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

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This Letter examines the relation between the spin-wave instabilities of collinear magnetic phases and the resulting noncollinear 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 noncollinear phase. Our results are related to the observed multiferroic phase of Al-doped CuFeO2.

It is well known that the transition between different magnetic ground states may be signaled by the softening of a spin-wave (SW) mode [1]. In the simplest case of a conventional square-lattice antiferromagnet, the softening of a SW mode at wave vectors (0, 0) and (0, π) signals the spin flop and canting of the magnetic moments at a critical field. In the manganites [2], the SW instabilities of the ferromagnetic state have been used to construct the phase diagram for the antiferromagnetic (AFM) phases that appear with Sr doping. The softening of a SW excitation at (π, 0) signals the instability of the Néel state and the canting of the spins in a spin-1/2 union-jack lattice [3]. But the relation between the SW instabilities of a collinear phase and a resulting noncollinear phase is less clear when multiple SW instabilities occur simultaneously or when the noncollinear phase has a complex magnetic structure with several elastic peaks. This Letter 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 noncollinear phases that appear with decreasing anisotropy $D$. We show that 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 noncollinear phase [4]. One of the predicted noncollinear phases may be related to the multiferroic phase that appears in CuFeO$_2$ with Al doping [5].

Frustrated TLAs with AFM nearest-neighbor exchange $J_1 < 0$ exhibit a remarkable number of competing ground states [6]. With interactions $J_i$ up to third nearest neighbors (denoted in Fig. 1) and assuming Ising spins along the z direction, Takagi and Mekata [7] 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 CuFeO$_2$ in fields below 7 T, the ground state is the 4-SL phase [8,9] and the black dot in Fig. 1 denotes the estimated ratio of exchange parameters $J_2/|J_1| = -0.44$ and $J_3/|J_1| = -0.57$ [10,11].

As demonstrated by the small SW gap of about 0.9 meV [5,10] on either side of the ordering wave vector $q = \pi x$, the spin fluctuations of CuFeO$_2$ 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 $J_j$. The observed softening of the SW modes in CuFe$_{1-x}$Al$_x$O$_2$ with Al doping [5] can be reproduced by lowering $D$ towards $D_c$ [12] 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 displays multiferroic properties [13–15].

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

$$H = -\sum_{\langle i,j \rangle} J_{ij} S_i \cdot S_j - D \sum_i S_{ii}^2,$$

(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 $\omega_k^{(i)}$ of the collinear phases are obtained by performing a Holstein-Primakoff 1/$S$ expansion about the classical limit. The SW intensities $W_k^{(i)}$ are the prefactors in the spin-spin correlation function $S(k, \omega) = \sum \omega_k^{(i)} \delta(\omega - \omega_k^{(i)})$ [16]. The intensities $W_k^{(i)}$ correspond to the strength of the spin fluctuations associated with the wave vector $k$ and SW branch $i$.

To mimic the process of thermal annealing, the system was slowly cooled to a final temperature (in units of $T_0$) ranging from $4 \times 10^{-3}$ to $1 \times 10^{-4}$. Lowering the final temperature further did not significantly change the result.

In Fig. 1, the 4-SL phase is separated into regions I and II by the curve $J_1/J_2 - 2 = J_2/J_3$. In region 4-SL II, the instability wave vectors are given by $k = (\pi \pm \pi/3)x$, independent of the exchange parameters; in region 4-SL I, the instability wave vectors depend on the exchange parameters [4]. 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 $D/|J_1| = 0.27$, below which MC simulations obtain the complex noncollinear (CNC) phase shown on the top of Fig. 2. While the CNC phase is translationally invariant in the $y$ direction, it retains some of the FM correlations present in the 4-SL phase: the same up or down spin frequently occurs at sites $R = mx + n\sqrt{3}y$ and $R' = R + x/2 + \sqrt{3}y/2$ or $R'' = R - x/2 + \sqrt{3}y/2$. Although the MC boundary conditions prevent a definitive determination, the CNC phase is probably incommensurate in the $x$ direction.

Below a second threshold value of $D/|J_1| = 0.08$, a cycloid like the one sketched in the bottom panel of Fig. 2 [17] 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 removed, then the cycloid would achieve a lower energy than the 4-SL phase below $D/|J_1| = 0.2$, still above the critical value $D_c/|J_1| = 0.15$ for the local stability of the 4-SL phase. The cycloid energy in Fig. 2 was obtained numerically with the cycloid restricted by finite boundary conditions.

