Critical anisotropies of a geometrically frustrated triangular-lattice antiferromagnet

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This work examines the critical anisotropy required for the local stability of the collinear ground states of a geometrically frustrated triangular-lattice antiferromagnet (TLA). Using a Holstein-Primakoff expansion, we calculate the spin-wave frequencies for the one-, two-, three-, four-, and eight-sublattice (SL) ground states of a TLA with up to third neighbor interactions. Local stability requires that all spin-wave frequencies are real and positive. The two-, four-, and eight-SL phases break up into several regions where the critical anisotropy is a different function of the exchange parameters. We find that the critical anisotropy is a continuous function everywhere except across the two-SL/three-SL and three-SL/four-SL phase boundaries, where the three-SL phase has the higher critical anisotropy.

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I. INTRODUCTION

Geometrically frustrated systems exhibit many novel characteristics including noncollinear ground states and multiferroic properties.1 One of the best realizations of a geometrically frustrated triangular-lattice antiferromagnet (TLA) is CuFeO2, which contains stacked hexagonal planes of spin-5/2 Fe3+ ions. Accompanied by a phase transition from a collinear four-sublattice (SL) ground state to a noncollinear phase,2–5 CuFeO2 exhibits multiferroic properties above a small anisotropy. Here, the exchange coupling Jij and j is antiferromagnetic when Jij<0. Employing a HP transformation, the spin operators are given by $S_i=\alpha_i^1a_i$, $S_{iz}=\sqrt{2}\alpha_i^2a_i$, and $S_{iz}=\sqrt{2}\alpha_i^3a_i$. Expanded about the classical limit in powers of $1/\sqrt{S}$, the Hamiltonian can be written as $H=E+H_1+H_2+\cdots$. The first-order term $H_1$ vanishes when the spin configuration minimizes the energy $E$. The second-order term $H_2$ provides the dynamics of noninteracting SWs. Higher-order terms $H_{n>2}$ reflect the interactions between SWs. They are unimportant at low temperature and for small

$$H = -\frac{1}{2} \sum_{i<j} J_{ij} S_i \cdot S_j - D \sum_i S_i^2. \tag{1}$$

where $S_i$ is the local moment on site $i$ and $D$ is the single-ion anisotropy. Here, the exchange coupling $J_{ij}$ between sites $i$ and $j$ is antiferromagnetic when $J_{ij}<0$. Employing a HP transformation, the spin operators are given by $S_i=\alpha_i^1a_i$, $S_{iz}=\sqrt{2}\alpha_i^2a_i$, and $S_{iz}=\sqrt{2}\alpha_i^3a_i$. Expanded about the classical limit in powers of $1/\sqrt{S}$, the Hamiltonian can be written as $H=E+H_1+H_2+\cdots$. The first-order term $H_1$ vanishes when the spin configuration minimizes the energy $E$. The second-order term $H_2$ provides the dynamics of noninteracting SWs. Higher-order terms $H_{n>2}$ reflect the interactions between SWs. They are unimportant at low temperature and for small

In this paper, we evaluate the critical anisotropies required for the local stability of the collinear magnetic phases in a model TLA with up to third nearest neighbors. As shown elsewhere,13 the wave vector of the dominant SW instabilities of a collinear phase coincides with the dominant wave vector of the noncollinear phase that appears with decreasing anisotropy. Therefore, an analysis of the critical anisotropies and wave vectors of a frustrated TLA can provide useful information about the noncollinear phases that appear at small anisotropy.

The collinear ground states of a TLA with strong anisotropy were first obtained by Takagi and Mekata,14 who examined an Ising model with interactions up to third nearest neighbors. The ground-state phase diagram consists of the five phases sketched in Fig. 1, where the energies of these five states are given in Table I. Using a Holstein-Primakoff (HP) expansion, we have calculated the SW frequencies and critical anisotropies for each of these phases.

The Hamiltonian for a TLA is given by

![FIG. 1. (Color online) The one-, two-, three-, four-, and eight-SL phases for the ground states of the geometrically frustrated TLA. Solid black lines denote the magnetic unit cell of each phase. Up and down spins are designated by light red and dark blue circles, respectively.]
TABLE I. Classical energies and critical anisotropies for TLA sublattices.

