Skyrmion crystal formation and temperature–magnetic-field phase diagram of the frustrated triangular lattice Heisenberg magnet with easy-axis magnetic anisotropy

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The nature of the skyrmion-crystal (SkX) formation and various multiple-q phases encompassing the SkX phase are investigated by extensive Monte Carlo simulations on the frustrated J_1 - J_3 triangular-lattice Heisenberg model with the weak easy-axis magnetic anisotropy. Phase diagrams in the temperature T vs magnetic-field H plane are constructed, leading to a rich variety of multiple-q phases. The anisotropy stabilizes the SkX state down to T = 0 at intermediate fields, while in the lower-field range the SkX state becomes only metastable, and new multiple-q states with a broken C_3 symmetry are instead stabilized. Implications to experiments are discussed.

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I. INTRODUCTION

Much attention has recently been paid to various types of topologically protected nanoscale spin textures in magnets, e.g., vortex, skyrmion and hedgehog, from both fundamental interest in topology-related physics and possible applications to spintronics. Skymion, a swirling noncoplanar spin texture characterized by an integer topological charge whose constituent spin directions wrap a sphere in spin space, has gotten special attention. In magnetically ordered states, skyrmions are often stabilized as a periodic array called the skyrmion crystal (SkX). At an earlier stage, the SkX state was discussed for noncentrosymmetric magnets as induced by the antisymmetric Dzyaloshinskii-Moriva (DM) interaction [1–5]. In 2012, it was theoretically proposed that the "symmetric" SkX is also possible in certain class of frustrated centrosymmetric magnets without the DM interaction, where the size of constituent skyrmions can be varied continuously from very small to infinitely large (corresponding to the continuum limit) by tuning the extent of frustration [6]. An interesting characteristic of frustration-induced symmetric skyrmions is that, due to the underlying chiral degeneracy, both skyrmions and antiskyrmions of mutually opposite signs of topological charge, or the scalar chirality, are equally possible, leading to the unique and rich electromagnetic responses [6].

In Ref. [6], the SkX was identified in a simplified model, i.e., the frustrated J_1 - J_3 (J_1 - J_2) isotropic Heisenberg model on the triangular lattice as a triple-q state stabilized by magnetic fields and thermal fluctuations. Subsequent experiments successfully observed the SkX for centrosymmetric triangular-lattice metallic magnets, e.g., Gd₂PdSi₃, accompanied by the pronounced topological Hall effect [7]. Recent Monte Carlo (MC) simulations indicated that the SkX could also be stabilized in the standard RKKY system with only the bilinear interaction modeling weak-coupling metals,

where the oscillating nature of the RKKY interaction bears frustration [8,9].

Of course, real material possesses various perturbative interactions not taken into account in a simplified model [6], e.g., the three-dimensionality (interplane coupling), the magnetic anisotropy, quantum fluctuations, etc. In particular, experiments have indicated that the SkX can be stabilized even at zero temperature (T = 0), where the effect of certain perturbative interactions, e.g., the magnetic anisotropy, was argued to play a role [7,10]. A possible mechanism leading to the T = 0 SkX state was theoretically discussed in the literature, including the biquadratic interaction arising from the higher-order perturbation beyond the second order (strongcoupling effect in itinerant metals) [11–13], quantum spin fluctuations [14], etc. Among them, the magnetic anisotropy prevails in real magnets, both classical and quantum, and generally exists even at the spin-bilinear order.

On the basis of the ground-state phase diagram of the frustrated J_1 - J_2 triangular Heisenberg model obtained by the simulated annealing, it was theoretically suggested that the easy-axis magnetic anisotropy stabilized the SkX state even at T = 0 [10]. While the effect of magnetic anisotropy on the SkX formation was examined further by various authors [14–21], most of them concentrated on the T =0 properties, with few studies on the temperature (T) vs magnetic-field (H) phase diagram (see [15,16,20], however). Even concerning the T = 0 properties, the proposed magnetic-anisotropy stabilization of the SkX state might deserve further careful examination, since the numerical method employed, e.g., the simulated annealing, might capture the metastable SkX state, while such a metastable, not truly stable SkX state was indeed reported under certain annealing conditions even experimentally [22,23].