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

![FIG. 2 (color online). Energy as a function of $D/|J_1|$ for the 4-SL (black dashed), CNC (blue diamonds), and cycloid I (red circles) phases with $J_3/J_2 = 1.3$ and $J_1/J_2 = 2.28$. The bottom diagram shows the cycloid for arbitrary $q$. The top diagram shows the CNC phase, where local moments range from red (purely up, $+z$) to blue (purely down, $-z$) and arrows indicate moment direction.](image)

![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 (dark blue), 8-SL (light green), CNC (light violet), cycloid I (variable green-orange), and cycloid II (solid red). 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 $q$ that range from $0.684\pi$ to $0.923\pi$ in intervals of $0.016\pi$.](image)
The wave vectors \((\pi \pm \pi/3)\mathbf{x}\). The SW intensity at the larger of these two wave vectors always dominates and the 4-SL phase evolves into cycloid II with wave vector \(4\pi x/3\).

In region 4-SL I, the 4-SL phase has three unique SW instabilities: one at wave vector \(\mathbf{k}_1\) along the \(x\) axis, another at \(\mathbf{k}_2\) rotated by \(\pi/3\), and a third at \(\mathbf{k}_3\) 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 \(\mathbf{k}_1\) always has a larger intensity than the “twins” at \(\mathbf{k}_2\) or \(\mathbf{k}_3\) 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. 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 4-SL and cycloid I phases is characterized by several elastic peaks shown in Fig. 4(c). Within the precision of our MC simulations, the dominant wave vector \(\mathbf{k} = 0.87\pi x\) of the CNC phase coincides with the dominant instability wave vector of the 4-SL phase that precedes it.

To demonstrate how the magnetic ground state evolves from cycloid II into the 4-SL phase and then into cycloid I, we plot in Figs. 4(a) and 4(b) the SW frequencies and intensities versus wave vector for the 4-SL phase with \(J_1/D = -5.5\), \(J_3/J_2 = 1.3\), and \(|J_2|/D\) varying from 1.30 to 2.67. As \(|J_2|/D\) approaches the lower limit for the stability of the 4-SL phase, the SW intensity dominates at the cycloid II wave vector \(\mathbf{q} = 4\pi x/3\). At the upper limit, the cycloid I wave vector \(\mathbf{q} = 0.83\pi x\) dominates. We concluded that the wave vectors of the SW instabilities for the collinear phases correspond to the ordering wave vectors of the noncollinear phases.

The CNC phase may be related to the multiferroic phase observed in Al-doped CuFeO\(_2\) [5], which was recently investigated by Nakajima et al. [19]. 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

<table>
<thead>
<tr>
<th>Phase</th>
<th>(A_1)</th>
<th>(A_2)</th>
<th>(A_3)</th>
<th>(A_0)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4-SL</td>
<td>1</td>
<td>-1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>8-SL</td>
<td>0</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>Cycloid I</td>
<td>(-\cos(q) + 2\cos(q/2))</td>
<td>(-(1 + 2\cos(3q/2)))</td>
<td>(-\cos(2q) + 2\cos(q))</td>
<td>1/2</td>
</tr>
<tr>
<td>Cycloid II</td>
<td>3/2</td>
<td>3</td>
<td>3/2</td>
<td>1/2</td>
</tr>
<tr>
<td>CNC</td>
<td>0.595 ± 0.001</td>
<td>-0.097 ± 0.001</td>
<td>1.159 ± 0.001</td>
<td>0.712 ± 0.001</td>
</tr>
</tbody>
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wave vectors on either side of $\pi x$, 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 $R'$, a pure cycloid with $\delta = 0$ and a single elastic peak always has lower energy than the phase proposed in Ref. [19] with $\delta = -q/2$. This conclusion has been verified by MC simulations.

Like the noncollinear phase proposed earlier [19], the CNC phase also contains FM correlations between sites $R$ and $R'$ or $R''$. So the CNC phase also has elastic peaks on either side of $\pi x$ at $k_x = 0.87\pi$ and $1.13\pi$, as shown in Fig. 4(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. [19].

To summarize, we demonstrate for the first time that the dominant ordering wave vector of a noncollinear phase corresponds to the dominant instability wave vector of the preceding collinear phase. Thus, complex noncollinear phases can be more easily characterized by studying the SW instabilities of collinear phases with stronger anisotropy. While the connection between the SW instabilities and the dominant wave vector of the noncollinear phases was demonstrated numerically for a frustrated TLA, we hope to establish that connection more generally in future work. We also argue that the CNC phase sketched in Fig. 2 is a more reasonable candidate for the multiferroic phase observed in Al-doped CuFeO$_2$ than the one previously proposed.

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[17] Because of the anisotropy, the cycloids will be slightly distorted by the tilting of the spins towards the $\pm z$ directions with an energy gain of order $D^2S^2/2|J_1|$ ($z = 6$ is the number of nearest neighbors). Within the precision of our MC simulations, this distortion is undetectable and a pure cycloid is obtained.