<table>
<thead>
<tr>
<th>SL</th>
<th>Energy</th>
<th>(D_c)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1-SL</td>
<td>(E^{(1)}_{1}/NS^2 = -3J_1 - 3J_2 - 3J_3 - D)</td>
<td>(D_c^{(1)} = 0)</td>
</tr>
<tr>
<td>2-SL</td>
<td>(E^{(2)}_{1}/NS^2 = J_1 + J_2 - 3J_3 - D)</td>
<td>(D_c^{(2)} ) [Eq. (6)]</td>
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<td></td>
<td>(D_c^{(2)} ) [Eq. (6)]</td>
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<td></td>
<td>(D_c^{(2)} ) [Eq. (10)]</td>
<td>(D_c^{(2)} ) [Eq. (10)]</td>
</tr>
<tr>
<td>3-SL</td>
<td>(E^{(3)}_{1}/NS^2 = J_1 - 3J_2 + J_3 - D)</td>
<td>(D_c^{(3)} ) [Eq. (15)]</td>
</tr>
<tr>
<td>4-SL</td>
<td>(E^{(4)}_{1}/NS^2 = J_1 - J_2 + J_3 - D)</td>
<td>(D_c^{(4)} ) [Eq. (18)]</td>
</tr>
<tr>
<td>5-SL</td>
<td>(E^{(5)}_{1}/NS^2 = J_1 + J_2 - J_3 - D)</td>
<td>(D_c^{(5)} ) [Eq. (20)]</td>
</tr>
</tbody>
</table>

1/S. Similar to Takagi and Mekata, we consider nearest-neighbor \(J_1\), next-nearest-neighbor \(J_2\), and next-next-nearest-neighbor \(J_3\) exchange interactions, as sketched in Fig. 1. Whereas \(J_1 < 0\) is always antiferromagnetic, \(J_2\) and \(J_3\) can be either positive or negative.

To determine the SW frequencies \(\omega_{k}\), we solve the equation of motion for the vectors \(v_k = [a_k^{(1)}, a_k^{(3)}, a_k^{(5)}, ...]\), which may be written in terms of the \(2N \times 2N\) matrix \(M(k)\) as \(i d v_k / d t = - [H_2, v_k] = M(k)v_k\), where \(N\) is the number of spin sites in the unit cell. The SW frequencies are then determined from the condition \(\det[M(k) - \omega_{k}I] = 0\).

Two conditions are required for the local stability of any magnetic phase: all SW frequencies must be real and positive and all SW weights must be positive. The SW weights \(W_{k}\) are coefficients of the spin-spin correlation function

\[
S(k, \omega) = \frac{1}{N} \int dt e^{-i \omega t} \sum_{i,j} e^{i k d_{ij}} \langle S_i^x S_j^x (t) \rangle + \langle S_i^y S_j^y (t) \rangle.
\]

where \(s\) denotes a branch of the SW spectrum and \(d_{ij}\) is defined as the vector pointing from site \(i\) to site \(j\). The weights \(W_{k}\) were evaluated within the HP formalism by solving the equations of motion for coupled spin Green’s functions. In zero field, the condition that the SW weights are positive for all \(k\) is equivalent to the condition that all SW frequencies are positive.

We obtained analytic expressions for the SW frequencies for all phases shown in Fig. 1 with the exception of the eight-SL phase, which was solved numerically. Analyzed of the SW frequencies yields the critical anisotropy \(D_c\) and the critical wave vectors \(k_c\) where the SW frequencies vanish. To simplify the following discussion, the SW and anisotropy coefficients are provided in the Appendix.

II. ONE-SUBLATTICE PHASE

The one-SL phase [Fig. 1(a)] is a ferromagnet with SW frequencies

\[
\omega_{k}^{(1)} = 2S(D + A_{1k}).
\]

Since the one-SL phase is locally stable for any positive value of the anisotropy, \(D_c = 0\). The SW intensity \(W_{k}\) is constant throughout \(k\) for all interactions.