Under these circumstances, we study by extensive MC simulations the SkX formation and the T-H phase diagram of the frustrated J_1-J_3 Heisenberg model on the triangular lattice with the easy-axis magnetic anisotropy, an anisotropic extension of the isotropic model of Ref. [6]. We wish to clarify how the T-H phase diagram of the isotropic model changes

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by the magnetic anisotropy, paying special attention to the questions of whether the SkX state is truly stabilized at T = 0, whether some new phases appear induced by the anisotropy and, if any, the nature of these phases. We then find that the SkX state is stabilized in intermediate fields at T = 0, while its stability range is considerably reduced compared with that obtained by the simulated annealing, and in the region where the SkX phase turns out to be only metastable, two new anisotropy-induced multiple-q phases with a broken C_3 symmetry emerge as stable phases.

II. MODEL AND METHOD

We consider the J_1 - J_3 classical Heisenberg model on the two-dimensional triangular lattice with the easy-axis uniaxial anisotropy. The Hamiltonian is given by

$$\mathcal{H} = -J_1 \sum_{\langle i,j \rangle_1} (S_{ix} S_{jx} + S_{iy} S_{jy} + \gamma S_{iz} S_{jz}) + -J_3 \sum_{\langle i,j \rangle_3} (S_{ix} S_{jx} + S_{iy} S_{jy} + \gamma S_{iz} S_{jz}) - H \sum_i S_{iz}, \quad (1)$$

where $J_1 > 0$ is the ferromagnetic nearest-neighbor coupling, $J_3 < 0$ the antiferromagnetic third-neighbor coupling, $S_i = (S_{ix}, S_{iy}, S_{iz})$ a three-component unit vector at site *i*, magnetic field is applied along the easy axis with *H* the magnetic-field intensity, and γ the uniaxial exchange anisotropy parameter. We assume a rather weak easy-axis anisotropy and set $\gamma = 1.1$, i.e., 10% anisotropy. Following Ref. [6], we set $J_1/J_3 = -1/3$, and J_1 , *T*, and *H* are given in units of $|J_3|$, hereafter.

MC simulation based on the standard heat-bath method combined with the over-relaxation method is performed. In addition, fully equilibrated temperature-exchange simulations are also made in the higher-*T* range. The lattice is a $L \times L$ triangular lattice with L = 144, 180, 216, 288 with periodic boundary conditions. Unit MC step consists of one heat bath and *L* over-relaxation sweeps. Typically, each run contains 2×10^5 MC steps per spin (MCS) at each temperature, the first half discarded for thermalization.

To reach a given (T^*, H^*) state, together with the field-cooling (FC) run, i.e., the gradual cooling simulatedannealing run at fixed H^* , various other computation protocols are tried by combining H and T sweeps in search for the stable state. Since, at sufficiently low $T = T_0$, a truly stable state should have the lowest energy among several metastable states generated by different protocols, it can be determined by comparing their energies. One standard protocol might be the zero-field cooling (ZFC) run to (T_0, H^*) , i.e., gradual cooling in zero field (or weak fields of $H \lesssim 1.5$) to a low $T = T_0$ (we set here $T_0 = 0.1, 0.05$) followed by the gradual increase of H to H^* at $T = T_0$. Such ZFC runs are repeated $10 \sim 20$ times in search for the lower-energy state by changing the random numbers and the way of H application. If the ZFC protocol yields a stable state with the lowest energy at (T_0, H^*) , a gradual warming run from that state is also performed to higher $T = T^*$ at fixed H^* (sometimes a further cooling run is also made). Consistency is then checked by confirming the obtained state to be compatible with that obtained by the T-exchange simulations at moderately high T.



FIG. 1. The phase diagram of the model with $J_1/J_3 = -1/3$ and $\gamma = 1.1$ in the temperature vs magnetic-field plane. The notations (mq, nq), D and U are explained in the text.

III. RESULTS

The T-H phase diagram obtained in this way is shown in Fig. 1. It contains ten distinct ordered phases. Although it might look rather complicated, all the phases appearing in the isotropic phase diagram [6] also appear here. The triple-qSkX state, described as (3q, 3q) in Fig. 1, is stabilized down to T = 0 in the intermediate field range, where m and n (=1,2,3) in (mq, nq) represent the number of the dominant (quasi-)Bragg peaks except for $\mathbf{q} = 0$ in the transverse (S_{xy}) and longitudinal (S_z) spin structure factors, $S_{\perp}(q)$ and $S_{\parallel}(q)$ [24]. The SkX state is essentially of the same type as the one of the isotropic model, as demonstrated in the real-space spin and scalar-chirality configurations of Figs. 2(c) and 2(d). The SkX state is characterized by the nonzero total scalar chirality $\chi_{tot} > 0$, leading to the topological Hall effect. The T dependence of the specific heat and χ_{tot} at H = 3.5 are shown in Figs. 2(a) and 2(b), the definitions of χ_{tot} and $S(\mathbf{q})$ being



FIG. 2. The temperature and size dependence of (a) the specific heat, and of (b) the total scalar chirality, at H = 3.5. The data of the FC runs and of the temperature-exchange runs (T_{ex}) are shown. The typical real-space configurations of (c) the transverse components of the spin, and of (d) the scalar chirality, in the SkX state at H = 3.5 and T = 0.0001 are shown for a common part of the L = 180 lattice.



