



## Article

# Skyrmion Crystal in Bilinear–Biquadratic–Bicubic Model on a Centrosymmetric Triangular Lattice

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## Abstract

We numerically investigate the effect of multi-spin interactions on the stability of skyrmion crystals and other multiple- $Q$  magnetic states, with a particular emphasis on the momentum-resolved bicubic interaction. By performing simulated annealing for an effective spin model that incorporates bilinear, biquadratic, and bicubic interactions on a two-dimensional triangular lattice, we construct the corresponding low-temperature phase diagram. Our results reveal that a positive bicubic interaction stabilizes a skyrmion crystal with a skyrmion number of two, whereas a negative bicubic interaction favors a single- $Q$  spiral state. Moreover, we demonstrate that the stability region of the field-induced skyrmion crystal with the skyrmion number of one is largely enlarged in the presence of a positive bicubic interaction.

**Keywords:** multi-spin interaction; skyrmion crystal; bicubic interaction; topological magnetism; centrosymmetric magnets; phase diagram

## 1. Introduction

Magnetism in solids emerges from the intricate interplay among various electronic degrees of freedom, such as spin, charge, and orbital, as well as the underlying crystal structure. Among the different classes of magnetic materials, itinerant magnets in which magnetic ordering originates from the itinerancy of electrons exhibit a rich variety of magnetic phases that are highly sensitive to the details of electronic band structures and Fermi surface topology [1–3]. In contrast to localized spin systems that involve only the spin degree of freedom, itinerant magnets are governed by both direct and indirect exchange interactions, the latter of which arise from kinetic-driven effects, such as Fermi surface nesting. These competing interactions often lead to multiple complex magnetic phases, including spin density waves, helical spiral states, and multiple- $Q$  states [4–20].

A notable feature of itinerant magnets is the presence of intrinsic multi-spin interactions that arise naturally from the itinerant nature of the electrons. These higher-order spin couplings, such as four-spin interactions and six-spin chiral interactions, often emerge from the integration of the charge degree of freedom in itinerant magnets [21–24]. As a consequence, even minimal models of itinerant magnets can host complex magnetic textures stabilized by effective multi-spin interactions that go beyond conventional Heisenberg-type bilinear exchange interactions.

Recent advances in spintronics and topological materials have led to a surge of interest in topological magnetism, where magnetic textures give rise to nontrivial topological properties [25–27]. In particular, noncoplanar spin textures, such as skyrmions and hedgehogs [28–31], can generate emergent magnetic fields that act on itinerant electrons, resulting



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in phenomena like the topological Hall effect [32–43]. These effects originate from the scalar spin chirality, a triple product of neighboring spins, which serves as a key microscopic degree of freedom in such noncoplanar magnetic systems.

Itinerant magnets provide particularly fertile ground for realizing topological magnetic phases due to the strong coupling between spin and charge degrees of freedom. Indeed, several topological magnetic phases have been discovered in itinerant magnets, such as  $\text{MnSc}_2\text{S}_4$  [44,45],  $\text{Gd}_2\text{PdSi}_3$  [46–50],  $\text{Gd}_3\text{Ru}_4\text{Al}_{12}$  [51–54],  $\text{GdRu}_2\text{Si}_2$  [55–60], and  $\text{EuPtSi}$  [61–68] that host the SkX and  $\text{MnSi}_{1-x}\text{Ge}_x$  [69–72] and  $\text{SrFeO}_3$  [73–76] that host the hedgehog crystal. Especially, the importance of the Fermi surface nesting has been demonstrated in  $\text{GdRu}_2\text{Si}_2$  by angle-resolved photoemission spectroscopy measurements [77]. From a theoretical perspective, it remains important to clarify the stabilization mechanisms of such topological magnetic states in itinerant magnets based on the fundamental model, although most previous studies have focused on multi-spin interactions up to the four-spin level.

