Global stability and the magnetic phase diagram of a geometrically frustrated triangular lattice antiferromagnet

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While a magnetic phase may be both locally stable and globally unstable, global stability always implies local stability. The distinction between local and global stability is studied on a geometrically-frustrated triangular lattice antiferromagnet with single-ion anisotropy D that favors alignment along the z axis. Whereas the critical value $D_c^{\rm loc}$ for local stability may be discontinuous across a magnetic phase boundary, the critical value $D_c^{\rm glo} \geq D_c^{\rm loc}$ for global stability must be continuous. We demonstrate this behavior across the phase boundary between collinear three and four sublattice phases that are stable for large D. © *2011 American Institute of Physics*. [doi:10.1063/1.3553780]

Although quite well understood in many contexts, the distinction between local and global stability is seldom applied to the magnetic phase diagram of a complex system. This paper studies the distinction between local and global stability on a frustrated triangular-lattice (TL) antiferromagnet (AF) with single-ion anisotropy D that favors the alignment of classical spins along the z axis. With decreasing D, the collinear magnetic phases eventually become unstable to noncollinear spin states. The critical value for global stability $D_c^{\rm glo}$ must exceed or equal the critical value for local stability $D_c^{\rm loc}$ of any collinear phase. Whereas $D_c^{\rm loc}$ may be discontinuous across a magnetic phase boundary, $D_c^{\rm glo}$ must be a continuous function of the exchange parameters J_{ij}.

A TLAF with exchange interactions $J_1 < 0$, J_2, and J_3 (up to third nearest neighbors) is sketched in the inset to Fig. 1. Even for Ising spins, the TLAF has a rather complex phase diagram containing five collinear magnetic phases with 1, 2, 3, 5, or 8 sublattices (SLs).[1] A portion of the magnetic phase diagram with $J_2 > -|J_1|/2$ and $J_3 < 0$, shown in Fig. 1, contains 2SL, 3SL, and 4SL phases. The 2SL phase is a simple AF; the 3SL and 4SL phases are sketched in the inset to Fig. 1. The 4SL or $\uparrow\uparrow\downarrow\downarrow$ phase is particularly important because it appears at low temperatures in the hexagonal planes of pure $CuFeO_2$.[2]

The energy of a TLAF with anisotropy D and classical spins $\mathbf{S}_i$ is given by

$$E = -\frac{1}{2}\sum_{i\neq j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j - D\sum_i {S_{iz}}^2. \qquad (1)$$

The Ising limit is obtained as $D\rightarrow\infty$, which confines the spins to the z axis. As D decreases, the collinear phases eventually become unstable, first globally and then locally.

Local stability of a magnetic phase with classical spins can be tested by performing a $1/S$ expansion about the classical limit. A phase is locally stable if the spin-wave (SW) frequencies are real and the SW weights are positive.[3] The softening of a SW mode signals that the magnetic phase is no longer locally stable.[4] In recent work,[5] we evaluated the critical values $D_c^{\rm loc}$ for the local stability of all five collinear phases in the TLAF.

Surprisingly, the 2SL, 4SL, and 8SL regions break into subregions where the conditions for local stability are different. There are two distinct subregions of the 4SL phase. In subregion 4II, bordered by the solid lines and and to the right of the dashed curve $J_3 = J_2^2/(J_1 - 2J_2)$ in Fig. 1, the SWs soften at the wavevector $\mathbf{Q}_{\rm SW} = (4\pi/3)\mathbf{x}$ in the $\mathbf{x}$ direction (or in the equivalent hexagonal directions rotated by $\pm\pi/3$), regardless of the exchange interactions. But in subregion 4I, bordered by the solid lines and to the left of the dashed curve, the SWs soften at a wavevector $\mathbf{Q}_{\rm SW}$ that sensitively depends on the exchange interactions.[6] It is believed[7] that the exchange parameters of pure $CuFeO_2$ (neglecting the intralayer exchange) occupy subregion 4I with $\mathbf{Q}_{\rm SW} \approx 0.85\pi\mathbf{x}$.