FIG. 3. The spin structure factors (a) $S_{\perp}(q)$ and (b) $S_{\parallel}(q)$, and the projected spin (c) (S_x, S_y) and (d) (S_x, S_z) components in the (1q, 2q) phase at H = 3 and T = 0.0001. (e)–(h): The corresponding plots in the (2q, 2q) phase at H = 1.5 and T = 0.0001. The lattice size is L = 180.

given in the Supplemental Material (SM) [25]. In the higher-*T* region, the so-called *Z* phase, the random domain state of the SkX, and the anti-SkX [6], also appears as the collinear triple-q (D, 3q) state [D means disordered, i.e., the absence of sharp (quasi-)Bragg peak in S(q)]; see Fig. S7 [25].

The single-q spiral states also appear both in zero (or sufficiently weak) field and in higher fields. The first type, the (1q, 1q) state, is a vertical spiral (VS) induced by the easy-axis anisotropy, exhibiting a 90° rotation from the conical spiral (CS) stabilized in the isotropic model [6]. In zero field, this VS state in the $T \rightarrow 0$ limit is vertically coplanar, but it becomes weakly noncoplanar in nonzero fields, the latter corresponding to the "M state" of Ref. [10]. By contrast, the high-field single-q state, the (1q, U) state [U means uniform, i.e., only the q = 0 peak in $S_{\parallel}(q)$], is essentially the same CS as that of the isotropic model [6]. In the high-field region, there also appears the double-q state, the (2q, 1q) state, essentially the same as that of the isotropic model [6]. Further details of these states are given in the SM [25].

Between the SkX phase at intermediate *H* and the VS phase at low enough *H*, there appear two new phases absent in the isotropic model, i.e., the (1q, 2q) and the (2q, 2q) phases, which persist even in the $T \rightarrow 0$ limit. The associated $S_{\perp}(q)$ and $S_{\parallel}(q)$ are respectively given in Figs. 3(a), 3(b), 3(e), and 3(f). Note that "1q" ("2q") here means S(q) possesses three (quasi-)Bragg peaks, but the C_3 symmetry is broken resulting in one (two) pair of higher-intensity peaks [24]. In both phases, χ_{tot} vanishes, meaning the absence of the topological Hall effect.

An important caveat might be in order here: If one makes FC simulated-annealing runs in the field range $2.0 \leq H \leq 3.6$ to low *T*, one ends up with the triple-*q* SkX state even if one makes a very slow cooling. By contrast, if one makes a ZFC run to (T_0, H^*) with H^* in the relevant range, one generally ends up with the C_3 -symmetry broken state. The energy (*e*) comparison indicates that, for the field $1.35 \leq H \leq 2.6$ the (2q, 2q) state reached by the ZFC run is stable [*e* is lower than *e* of the SkX state reached by the FC run by ~0.54% and by ~0.27% at H = 2 and 2.5, respectively, well beyond the typical error bar of order 0.001%]; for $2.6 \leq H \leq 3.2$ the (1q, 2q) state is stable [*e* is lower than *e* of the SkX state by ~0.12% at H = 3]; but for $3.2 \leq H \leq 4.1$ the (3q, 3q) SkX state is stable [*e* is lower than *e* of the (2q, 2q)/(1q, 2q) states by ~0.08% and by ~0.12% at H = 3.5 and 4, respectively.



FIG. 4. The temperature dependence of (a) the specific heat for various sizes, and of (b) the intensities of the three (quasi-)Bragg peaks of $S_{\perp}(q)$ and $S_{\parallel}(q)$ for L = 288, at H = 2. The data are taken by the gradual warming run from the $T = T_0$ state as explained in the text, while in (a) the data of the temperature-exchange run (T_{ex}) are also added. The projected vector-chirality (c) (κ_x , κ_y) components in the (1q, 2q) phase at H = 3 and T = 0.0001, and (d) (κ_x , κ_z) components in the (2q, 2q) phase at H = 1.5 and T = 0.0001. The projected (e) spin (S_x , S_z) and (f) vector-chirality (κ_x , κ_z) components in the (2q, 1q) phase at H = 4.5 and T = 0.0001.

tively]. In fact, the energy difference between the (2q, 2q) and (1q, 2q) states is rather small of order of the error bar, although we have observed a clear phase transition between these two states with varying *T*; see Figs. 4(b) and S2(b) [25].