In the present study, we focus on the role of bicubic interactions on the stability of topological magnetic states [78,79]. Our analysis is conducted on the basis of the effective bilinear–biquadratic spin model developed in earlier works, where the SkX is stabilized depending on the strength of the biquadratic interaction [23,80]. Meanwhile, the effect of higher-order spin interactions on the stabilization of the SkX has not been fully elucidated, although itinerant magnets inherently possess higher-order spin interactions owing to the interplay between spin and charge degrees of freedom. To clarify this point, we examine the low-temperature phase diagram when both biquadratic and bicubic interactions are varied on a two-dimensional triangular lattice by employing simulated annealing, obtaining the stability tendency of the SkX with the skyrmion number ( $n_{\text{sk}}$ ) of two, denoted as  $n_{\text{sk}} = 2$  SkX hereafter, against the bicubic interaction, which is stabilized in the bilinear–biquadratic model at zero external magnetic field [23]; the positive (negative) bicubic interaction enhances (suppresses) the stability region of the  $n_{\text{sk}} = 2$  SkX. Furthermore, we show that this stability tendency also holds for the field-induced SkX with the skyrmion number of one ( $n_{\text{sk}} = 1$  SkX). Our results show that multi-spin interactions that sometimes cannot be neglected in itinerant magnets affect the stability region of topological magnetic phases.

The rest of this paper is organized as follows. In Section 2, we present an effective spin model, which is derived from the itinerant electron model with spin–charge coupling in the weak-coupling regime. We also describe the numerical method based on simulated annealing, which is used to investigate the low-temperature phase diagram of the effective spin model. In Section 3, we show the stability of SkX and other multiple- $Q$  states in the presence of positive and negative bicubic interactions. We discuss the trend of the phase diagram in the presence and absence of an external magnetic field. Finally, we conclude the present results in Section 4.

## 2. Model and Methods

We begin with the Kondo lattice model, a prototypical framework for describing itinerant magnetism [81–83]. The model Hamiltonian incorporates both itinerant electrons and localized spins, which is expressed as

$$\mathcal{H}_{\text{KLM}} = - \sum_{i,j,\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + J_{\text{K}} \sum_{i,\sigma,\sigma'} c_{i\sigma}^\dagger \sigma_{\sigma\sigma'} c_{i\sigma'} \cdot \mathbf{S}_i, \quad (1)$$

where the itinerant electron degree of freedom is described by creation and annihilation operators,  $c_{i\sigma}^\dagger$  and  $c_{i\sigma}$ , at site  $i$  and spin  $\sigma$ , and localized spin degree of freedom is described by classical spin vectors,  $\mathbf{S}_i$ , at site  $i$ ; the magnitude of  $\mathbf{S}_i$  is normalized such that  $|\mathbf{S}_i| = 1$ .

The first term describes the kinetic energy of itinerant electrons with the hopping amplitude  $t_{ij}$ . The second term represents the on-site exchange coupling between itinerant electron spins and localized spins  $S_i$ , characterized by the coupling constant  $J_K$ . Here,  $\sigma = (\sigma^x, \sigma^y, \sigma^z)$  denotes the vector of Pauli matrices. The competition between the itinerant electron motion (first term) and the local exchange interaction (second term) gives rise to effective interactions between localized spins in an indirect way; we ignore any direct-type exchange interactions for simplicity.

By applying the Fourier transformation to the Kondo lattice model in Equation (1), its momentum representation is given by

$$\mathcal{H}_{\text{KLM}} = \sum_{k\sigma} (\varepsilon_k - \mu) c_{k\sigma}^\dagger c_{k\sigma} + \frac{J_K}{\sqrt{N}} \sum_{kq\sigma\sigma'} c_{k\sigma}^\dagger \sigma_{\sigma\sigma'} c_{k+q\sigma'} \cdot S_q, \quad (2)$$

where  $c_{k\sigma}^\dagger$  and  $c_{k\sigma}$  at wave vector  $k$  and spin  $\sigma$  are the counterparts of  $c_{i\sigma}^\dagger$  and  $c_{i\sigma}$  in real space, respectively, and  $S_q$  is the Fourier transform of a localized spin  $S_i$ . In the first term,  $\varepsilon_k$  is the energy dispersion and  $\mu$  is the chemical potential. For example,  $\varepsilon_k = -2t[\cos k_x + 2 \cos(k_x/2) \cos(\sqrt{3}k_y/2)]$  for the nearest-neighbor hopping on the triangular lattice, where we set the lattice constant to unity. In the second term,  $N$  stands for the number of sites.

