Skyrmion Crystals in Frustrated Shastry–Sutherland Magnets

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The phase diagrams of the frustrated classical spin model with the Dzyaloshinskii–Moriya (DM) interaction on the Shastry–Sutherland lattice are studied by means of Monte Carlo simulations. For ferromagnetic next-nearest-neighboring ($J_2$) interactions, the introduced exchange frustration enhances the effect of the DM interaction, which enlarges the magnetic field-range with the skyrmion lattice phase and increases the skyrmion density. For antiferromagnetic $J_2$ interactions, the so-called 2$q$ phase (two-sublattice skyrmion crystal) and the spin-flop phase are observed, and their stabilizations are closely dependent on the DM interaction and $J_2$ interaction, respectively. The simulated results are qualitatively explained from the energy competitions among these couplings, which provide an important guidance for finding skyrmion crystals in frustrated magnets.

During the past few years, skyrmions have drawn extensive attention for their potential applications in the future memory technology because of the outstanding merits including the nanoscale sizes, the topological protection, and the ultralow critical drive currents. Magnetic skyrmions have been discovered experimentally in chiral magnets including MnSi, MnGe and FeGe and have been proven theoretically to be stabilized by the competition among the ferromagnetic (FM) exchange, Dzyaloshinskii–Moriya (DM), and Zeeman couplings in the presence of thermal fluctuations. Otherwise, strong interfacial DM interactions are induced at heavy metal (HM)/FM interfaces in films such as Pt/Co/MgO heterostructure and Pt/Co/Ta thin film due to the broken inversion symmetry and strong spin-orbit coupling, contributing to the stabilization of Neel-type magnetic skyrmions even at room temperature (T). More recently, it has been reported that the exchange bias at the antiferromagnetic (AFM)/FM interface of the IrMn/CoFeB/MgO heterostructure can stabilize the skyrmions even in the absence of an external magnetic field ($B$). More interestingly, the DM interaction at the interface can be significantly enhanced by a factor of seven through increasing the IrMn thickness, which is very meaningful for spintronic applications. In addition, several frustrated magnets such as the triangular antiferromagnets are predicted to host the skyrmion lattice phase. Instead of the DM interaction, the competition between the nearest-neighbor (NN) and next-nearest-neighbor (NNN) interactions is essential for stabilizing the skyrmions. Moreover, the first observation of skyrmionic magnetic bubbles formed at room temperature has been reported experimentally in the frustrated kagome Fe$_3$Sn$_2$ magnet, well confirming the earlier predictions. It is noted that finding materials with a stabilized skyrmion phase in a wide $B$ range is essential both in basic physical research and for potential applications. Frustrated magnets such as the Shastry–Sutherland (S–S) magnets may be promising candidates, as will be explained below.

On the one hand, quite a few rare-earth tetraborides RB$_4$ (R = Tb, Dy, Ho, Tm, etc.) are the famous representatives of the S–S magnets. In these materials, DM interactions could be induced by spin-orbit coupling between rare-earth ions and/or by structural inversion asymmetry through the S–S magnet film design, which may play an important role in stabilizing noncollinear phases. In the S–S Kondo lattice model, for instance, the DM interactions are suggested to be necessary for the emergence of the chiral spin structures. On the other hand, the exchange frustration is proven to be important in understanding the intriguing magnetization behaviors observed in RB$_4$. More importantly, it is expected that the DM effect could be enhanced by the effective exchange frustration and, in turn, further stabilizes the skyrmion phase. As a matter of fact, a similar phenomenon has been observed in the frustrated spin model on the square lattice, and the enhancement of the field-induced skyrmion phase caused by the interaction frustration has been reported.

Thus, there is still an urgent need to elucidate complex magnetic orders in frustrated S–S magnets with DM interactions.

In this work, we study the phase diagrams of the frustrated spin model with a DM interaction on the S–S lattice. For the FM NNN interaction ($J_3$), the introduced exchange frustration enhances the effect of the DM interaction, leading to the enlargement of the magnetic field-range with the skyrmion lattice phase and the increase in the skyrmion density. For the AFM $J_2$ interaction, the 2$q$ phase (two-sublattice skyrmion crystal) and the spin-flop phase are stabilized by the DM interaction and $J_2$.
interaction, respectively. The simulated results are qualitatively explained from the energy landscape.