Our observation then indicates that the easy-axis anisotropy stabilizes the triple-q SkX state even at T = 0 at intermediate H (3.2 $\leq H \leq 4.1$), where its stability range is considerably reduced compared with that obtained by the simulated annealing, and the truly stable state in the lower-Hregion turns out to be the (1q, 2q) state for 2.6 $\leq H \leq 3.2$, and the (2q, 2q) states for 1.35 $\leq H \leq 2.6$. To the author's knowledge, these two states, the (1q, 2q) and (2q, 2q) states, are unnoticed so far. The observed strong hysteretic effect might give the reason why these states were not reported in the T = 0 phase diagram constructed by, e.g., the simulated annealing [10].

Let us further look into the nature of these new phases. Although the (1q, 2q) state is a vertical coplanar state as shown in Figs. 3(c) and 3(d) [26], it is not a simple VS (1q, 1q) state. In fact, as can be seen from the vector-chirality (κ_x, κ_y) projection shown in Fig. 4(c), where κ is the vector chirality defined on each upward triangle by $\kappa = (2/3\sqrt{3}) \sum_{\langle ij \rangle} S_i \times S_j$ (the summation taken over three clockwise bonds on each triangle) changing sign under the spatial inversion, κ mostly exhibits parallel alignment in the direction perpendicular to the coplanar spin plane, but some κ exhibits antiparallel alignment in the opposite direction, indicating that the spins rotate mostly in a certain (say, clockwise) direction, but occasionally rotate in an opposite (say, counter-clockwise) direction. Closer inspection reveals that such a counter-rotation occurs when the spins stay in the vicinity of the H direction to gain the Zeeman energy.

By contrast, the (2q, 2q) state is a noncoplanar state as shown in Figs. 3(g) and 3(h). As can be seen from the (κ_x, κ_z) projection of Fig. 4(d), κ is dominated by the horizontal (perpendicular to the field) component, which suggests that the associated noncoplanar spin configuration is basically "vertical." If one compares this with the corresponding plot for another double-q state, i.e., the (2q, 1q) state in high fields, κ in the latter rather lacks in the horizontal component [see Fig. 4(f)], consistently with the meronlike "conical" character of its spin state [see Fig. 4(e)]. Hence, the two double-q states, the (2q, 2q) and (2q, 1q) states, are different kinds of states, i.e., "vertical" vs "conical".

In the higher-*T* range, phases absent in the isotropic model also appear, including the collinear single-*q* (D, 1*q*) and the collinear double-*q* (D, 2*q*) phases. In Fig. 1, the collinear single-*q* phase appears in two distinct regions, i.e., at intermediate fields and at zero and weaker fields, each represented by (D, 1*q*) and (D, 1*q*), which are not connected in the phase diagram. Indeed, in the (D, 1*q*) state, $S_{\parallel}(q)$ possesses three pairs of (quasi-)Bragg peaks, among which single pair $\pm q_1^*$ exceeds the other two $\pm q_2^*$ and $\pm q_3^*$ by factor of $2 \sim 3$ in their intensities (refer to Figs. 4(b), S2(b) and S6 [25]), while, in the (D, 1*q*)' state, $S_{\parallel}(q)$ possesses only one pair of (quasi-)Bragg peaks.

The richness of the phase diagram suggests that even a simple cut of the phase diagram could yield many phases and phase transitions among them. We demonstrate such richness by showing the *T* dependence of physical quantities at a representative field H = 2. The data are taken by the gradual warming runs from the $T = T_0$ state prepared by the ZFC run explained above. On increasing *T* from T_0 , one encounters the (2q, 2q), (1q, 2q), (D, 2q), (D, 1q), and (D, 3q) states before finally reaching the paramagnetic state. We show in Fig. 4 the *T* dependences of (a) the specific heat and of (b) the intensities of the three relevant (quasi-)Bragg peaks q_i^* (i = 1, 2, 3) of the spin structure factor which are ordered according to their intensities [27]. Similar data for H = 2.5 are also given in Fig. S2 of the SM [25]. As can be seen from the figures, the system indeed exhibits a rich phase structure.