In the weak-coupling regime, where the exchange coupling  $J_K$  is much smaller than the bandwidth of itinerant electrons, the model in Equation (2) can be effectively reduced to a spin model by tracing out the itinerant electron degree of freedom [21–23,84]. Assuming that the dominant contributions to the  $n$ th-order spin interactions originate from the momentum channel of the form  $(S_q \cdot S_{-q})^{n/2}$ , the effective spin Hamiltonian up to sixth-order interactions can be written as [23]

$$\mathcal{H} = - \sum_q J_q S_q \cdot S_{-q} + \frac{1}{N} \sum_q K_q (S_q \cdot S_{-q})^2 + \frac{1}{N^2} \sum_q L_q (S_q \cdot S_{-q})^3, \quad (3)$$

where the coupling constants for the bilinear ( $J_q$ ), biquadratic ( $K_q$ ), and bicubic terms ( $L_q$ ) are given by

$$J_q = \frac{2J^2}{N} T \sum_{k,q,\omega_p} G_k G_{k+q}, \quad (4)$$

$$K_q = \frac{2J^4}{N} T \sum_{k,q,\omega_p} G_k^2 G_{k+q}^2, \quad (5)$$

$$L_q = \frac{8J^6}{3N} T \sum_{k,q,\omega_p} G_k^3 G_{k+q}^3. \quad (6)$$

Here,  $G_k(i\omega_p) = [i\omega_p - (\varepsilon_k - \mu)]^{-1}$  is the noninteracting Green's function, and  $\omega_p$  denotes the Matsubara frequency. The first, second, and third terms in Equation (3) stand for the bilinear, biquadratic, and bicubic interactions, respectively, which are derived from the second-, fourth-, and sixth-order perturbations. Among them, the second-order bilinear interaction corresponds to the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction [85–87], whereas the fourth-order biquadratic and sixth-order bicubic interactions provide the correction of the RKKY interaction. It is noted that all the spin interactions appearing in Equation (3) are isotropic in spin space, reflecting the absence of the spin–orbit coupling. Consequently, the model does not include chiral spin interactions, such as the Dzyaloshinskii–Moriya interaction [88,89], chiral biquadratic interaction [90–95], and other types of anisotropic magnetic interactions [96–101].

We investigate the ground-state spin configurations of the model in Equation (3) by focusing on sufficiently low temperatures close to the ground state so that the entropic effect can be neglected. In such a situation, it is enough to consider the spin interactions at particular wave vectors that yield the maximum of  $J_q$  since the perturbative hierarchy  $J_q > K_q > L_q$  implies that the bilinear term dominates. Specifically, we assume that the leading magnetic instability occurs at the wave vector  $Q_1 = (\pi/3, 0)$ , motivated by a presumed nesting feature of the Fermi surface at  $Q_1$ . From the threefold rotational symmetry of the triangular lattice structure, the interactions at  $Q_2 = (-\pi/6, \sqrt{3}\pi/6)$  and  $Q_3 = (-\pi/6, -\sqrt{3}\pi/6)$  are equivalent to that at  $Q_1$ . Under this assumption, the effective spin model in Equation (3) can be further simplified as

$$\mathcal{H}^{\text{eff}} = -2J \sum_{\nu} S_{Q_{\nu}} \cdot S_{-Q_{\nu}} + 2\frac{K}{N} \sum_{\nu} (S_{Q_{\nu}} \cdot S_{-Q_{\nu}})^2 + 2\frac{L}{N^2} \sum_{\nu} (S_{Q_{\nu}} \cdot S_{-Q_{\nu}})^3, \quad (7)$$