III. TWO-SUBLATTICE PHASE

For the two-SL phase [shown in Fig. 1(b)], the SW frequencies are given by

\[
\omega_{k}^{(2)} = 2S\sqrt{A_{2k}^2 - A_{3k}^2}.
\]

The SW weights for the two-SL phase are

\[
W_{k}^{(2)} = \sqrt{\frac{A_{2k} + A_{3k}}{A_{2k} - A_{3k}}}.
\]

From Eq. (4), the condition for the local stability of a two-SL phase is \(A_{2k}^2 - A_{3k}^2 > 0\). At \(D_c, A_{2k} = A_{3k}\). This condition is satisfied when \(D_c = 0\) in most of the two-SL phase. But approaching the three-, four-, and eight-SL phase boundaries, nonzero anisotropy is required for local stability. As shown in Fig. 2(a), the critical anisotropy is continuous across the four-SL and eight-SL boundaries but is discontinuous across the three-SL boundary.

Upon closer examination [Fig. 2(c)], we find that \(D_c\) depends differently on the exchange parameters in the three regions designated by roman numerals. In region 2I [bounded by \(J_3 = J_2/2, J_3 = (9J_2 - J_1)/12, J_3 = J_2/(J_1 - 2J_2)\)],

\[
D_c^{(2)} = \frac{1}{(4J_3)^3} \left[ -272J_1^2 + 64J_1^2 J_2 + 48J_1 J_2^2 + 72J_1 J_2^2 - 48J_2 J_1 J_2 - 8J_2^2 J_1^2 + 36J_2 J_1^2 J_2 - 27J_2^2 - (2J_2 - J_1)C^3 \right],
\]

where

\[
C = \sqrt{(2J_2 + 3J_1)^2 - 8J_2 J_1}.
\]

In region 2II [bounded by \(J_3 = J_2/2, J_3 = J_2, J_3 = (8J_2 - J_1)/9, J_3 = J_2/(J_1 - 2J_2)\)],

\[
D_c^{(2)} = 4J_2 - \frac{9}{2}J_3 - \frac{1}{2}J_1.
\]

Finally, in region 2III [bounded by \(J_3 = J_2/2, J_3 = J_1/4, \) and \(J_3 = (J_1 - J_2)/4\)],

\[
D_c^{(2)} = - \frac{(4J_1 + J_2 - J_3)^2}{2(J_2 + 2J_3)}.
\]

The critical wave vectors \(k_c\) for the SW instabilities in region 2I are

\[
k_c^{(2I,a)} = 2 \arccos \left( \frac{3J_2 - 2J_3 - C}{8J_3} \right).
\]
CRITICAL ANISOTROPIES OF A GEOMETRICALLY...

Two other instabilities $k^{(2b)}$ and $k^{(2c)}$ are related to $k^{(2a)}$ by $\pm \pi/3$ rotations and can be considered twins of the $k^{(2a)}$ instabilities. All three instabilities occur at the same critical anisotropy $D_c^{(2)}$. For regions 2II and 2III, the SW instabilities occur at

$$k_x^{(2II)} a = \pi \pm m/3,$$

$$k_y^{(2II)} a = 0$$

and

$$k_z^{(2III)} a = 0,$$

along the $k_x$ and $k_z$ axes, respectively. Region 2IV is bounded by $J_2=0$, $J_3=(8J_2-J_1)/9$, and $J_4=(J_1-J_2)/4$ as shown in Figs. 2(b) and 2(c). This region has no critical anisotropy $D_c^{(2IV)}$ and is therefore a stable collinear phase for all $D \geq 0$.