The critical values for local stability of the collinear phases were obtained throughout the $\{J_2/|J_1|, J_3/|J_1|\}$ phase diagram with $J_1 < 0$.[5] We found that $D_c^{\rm loc}$ was continuous across all phase boundaries *except* those involving the 3SL phase: $D_c^{\rm loc}$ was discontinuous across both the 2SL-3SL and the 4SL-3SL phase boundaries with $D_c^{\rm loc}$ three times higher in the 3SL phase than in the neighboring 2SL and 4SL phases.

It is easy to prove that the critical anisotropy for global stability must be continuous across any phase boundary. Imagine that phases 1 and 2 have different values of $D_c^{\rm glo}$ such that phases 1 or $1'$ are globally stable for $J > 0$ when $D > D_{c1}$ or $D < D_{c1}$ and phase 2 is globally stable for $J < 0$ when $D > D_{c2}$ where $D_{c2} < D_{c1}$. Since phases 1 and 2 are degenerate at the $J = 0$ phase boundary for $D \geq D_{c1}$, their energies E_1 and E_2 must be equal when $D = D_{c1}$. However, for $D_{c1} > D > D_{c2}$ and $J = 0$, the energy $E_{1'}$ of phase $1'$ must be smaller than the energy E_2 of phase 2. Since the

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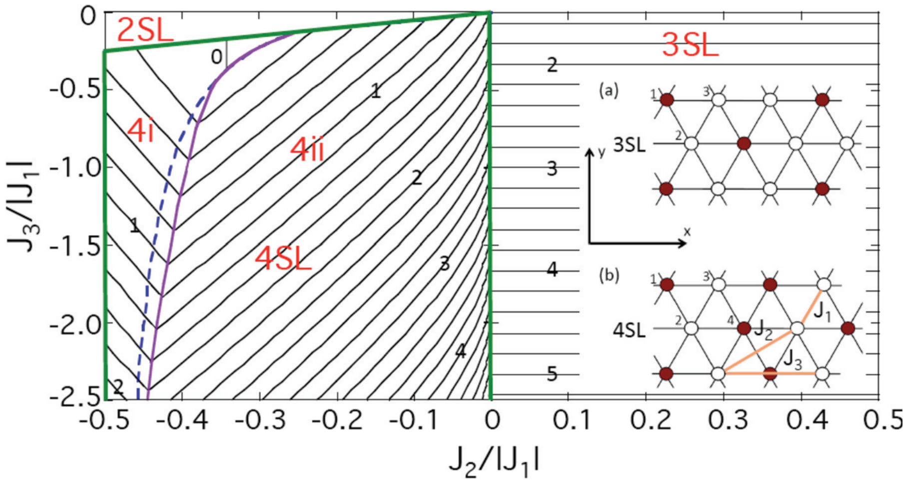

FIG. 1. (Color online) The phase diagram of a TLAF with interactions $J_1 < 0$, J_2, and J_3 denoted in the bottom inset. The region of stability for the 4SL phase with strong anisotropy D is bordered by the solid lines. The thin iso–anisotropy curves provide values for the critical global anisotropy $D_c^{\rm glo}/|J_1|$ in increments of 0.2. Spin states of the 3SL and 4SL phases with up (filled circles) and down (empty circles) spins are sketched in the inset.

energy must be a continuous function of J, this leads to a contradiction. Hence, the critical value for global stability must be continuous across the $J = 0$ phase boundary.

We conclude that the critical anisotropy for the global stability of the 4SL phase near the 4SL-3SL phase boundary must be at least three times higher then the critical anisotropy for its local stability. To obtain the globally stable phase, we use the recently developed technique of Fishman and Okamoto[8] to construct trial spin states containing odd-order harmonics of the fundamental wavevector Q:

$$S_z(\mathbf{R}) = A\left\{\cos(Qx) + \sum_{l=1}^{\infty} C_{2l+1}\cos(Q(2l+1)x)\right\}, \quad (2)$$

$$S_y(\mathbf{R}) = \sqrt{1 - S_z(\mathbf{R})^2}\,\mathrm{sgn}(\sin(Qx)), \quad (3)$$

where the amplitude A is fixed by the constraint that $\max|S_z(x)| = 1$ and the lattice constant is set to 1. Notice that $\mathbf{S}(\mathbf{R}) = \mathbf{S}(x)$ depends only on x. The anharmonic coefficients $C_{2l+1>1}$ reflect the deviation from a pure cycloid with $\mathbf{S}(x) = (0, \sin(Qx), \cos(Qx))$. The coefficients C_{2l+1} and the wavevector Q are treated as variational parameters that minimize the energy E of Eq.(1).