where  $\nu = 1-3$ ,  $J \equiv J_{Q_1} = J_{Q_2} = J_{Q_3}$ ,  $K \equiv K_{Q_1} = K_{Q_2} = K_{Q_3}$ , and  $L \equiv L_{Q_1} = L_{Q_2} = L_{Q_3}$ . The prefactor of 2 accounts for the contributions from  $-Q_1$ ,  $-Q_2$ , and  $-Q_3$ . This momentum-resolved effective spin model provides a minimal framework to efficiently search for the instabilities toward multiple- $Q$  magnetic states [102]. In the following analysis, we compute the low-temperature magnetic phases by varying the parameters  $K$  and  $L$  while fixing  $J = 1$  as the unit of energy. Although these parameters originate from a perturbative expansion, we treat both  $K$  and  $L$  as phenomenological parameters, allowing for regimes where  $|J| > |K|, |L|$  even beyond the strict perturbative limit [103]. In particular, we focus on the case of positive  $K$ , which is known to promote multiple- $Q$  instabilities [23], including the SkX observed in  $\text{GdRu}_2\text{Si}_2$  [56,104] and the vortex crystal observed in  $\text{Y}_3\text{Co}_8\text{Sn}_4$  [105].

In addition to the exchange interactions, we incorporate the effect of an external magnetic field along the out-of-plane  $z$  direction. This is introduced via the Zeeman coupling term:

$$\mathcal{H}^Z = -H \sum_i S_i^z, \quad (8)$$

where  $H$  stands for the magnitude of the magnetic field. Accordingly, the total Hamiltonian is expressed as

$$\mathcal{H} = \mathcal{H}^{\text{eff}} + \mathcal{H}^Z. \quad (9)$$

To explore the low-temperature magnetic properties of the model in Equation (9), we consider two situations: one is the absence of the magnetic field ( $H = 0$ ), and the other is the presence of the magnetic field ( $H \neq 0$ ). In the zero-field case, we construct the phase diagram by varying the biquadratic interaction  $K$  and the bicubic interaction  $L$ . This allows us to examine the instabilities toward zero-field topologically nontrivial magnetic states driven by multi-spin interactions. In the finite-field case, we search for the emergence of field-induced topologically nontrivial magnetic states by the interplay between multi-spin interactions and Zeeman coupling. The phase diagrams are determined via simulated annealing using the Metropolis algorithm with local spin updates. Calculations are carried out on the two-dimensional triangular lattice of size  $N = 24^2$  under periodic boundary conditions. Each simulation begins with a randomly initialized spin configuration and proceeds via gradual temperature reduction from an initial temperature  $T_0$ , which is typically taken as 1–5 so as to be comparable to or higher than  $J$ , to a final temperature  $T = 0.0001$ , which is low enough compared to  $J$  to approximate the ground state. The temperature is decreased stepwise according to the annealing schedule  $T_{n+1} = 0.999999T_n$ , where  $T_n$  denotes the temperature at the  $n$ th step; for the annealing schedule rate, it

is desirable to choose a value as close to 1 as possible in order to avoid being trapped in metastable states. At each temperature step, spin configurations are updated locally in real space. Once the system reaches the final temperature, we perform additional Monte Carlo sweeps by  $10^5$ – $10^6$  steps to refine the spin configuration. This procedure is independently repeated for each combination of model parameters  $(K, L, H)$ . To further ensure accuracy near phase boundaries and avoid convergence to metastable states, we also carry out simulations initialized from previously obtained low-temperature spin configurations. This approach enables a more reliable identification of thermodynamically stable magnetic phases.