In the presence of the additional DM interaction, the Hamiltonian is given by

\[ H = -\sum_{i,\delta_n} J_n S_i \cdot S_j - \sum_{\langle i,j \rangle} D_{ij} \cdot (S_i \times S_j) - \sum_i B \cdot S_i^z \]  

(1)

The first term is the exchange interactions where \( j = i + \delta_n \) and \( \delta_n \) connects site i and its n-th nearest neighbor sites with \( n = 1, 2, 3 \). Here, \( J_n \) is the exchange coupling and \( S_i \) represents the Heisenberg spin with unit length on site i. The second term describes that the interfacial DM interaction arises from the structural inversion asymmetry along with the thin-film normal direction. In detail, the DM vectors are set to be \( D_1 = -D_3 = D\hat{y} \) and \( D_4 = -D_2 = D\hat{x} \), as clearly shown in Figure 1. The last term is the Zeeman coupling with \( B \) applied along the z direction.

Our simulation is performed on an \( N = 24 \times 24 \) S–S lattice with period boundary conditions using the standard Metropolis algorithm and the temperature exchange method. Here, the temperature exchange method is used to prevent the system from trapping in metastable free-energy minima caused by the frustration and to relax the system to the equilibrium state. The finite-size effect on the phase transitions is confirmed to be negligible. We took an exchange sampling after every ten standard Monte Carlo (MC) steps. Generally, the initial 5 \( \times 10^3 \) MC steps are discarded for equilibrium consideration and another 5 \( \times 10^3 \) MC steps are retained for statistical averaging of the simulation. To explore the phases in the system, in addition to the well-known magnetization \( M \) and magnetic susceptibility \( \chi \), we also characterized the spin structures by performing the Fourier transform \( A_k = 1/\sqrt{N_S} \sum_i S_i \exp(-i \mathbf{k} \cdot \mathbf{r}_i) \) with \( \mathbf{r}_i \) as the spatial coordinate of site i and then calculating the intensity profile \( |A_k|^2 \). In addition, we also calculated the topological winding number \( Q_{\text{sk}} \) defined by

\[ Q_{\text{sk}} = \frac{1}{4\pi} \int \int S_i \cdot (\partial_x S_i \times \partial_y S_i) \text{d}x \text{d}y \]  

(2)

to characterize the number of skyrmions.

First, the effect of the exchange frustration on the stabilization of skyrmions at a low temperature \( T = 0.01 \) is investigated. Without the loss of generality, we fixed FM \( J_2 = 0.5 \) and \( D = 0.73 \) consistent with the earlier work and modulated the frustration by introducing AFM couplings \( J_1 = 2J_3 = -\alpha \). Here, the ratio of \( D \) to \( J_2 \) is chosen to be the same as in the square lattice system to help one understand the frustration effect more clearly, whereas other values hardly change our main conclusions. Figure 2a, b shows the magnetization \( M(B) \) curves and susceptibility \( \chi(B) \) curves, respectively, for various \( \alpha \). In the absence of the exchange frustration at \( \alpha = 0 \), two discontinuities are clearly observed in the simulated \( M(B) \) curve, indicating two subsequent phase transitions with the increase in \( B \). It is noted that the ground state under zero \( B \) for \( \alpha = 0 \) is the spiral phase with the wave vector \( \mathbf{k} = \arctan(D/\sqrt{2}J_2) \) (1, 1). When \( B \) increases to a critical value, the phase transition from the spiral phase to the skyrmion lattice phase occurs. Subsequently, the FM order is stabilized under strong enough \( B \).

The two critical fields can be estimated from the positions of the two peaks in the simulated \( \chi(B) \) curve. With the increase in \( \alpha \), the second peak significantly shifts toward the high \( B \) side, whereas the first peak is almost invariant, clearly demonstrating the enlargement of the \( B \)-range with the skyrmion lattice phase. This phenomenon can be understood from the following two aspects. On the one hand, when AFM \( J_1 \) and \( J_3 \) couplings are considered, the energy loss from the \( J_1 \) coupling energy \( H_1 \) and the \( J_3 \) coupling energy \( H_3 \) due to the phase transition from the skyrmion lattice phase to the FM phase is introduced, resulting in the increase in the saturation field. Thus, rather than the FM phase, the skyrmion lattice phase can be further stabilized when \( \alpha \) is increased, as clearly shown in our simulations. On the other hand, \( H_1 \) and \( H_3 \) are almost invariant in the phase transition from the spiral phase to the skyrmion lattice phase, and the first critical field is rather stable as \( \alpha \) increases. As a result, the \( B \)-range with the skyrmion lattice phase is obviously enlarged, which is also confirmed in the simulated \( Q_{\text{sk}}(B) \) curves for various \( \alpha \) presented in Figure 2c. Moreover, the DM interaction aligns noncollinear spin textures with a selective direction, and the sign of \( Q_{\text{sk}} \) depends on the sign of \( D \), i.e., a positive \( D \) leads to a negative \( Q_{\text{sk}} \).