Figure 3(a) shows three representative SWs for all two-SL regions. The interaction parameters for region 2I are $J_2/|J_1|=-0.25$, $J_3/|J_1|=-0.12$, and $D_c/|J_1|=0.04$. For region 2II, $J_2/|J_1|=-0.10$, $J_3/|J_1|=-0.05$, and $D_c/|J_1|=0.325$. For region 2III, $J_2/|J_1|=-0.75$, $J_3/|J_1|=-0.125$, and $D_c/|J_1|=0.301$. Finally, for region 2IV, the interaction parameters are $J_2/|J_1|=-1.0$, $J_3/|J_1|=-0.125$, and $D_c/|J_1|=0.0$. Regions I, II, and IV were evaluated with $k_y a=0$ while region III was evaluated at $k_y a=0.186 \pi$ as explained above.

In Figs. 2(b) and 2(c), we examine the critical anisotropy of the two-SL phase along the $J_1/|J_1|=0$ axis. The critical anisotropy vanishes for $-1 < J_2/|J_1| < -1/8$ but is nonzero outside this region. Therefore, noncollinear phases should appear for $J_2/|J_1| < -1$ and $J_1/|J_1| > -1/8$ when $D < D_c$. This agrees with Jolicoeur et al.,$^{15}$ who studied a TLA with nearest- and next-nearest-neighbor exchange interactions and $D=0$. They obtain a Néel state up to $J_2/|J_1|=-1/8$ and an incommensurate spiral for $J_2/|J_1|<-1$. Similar results have been obtained on square lattices.$^{18,19}$

IV. THREE-SUBLATTICE PHASE

For the three-SL phase [shown in Fig. 1(c)], the SW frequencies are

$$\omega_{k}^{(3)} = 6S\sqrt{R_{4k}} \cos(\theta/3 + 2m/3 \pi) + R_{2k}/3,$$  

where $m$ is an integer (0,1,2) distinguishing the three separate SW dispersion relations and
The critical anisotropy of the three-SL phase is independent of $J_2$ and given by

$$D_c^{(3)} = -\frac{3}{2}(J_1 + J_3).$$

(15)

Notice that $D_c^{(3)}=0$ along the three-SL/one-SL boundary. Again, $D_c$ is discontinuous along the two-SL/three-SL and three-SL/four-SL boundaries: the anisotropy required for the local stability of the three-SL phase is 3 times the critical anisotropy of the two- or four-SL phases. As discussed further below, the discontinuities at the two-SL/three-SL and three-SL/four-SL phase boundaries are related to the distinction between the conditions for global and local stabilities.

In Fig. 3(b), we plot a SW dispersion in the three-SL phase with interaction parameters $J_2/|J_1|=0.5$, $J_3/|J_1|=-0.5$, and $D_c/|J_1|=2.25$. Since the three-SL phase has a net moment, the SW frequencies are quadratic functions of $\mathbf{k}$ near the instability wave vectors.

V. FOUR-SUBLATTICE PHASE

The SW frequencies for the four-SL phase [shown in Fig. 1(d)] were evaluated in Ref. 12 and are given by

$$\omega_k^{(4)} = 2S(A_{5k}^2 - A_{7k}^2) + [(F_{2k} - F_{2k}^2)^2$$

$$+ 4A_{6k}F_{2k} - A_{7k}F_{2k}^2(2\eta^2)^2)^{1/2}.\]$$

(16)

The SW weights of the four-SL phase are

$$W_k^{(4)} = [R_{5k}(A_{7k} - A_{6k}) + (F_{2k} + F_{2k}^2)(A_{6k} - A_{7k})^2$$

$$+ (F_{2k} - F_{2k}^2)^2(F_{2k} + F_{2k}^2 - A_{6k} - A_{7k})]$$

$$\times [R_{5k}A_{7k}^2 - A_{7k}^2 - R_{7k}^2]^{-1}.\]$$

(17)

As for the two-SL phase, the critical anisotropy $D_c$ for the four-SL phase depends differently on the interaction parameters in two regions, again denoted by roman numerals I and II [Fig. 2(b)]. In region I [bounded by $J_3=J_2/2$, $J_2=J_1/2$, and $J_3=J_2/(J_1-2J_2)$],

$$D_c^{(4I)} = \frac{1}{4J_3^2}[-16J_2^3 - 64J_2J_3^2 + 48J_2^2J_1 + 72J_2^2J_3 - 8J_2^3J_1$$

$$- 48J_2^2J_1J_3 + 36J_2^2J_3J_2 - 27J_2^2 + (2J_2 - J_3)C^3],$$

(18)

and in region II [bounded by $J_3=J_2/2$, $J_2=0$, and $J_3=J_2/(J_1-2J_2)$],

$$D_c^{(4II)} = 2J_2 - \frac{1}{2}J_1 - \frac{1}{2}J_1.\]$$

(19)

