | 1       | -1      | 1       | 1      |
| 8-SL    | 0       | 1       | 1       | 1      |
| Cycloid I | $-(\cos(q) + 2\cos(q/2))$ | $-(1 + 2\cos(3q/2))$ | $-(\cos(2q) + 2\cos(q))$ | 1/2 |
| Cycloid II | 3/2     | -3      | 3/2     | 1/2    |
| CNC     | 0.595±0.001 | -0.097±0.001 | 1.161±0.001 | 0.712±0.001 |

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