Figure 2d shows the simulated phase diagram in the \((\alpha, B)\) parameter plane as well as the density of winding numbers. For a fixed \( B \), the density of \( Q_{\text{sk}} \) increases with the increasing \( \alpha \), indicating an increase/decrease in the density/size of the skyrmion, as clearly shown in Figure 3a, b, which gives the spin configuration and the Bragg intensity \( |A_k|^2 \) under \( \alpha = 0.55 \) for \( \alpha = 0.55 \) and \( 0.4 \), respectively. It is noted that for \( \alpha = 0 \), the size of the skyrmion is mainly determined by the \( J_2/D \) value, i.e., a larger \( J_2/D \) results in a smaller size under a fixed \( B \). The consideration of the competing \( J_1 \) and \( J_3 \) interactions effectively reduces the neighboring spin interactions and enhances the importance of the DM interaction, resulting in the decrease in the size and the increase in the density of the skyrmion. Thus, our work clearly demonstrates that both the \( B \)-range and the skyrmion size can be effectively modulated by tuning the magnitude of the frustration in frustrated S–S magnets.

Most recently, the \( 2q \) phase is observed in the square-lattice antiferromagnets, which shows strong similarity to skyrmions in FM films. In this part, we investigated the field-induced spin

[Image: Figure 1. Effective model on the S–S lattice model with the exchange and additional DM interactions. The DM vectors \( D_1 = -D_3 = D\hat{y}, D_4 = -D_2 = D\hat{x} \) are depicted by the pink arrows.]

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orders for AFM $J_2$ and paid particular attention to the effect of $J_2$ on the low $T$ phase diagram. The simulated phase diagram in the $(J_2, B)$ parameter plane at $T = 0.01$ for FM $J_1 = 2J_3 = 0.5$ is shown in Figure 4, in which the critical fields are similarly estimated from the positions of the peaks in $\chi(B)$ curves. Four phases including the AFM-spiral phase, the $2q$ phase, the spin-flop phase, and the FM phase exist in the phase diagram, and detailed spin configurations and results analysis are given subsequently.

It is noted that spins on the square/S–S lattice tend to form a two sublattice structure. Under low $B$, the AFM-spiral phase is stabilized, in which the staggered magnetization forms the spiral order, as clearly shown in Figure 5a, which shows a snapshot (top) and spin configurations on the sublattices A and B (bottom) of the AFM-spiral phase for $J_2 = 0.4$ under $B = 0.4$. The two interpenetrating spirals on sublattices A and B are with a same ordering wave vector, as clearly demonstrated in the intensity of

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**Figure 2.** The calculated a) magnetization $M$, b) magnetic susceptibility $\chi$, and c) the topological winding number $Q_{sk}$ as functions of $B$ for various $\alpha$ at $T = 0.01$ and $J_2 = 0.5$. d) The summarized phase diagram and the value of $Q_{sk}/N$ in the $(\alpha, B)$ parameter plane.

**Figure 3.** Typical snapshot of the spin configurations of the skyrmion lattice phase for $J_2 = 0.5$ at $T = 0.01$ for a) $(\alpha, B) = (0.0, 0.55)$ and b) $(0.4, 0.55)$. The insets show the Bragg intensity $|A_k|^2$.
the spin structure factor (inset in Figure 5a). When $B$ increases to a critical value, the phase transition from the AFM-spiral phase to the 2$q$ phase occurs. In the 2$q$ phase of which typical configuration under $B = 1.8$ is given in Figure 5b, the sublattice spin configurations show similar characteristics as FM spins in the skyrmion lattice phase, except that the skyrmion density maps are checker-board-like rather than hexagonal. Moreover, the critical field increases as the magnitude of $J_2$ increases. With the further increase in $B$, the 2$q$ phase is directly replaced by the FM phase for $J_2 < 0.15$ to save the Zeeman energy $H_{\text{zee}}$. For a large $J_2$ ($J_2 > 0.15$), the spin-flop phase sandwiched between the 2$q$ phase and the FM phase is stabilized. All the spins in the spin-flop phase have a same nonzero $z$ component, and their $xy$ components form the AFM structure, as clearly shown in Figure 5c, which gives the spin configuration of the spin-flop phase under $B = 2.2$. In addition, the critical $B$ differentiating the spin-flop and FM phase (black triangular points in the phase diagram) significantly increases with the increase in $J_2$, resulting in the enlargement of the $B$-range with the spin-flop phase. As a matter of fact, the critical $B$ at zero temperature can be exactly obtained by the mean-field theory, as explained in detail in Supporting Information S1. Furthermore, the transition from the spin-flop to FM phase is suggested to be a second-order phase transition based on the theoretical calculations.