The critical wave vectors for the four-SL phase are the same as those in the respective region of the two-SL phase, including the multiple instabilities in region 2I: $k^\text{II}(4I) = k^\text{II}(4II), k^\text{II}(4I) = k^\text{II}(4II)$, and $k^\text{II}(4I) = k^\text{II}(4II)$. Figure 3(c) shows two representative SWs for regions 4I and 4II with $k_a=0$. The interaction parameters for region 4I are $J_2/|J_1|=-0.439$, $J_3/|J_1|=-0.570$, and $D_c/|J_1|=0.105$. For region 4II, they are

$$D_c^{(8I)} = D_c^{(8II)} + 4J_3 - J_1.\]$$

(20)

VI. EIGHT-SUBLATTICE PHASE

For the eight-SL phase [shown in Fig. 1(e)], we have determined the SW dispersion relations numerically. The critical anisotropy values for this phase are shown in Fig. 2(a). Notice that $D_c$ has a cusp dividing the phase into regions 8I and 8II [Fig. 2(b)], separated by $J_3=J_2/2$. Looking more closely at the numerical results, the critical anisotropies in the eight-SL regions are closely related to those of their respective neighbors and are given by

$$D_c^{(8I)} = D_c^{(8II)} + 4J_3 - J_1.\]$$

(20)
which clearly show that the critical anisotropies are continuous across the phase boundaries. In region 8II, the wave-vector instabilities occur for $k_y=0$ (as in region 4I); in region 8I, the wave-vector instabilities occur for nonzero $k_y$ (as in region 2II). Figure 3(d) shows two representative SWs for regions 8I and 8II. The interaction parameters for region 8I are $J_2/|J_1|=-1.5$, $J_3/|J_1|=-0.50$, and $D_c/|J_1|=0.25$. For region 8II, they are $J_2/|J_1|=-0.75$, $J_3/|J_1|=-0.50$, and $D_c/|J_1|=0.62$. Whereas $k_y a=0$ for region 8II and $k_y a=0.382\pi$ for region 8I as explained above.

To better understand the discontinuities along the two-SL/three-SL and three-SL/four-SL phase boundaries, we consider the relationship between local and global stabilities. Our SW calculations only guarantee the local stability of each collinear phase. But even when a phase is locally stable, it can still be globally unstable to a lower-energy spin configuration. Hence, the critical anisotropy $D_c$ for global stability must be greater than or equal to the critical anisotropy $\bar{D}_c$ for local stability. Unlike $D_c$, $\bar{D}_c$ must also be a continuous function of $J_1$, $J_2$, and $J_3$. So when $D_c$ is discontinuous, the phase with the lower critical anisotropy cannot be globally stable. Since the three-SL phase has a higher critical anisotropy along the two- and four-SL boundaries, the two-SL and four-SL phases cannot be globally stable along those boundaries when $D_c^{(4I)} < D_c^{(3)}$ or $D_c^{(4II)} < D_c^{(3)}$. Therefore, our results for the local stability of the collinear phases also have implications for the global stability of those phases.

\section{VII. Conclusion}

We have examined the critical anisotropy for a geometrically frustrated TLA. Based on the Takagi-Mekata phase diagram, we calculated the SW frequencies for all five phases. Imposing the two conditions for local stability, we obtained the critical anisotropies and wave-vector instabilities for all phases as functions of the exchange interactions.

Surprisingly, these results are highly dependent on the longer-range exchange interactions and most phases break into several regions where the anisotropy has a distinct dependence on the exchange parameters. As discussed for the two-SL and four-SL phases, the critical anisotropies and wave vectors for the local stability of the collinear phases provide useful information about the noncollinear phases that appear at small anisotropy. We have also shown that the discontinuity of the critical anisotropy at the two-SL/three-SL and three-SL/four-SL phase boundaries has implications for the global stability of the two-SL and four-SL phases with the smaller critical anisotropies.