In view of the complicated appearance of the phase diagram, we now try to give a rough and intuitive picture of the complicated phase diagram, together with an intuitive reason why the easy-axis anisotropy energetically stabilizes the SkX phase at intermediate fields. Depending on the relative strength of the easy-axis anisotropy and the magnetic field, the phase diagram might be divided into three regimes, i.e., (i) the high-field regime where the field exceeds the anisotropy, (ii) the low-field regime where the anisotropy exceeds the field, and (iii) the medium-filed region where both compete. In the region (i) involving the double-q(2q, 1q) and the CS (1q,U) states, the spin states tend to be "conical" induced by the field, while in the region (ii) involving the VS (1q, 1q), the double-q (2q, 2q), and the single-q (1q, 2q) states, the spin states tend to be "vertical" induced by the anisotropy. Since the conical and vertical states compete with each other, the states in between tend to be virtually "spherical," setting the stage for stabilization of the SkX state.

IV. SUMMARY AND DISCUSSION

In summary, by means of extensive MC simulations on the frustrated J_1 - J_3 triangular-lattice Heisenberg model with the easy-axis exchange anisotropy, we have constructed the T-H phase diagram containing a rich variety of multiple-qphases. The easy-axis anisotropy stabilizes the triple-q SkX state down to T = 0 at intermediate fields. As the field gets weaker, the SkX state becomes only metastable, and new multiple-q states with a broken C_3 symmetry, the (2q, 2q) and (1q, 2q) states, are instead stabilized. In the high-T regime, in addition to the collinear triple-q phase (Z phase), the collinear single-q and double-q states absent in the isotropic model are stabilized by the easy-axis anisotropy.

Finally, we discuss experimental implications of the present result. Concerning the stability of the SkX and the multiple-q states encompassing it, while the weak easy-axis magnetic anisotropy enhances the SkX formation even at T = 0, it often accompanies a strong hysteretic effect associated with the C_3 breaking. Thus, in order to experimentally clarify the SkX-related phase structure, one needs to examine carefully the possible dependence of the state on the T-cooling/H-application protocols. Especially when different final states are to be obtained by different protocols to a common (T, H), one should determine which state is truly stable. Since the direct comparison of the energies as we did in the present analysis would be difficult experimentally, the long-time off-equilibrium measurements toward equilibrium might eventually be required. Experimental distinction among (1q, 2q), (2q, 2q), and (3q, 3q) from S(q) measurements might sometimes be not easy due to the domain problem, whereas the absence of the topological Hall effect in the former two could be used as a signature to distinguish them from the SkX state.

Of course, features of the phase diagram might well depend on the type and the strength of the anisotropy, e.g., the γ value, as well as on other perturbative interactions not taken into account in the present model, e.g., the dipolar interaction, quantum fluctuations, higher-order exchange interactions, etc. For example, since the energy difference between the two new C_3 -broken states, the (2q, 2q) and (1q, 2q) states, is rather small, a small change in γ and/or other perturbative effects might affect their relative stability. While further theoretical and experimental studies are desirable to fully clarify the effects of these perturbative interactions, the present work might hopefully serve as a useful starting reference.

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- [24] For example, if S(q) possesses three (quasi-)Bragg peaks at q_1 , q_2 , and q_3 with the relation $S(q_1) = S(q_2) > S(q_3) > 0$, we call it 2q.
- [25] See Supplemental Material at http://link.aps.org/supplemental/ 10.1103/PhysRevB.110.014424 for (I) definitions of physical quantities, (II) the temperature dependence of physical quantities in magnetic fields, and (III) properties of each ordered phase.
- [26] Note that the real-space (x, y) [or the *q*-space (q_x, q_y)] part and the spin-space (S_x, S_y) part are totally uncoupled in the present model so that any spin configuration generated by arbitrary global spin rotation around the magnetic-field (S_z) axis is equally possible. In Figs. 3(c) and 3(d), just for illustration, we take a special choice that the coplanar spin place coincides with the (S_x, S_z) plane. The same convention is employed also in Fig. 4(c).
- [27] The $(2q, 2q) \leftrightarrow (1q, 2q)$ transition temperature is estimated to be $T_c \simeq 0.23$ from Figs. 4(a) and 4(b). Even within the (2q, 2q)phase, however, some steplike "structure" or "weak anomaly" exists in Fig. 4(b) just below T_c , while the basic (2q, 2q) character of S(q) remains the same. We regard such "structure" only as a secondary one associated with, e.g., the possible incommensurability effect or the domain formation, although we cannot completely rule out the possibility that another phase with the same (2q, 2q) ordering pattern intervenes between the (2q, 2q)and (1q, 2q) phases just below T_c .