The simulated phase diagram can be well understood from the energy competitions among these couplings. Figure 6a gives the calculated $B$-dependence of $H_1$, the $J_2$ coupling energy $H_2$, $H_3$, the DM interaction energy $H_{\text{DM}}$, and $H_{\text{zee}}$ for $J_2 = 0.25$, and the corresponding magnetization and susceptibility curves are also shown in Figure 6b. Under small $B$ ($B < 1.2$), the AFM-spiral phase is stabilized by the cooperation of the exchange and DM interactions. Within certain $B$ range ($1.2 < B < 1.5$), the energy loss from $H_2$ and $H_{\text{DM}}$ due to the transition from the AFM-spiral phase to the 2$q$ phase is overtaken by the energy gain mainly from $H_{\text{zee}}$, resulting in the stabilization of the 2$q$ phase. With the further increase in $B$, the sizes of the sublattice skyrmions are decreased to increase the magnetization and to save $H_{\text{zee}}$. Moreover, when $B$ increases to $B > 1.5$, the transition from the 2$q$ phase to the spin-flop phase occurs to save $H_1$, $H_3$, and $H_{\text{zee}}$ in the expense of $H_2$ and $H_{\text{DM}}$. The $z$ component of the spins of the spin-flop phase linearly increases with $B$ until the emergence of the FM phase, as clearly shown in the simulated magnetization curve in Figure 6b.
At last, we tend to discuss the transitions of the critical fields with the enhancement of the $J_2$ interaction. It is noted that $H_2$ increases in the successive two or three phase transitions, as shown in Figure 6c, which gives the simulated local energies for $J_2 = 0.4$. Thus, the critical fields increase when $J_2$ increases, as shown in Figure 6d, which gives the magnetization and susceptibility as a function of $B$ for $J_2 = 0.4$. Furthermore, for a fixed $J_2$, the energy loss from $H_2$ due to the transition from the initial spin-flop phase to the FM phase is rather larger than the energy loss due to the transition from the initial $2q$ phase to the spin-flop phase. As a result, the critical $B$ differentiating the spin-flop and FM phase increases more quickly than the second critical one with the increasing $J_2$, resulting in an enlargement of the $B$-range with the spin-flop phase, as shown in the simulated phase diagram.

Importantly, it is clearly shown that $H_{DM}$ significantly increases with $B$ in the $2q$ phase, whereas it is invariant in the spin-flop phase. Therefore, the $2q$ phase can be enhanced by the DM interaction, resulting in an enlargement of the $B$-range with the $2q$ phase when $D$ is increased. This phenomenon has also been confirmed in our simulations, and the corresponding results are shown in Figure 7, which gives the simulated phase diagram in the $(D, B)$ parameter plane for $J_2 = -0.4$. It is clearly shown that the third transition $B$ is invariant, and the spin-flop phase is gradually replaced by the $2q$ phase with the increase in $D$.

In summary, we have studied the phase diagrams of the frustrated spin model with the DM interaction on the S–S lattice using MC simulation. For the FM next-nearest-neighboring $J_2$ interaction, the frustration introduced by considering the competing exchange interactions enhances the effect of the DM interaction, resulting in not only the enlargement of the field-range with the skyrmion lattice phase but also the increase in the skyrmion density. For the AFM $J_2$ interaction, the so-called $2q$ phase and the spin-flop phase are observed in the simulated phase diagram, and the magnetic field range with the $2q$/spin-flop phase is enlarged with the increase in the $J_2/D$. The simulated results are discussed in detail from the energy competitions among these couplings.

So far, it is suggested that the field-induced noncollinear phases including the skyrmion phase could exist in frustrated S–S magnetic films in which the interfacial DM interaction arises from the structural inversion asymmetry. The magnetization plateau phases in rare-earth tetraborides that probably result

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**Figure 6.** The local energies as functions of $B$ at $T = 0.01$ for a) $J_2 = 0.25$ and c) $J_2 = 0.4$. b,d) The corresponding magnetization and magnetic susceptibility curves.

**Figure 7.** The simulated phase diagram in the $(D, B)$ plane at $T = 0.01$ for FM $J_1 = 2J_3 = 0.5$ and $J_2 = -0.4$. 
from the collinear spin textures are not stabilized due to the DM interaction. More importantly, the stability of the skyrmion phase can be enhanced by combining the DM interaction and frustration. Thus, our work does provide a new guidance for finding skyrmion crystals in frustrated magnets. The uniaxial anisotropy has been confirmed to play an important role in understanding the magnetization behaviors in tetraboride TmB$_4$. The effect of the anisotropy on the phase diagram has been investigated in our work, and the corresponding results are shown in Supporting Information S2. With the enhancement of the anisotropy, the skyrmions phase or $2q$ phase is gradually replaced. Thus, the predictions remain to be checked by further experiments on particular rare-earth tetraboride film designing.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

Dzyaloshinskii–Moriya interaction, frustration, Shastry–Sutherland model, skyrmions

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