It is expected that this investigation of the collinear phases of the TLA enables one to characterize the possible noncollinear phases at small anisotropy. An examination of the critical wave vectors for each collinear phase can provide useful information about the dominant ordering wave vector of the underlying noncollinear phase that may arise. We believe that an analysis similar to that performed in this paper can also provide useful information for a variety of other physically important frustrated systems.

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\section{Appendix: Spin-Wave and Anisotropy Coefficients}

This appendix provides the coefficients that enter the SW frequencies and weights for each phase. The coefficients for the one-SL or ferromagnetic phase are
\[ A_{1k} = \frac{3}{2} (J_1 + J_2 + J_3) - J_1 \cos(k \cdot d_1) \cos(k \cdot d_2) \]
\[ + \cos(k \cdot d_3) - J_2 \cos(k \cdot d_4) \]
\[ + \cos(k \cdot d_6) - J_3 \cos(2k \cdot d_1) + \cos(2k \cdot d_2) \]
\[ + \cos(2k \cdot d_3), \]
\[ (A1) \]

where \( d_1 = ax_1 + \sqrt{3}/2a_y, \quad d_2 = -1/2ax + \sqrt{3}/2a_y, \quad d_3 = 3/2ax + \sqrt{3}/2a_y, \quad d_4 = -3/2ax + \sqrt{3}/2a_y. \)

The two-SL phase coefficients are
\[ A_{2k} = D + 3J_3 - J_1 \cos(k \cdot d_1) + J_2 \cos(k \cdot d_3) + 1 \]
\[ - J_3 \cos(2k \cdot d_1) + \cos(2k \cdot d_2) + \cos(2k \cdot d_3), \]
\[ (A2) \]

The three-SL phase coefficients are
\[ R_{1k} = R_{2k}^2 - 3R_{3k}, \]
\[ (A4) \]
\[ R_{2k} = 2A_{4k} + A_{5k}, \]
\[ (A5) \]
\[ R_{3k} = A_{1k}^2 + 2A_{4k}A_{5k} + |F_{1k}|^2, \]
\[ (A6) \]

As in Ref. 12, the four-SL phase coefficients are
\[ R_{5k} = 2D + 2J_2 (3 - \cos(k \cdot d_3) - \cos(k \cdot d_5) - \cos(k \cdot d_6)), \]
\[ (A7) \]
\[ A_{5k} = 4J_1 + 6J_3 - A_{3k}, \]
\[ (A8) \]
\[ F_{1k} = J_1 (e^{-ik \cdot d_2} + e^{ik \cdot d_1} + e^{ik \cdot d_4}) \]
\[ + J_2 (e^{2ik \cdot d_2} + e^{-2ik \cdot d_1} + e^{-2ik \cdot d_4}). \]
\[ (A9) \]

As in Ref. 12, the four-SL phase coefficients are
\[ R_{5k} = (F_{2k}^4 + F_{2k}^2 - 2(F_{2k}^2 + 2A_{6k}A_{7k})F_{2k}^2 \]
\[ + 4A_{7k}^2 + A_{7k}^2 |F_{2k}|^2 - 4A_{6k}A_{7k}F_{2k}^2)^{1/2}, \]
\[ (A11) \]

The following equations hold for the three-SL phase coefficients
\[ A_{5k} = D - J_1 + J_2 (1 - \cos(k \cdot d_3) - J_3 (1 + \cos(2k \cdot d_3)), \]
\[ (A12) \]
\[ A_{7k} = - \cos(k \cdot d_1) (J_1 + 2J_3 \cos(\sqrt{3}k \cdot d_3)), \]
\[ (A13) \]
\[ F_{2k} = - \cos(k \cdot d_3/2) (J_1 e^{ik \cdot d_1}/2 + J_2 e^{-3ik \cdot d_1/2}). \]
\[ (A14) \]