The energy E was minimized within a unit cell of length 5000 with open boundary conditions in the $\mathbf{x}$ direction. Doubling the length of the unit cell has no noticeable effect on the amplitudes C_{2l+1}. Throughout the phase space of Fig. 1, only the third and fifth harmonics C_3 and C_5 are significant and harmonics above C_5 can be neglected. The anharmonicity becomes weaker with decreasing D and pure spirals with $C_{2l+1>1} = 0$ are recovered as $D \to 0$.

In the 3SL region, the stable phase below $D_c^{\rm glo}$ has wavevector $Q = (4\pi/3)x$ with the spin configuration shown in the inset to Fig. 2(a). If the spin on site site 1 points up, the spins of neighboring sites 2 and 3 point at angles $\pm\theta$ toward the $-\mathbf{z}$ direction. Since odd multiples of $\mathbf{Q} = (4\pi/3)\mathbf{x}$ are either equivalent to the Bragg vector $4\pi\mathbf{x}$ or to $\mathbf{Q}$ itself, Eqs. (2) and (3) imply that this spin configuration has z component

$$S_z(\mathbf{R}) = \frac{\cos(4\pi x/3) + f}{1 + f}, \quad (4)$$

where f is a constant. Because the angle θ is given by the relation $\cos\theta = (f - 1/2)/(1+f)$, the 3SL phase with $\theta = \pi$ is recovered when $f = -1/4$.

The critical value $D_c^{\rm glo}/|J_1|$ in the 3SL region depends only on $J_3/|J_1|$ and not on $J_2/|J_1| > 0$. We find that the conditions for global and local stability coincide with $D_c^{\rm glo} = D_c^{\rm loc} = 3(|J_1| + |J_3|)/2$. Lines of constant critical anisotropy (iso–anisotropy curves) are sketched in Fig. 1 in increments

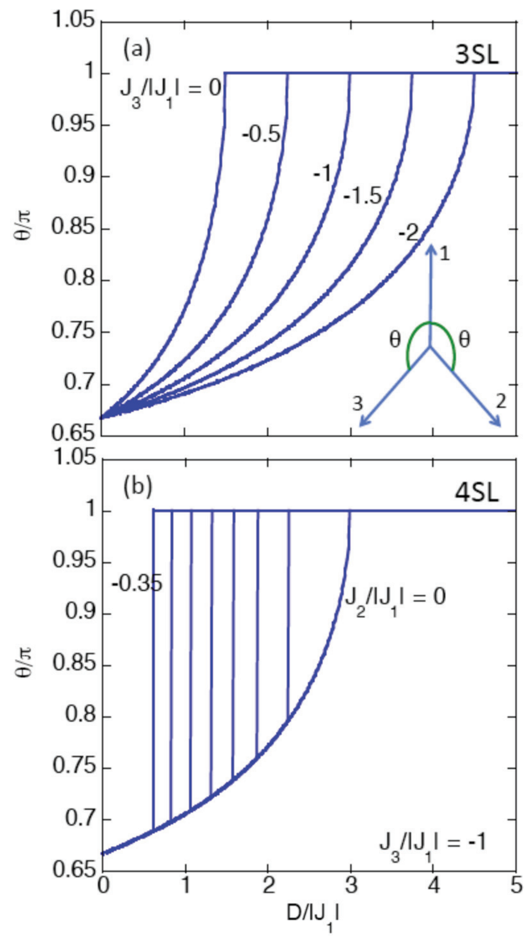

FIG. 2. (Color online) The angle θ for the noncollinear spin state with wavevector $4\pi/3$ versus $D/|J_1|$ (a) in region 3SL for any $J_2/|J_1| > 0$ and (b) in region 4ii with $J_3/|J_1| = -1$.

of 0.2. In Fig. 2(a), θ smoothly decreases from π (the 3SL phase) for $D > D_c^{\rm glo}$ to $2\pi/3$ (the ''120° phase'') as $D \to 0$.

On the left side of the $J_2 = 0$ phase boundary but to the right of the solid curve, the stable phase below $D_c^{\rm glo}$ is precisely the same $\mathbf{Q} = (4\pi/3)\mathbf{x}$ phase described above. For any $J_2 < 0$, the transition between the 4SL phase and the phase below $D_c^{\rm glo}$ is first order, as shown in Fig. 2(b), with $\theta < \pi$ just below $D_c^{\rm glo}$. The critical values for global stability $D_c^{\rm glo}$ always exceed the critical values for local stability $D_c^{\rm loc}$ calculated earlier.[5] Just to the left of the 4SL-3SL phase boundary, $D_c^{\rm glo}$ is precisely three times higher than $D_c^{\rm loc}$ so that the iso–anisotropy curves sketched in Fig. 1 continuously join those on the right side of the $J_2 = 0$ phase boundary. Hence, our results obey the theorem that the iso–anisotropy curves must be continuous across any phase boundary.

More surprisingly, the region of stability for the $4\pi/3$ phase does not extend all the way to the boundary of the 4I subregion evaluated earlier[5] using the conditions for local stability. We have denoted 4ii as the stable subregion for the $4\pi/3$ phase. To the left of the solid curve in subregion 4i, the low-D state is no longer the $4\pi/3$ state but rather is characterized by nonzero coefficients C_3 and C_5 and by a wavevector $\mathbf{Q}$ that depends sensitively on the exchange parameters. As seen in Fig. 1, the iso–anisotropy curves in region 4i continuously join the iso–anisotropy curves in subregion 4ii. So as expected, the critical values for global stability are continuous across the 4i-4ii phase boundary.

In earlier works,[5,6] we speculated that the dominant SW instability wavevector $\mathbf{Q}_{\rm SW}$ of the collinear phase just above $D_c^{\rm loc}$ corresponds to the dominant ordering wavevector $\mathbf{Q}_{\rm NC}$ of the noncollinear phase just below $D_c^{\rm glo}$. This seems to be the case when $D_c^{\rm loc} = D_c^{\rm glo}$ and the phase transition is second order, such as for the 3SL phase with parameters plotted in Fig. 2(a). However, this conjecture is violated, sometimes spectacularly, when $D_c^{\rm glo} > D_c^{\rm loc}$ and the phase transition is first order. For example, between the solid and dashed curves lying within subregion 4i sketched in Fig. 1, $\mathbf{Q}_{\rm SW} = (4\pi/3)\mathbf{x}$ but $\mathbf{Q}_{\rm NC}$ sensitively depends on the exchange parameters. Even within subregion 4i to the left of the dashed curve, where both $\mathbf{Q}_{\rm SW}$ and $\mathbf{Q}_{\rm NC}$ depend on the exchange parameters, $\mathbf{Q}_{\rm SW}$ and $\mathbf{Q}_{\rm NC}$ are not precisely equal.

Of course, it is possible that the trial spin state employed in this work does not provide the lowest-energy solution of Eq. (1). However, the continuous iso–anisotropy curves provide us with great confidence in our variational solutions. Like a jig-saw puzzle, the stable phases of the TLAF can be pieced together one subregion at a time, with the continuity condition providing assurance that the puzzle is being assembled correctly.

For example, the global critical values to the right of the $J_2 = -|J_1|/2$ phase boundary between the 4SL and 8SL phases are now substantially higher than the local critical values. This places severe constraints on the noncollinear spin state in the neighboring 8SL region. Along the $J_3 = J_2/2$ phase boundary between the 4SL and 2SL phases, the global critical values are again larger than the local critical values, except when $D_c^{\rm glo} = D_c^{\rm loc} = 0$ for $J_2 = -|J_1|/3$. So our results for the 4SL phase place severe constraints on the noncollinear spin state of the neighboring 2SL region. We hope that this work will prove useful in obtaining a complete solution of the classical TLAF in the future.

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