To characterize the magnetic phases obtained through simulated annealing, we evaluate both spin-related and topologically related quantities. In particular, we analyze the Fourier components of the spin structure to extract ordering tendencies at specific wave vectors  $Q_\nu$ . The magnetic order parameter is defined as the  $Q_\nu$  component of the spin moment, calculated as

$$m_{Q_\nu}^\eta = \sqrt{\frac{S_s^\eta(Q_\nu)}{N}}, \quad (10)$$

$$S_s^\eta(\mathbf{q}) = \frac{1}{N} \sum_{i,j} S_i^\eta S_j^\eta e^{i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)}, \quad (11)$$

where  $S_s^\eta(\mathbf{q})$  stands for the  $\eta = x, y, z$  component of the spin structure factor;  $\mathbf{r}_i$  denotes the real-space position vector at site  $i$ , and  $\mathbf{q}$  is the wave vector in the Brillouin zone. In the absence of the magnetic field, we further define the isotropic contribution as  $(m_{Q_\nu})^2 = (m_{Q_\nu}^x)^2 + (m_{Q_\nu}^y)^2 + (m_{Q_\nu}^z)^2$  and  $S_s(\mathbf{q}) = S_s^x(\mathbf{q}) + S_s^y(\mathbf{q}) + S_s^z(\mathbf{q})$ . In the presence of the magnetic field applied along the  $z$  direction, spin rotational symmetry is preserved in the  $xy$  plane. Accordingly, we consider the in-plane component as  $(m_{Q_\nu}^{xy})^2 = (m_{Q_\nu}^x)^2 + (m_{Q_\nu}^y)^2$  and  $S_s^{xy}(\mathbf{q}) = S_s^x(\mathbf{q}) + S_s^y(\mathbf{q})$ . The uniform (ferromagnetic) component of the magnetization is computed as the site-averaged spin:

$$M^\eta = \frac{1}{N} \sum_i S_i^\eta. \quad (12)$$

To probe the topological nature of the spin textures, we evaluate the scalar spin chirality, a key quantity that captures the degree of noncoplanarity in spin configurations. It is defined by averaging the local chirality over all elementary triangles in the two-dimensional lattice:

$$\chi_{\text{sc}} = \frac{1}{N} \sum_{\mu} \sum_{\mathbf{R} \in \mu} \mathbf{S}_i \cdot (\mathbf{S}_j \times \mathbf{S}_k), \quad (13)$$

where  $\mathbf{R}$  denotes the position of the triangle center, and  $\mu = (u, d)$  indices upward and downward triangles on the triangular lattice. The indices  $(i, j, k)$  refer to the three vertices of a given triangle at position  $\mathbf{R}$ , ordered counterclockwise. A nonzero value of  $\chi_{\text{sc}}$  signals a topologically nontrivial magnetic configuration, characterized by noncoplanar spin textures. Such states give rise to an emergent magnetic field for itinerant electrons via the spin Berry phase mechanism, leading to observable consequences, such as the topological Hall effect [32–43]. The scalar spin chirality is closely related to the skyrmion number  $n_{\text{sk}}$ , which quantifies the topological winding of the spin texture. Its formulation in discrete lattice systems can be found in Ref. [106].

### 3. Results

We discuss the instabilities toward topologically nontrivial magnetic states based on the results obtained from simulated annealing. We present the results in the absence of the magnetic field in Section 3.1 and show the effect of the magnetic field in Section 3.2.

#### 3.1. Zero Magnetic Field

Let us consider the case without the magnetic field, i.e.,  $H = 0$ . Figure 1 presents the low-temperature phase diagram on the plane of the biquadratic interaction  $K$  and the bicubic interaction  $L$ , which is obtained by performing simulated annealing. Three distinct phases appear in the phase diagram, which are denoted as the single- $Q$  spiral (1 $Q$ ) state, double- $Q$  chiral stripe (2 $Q$  CS) state, and  $n_{\text{sk}} = 2$  SkX (SkX II). In the case of  $L = 0$ , where the model reduces to the bilinear–biquadratic form, the single- $Q$  spiral state appears for  $K = 0$ , the double- $Q$  chiral stripe state appears for  $0 < K \leq 0.19$ , and the  $n_{\text{sk}} = 2$  SkX appears for  $K \geq 0.19$ , which is consistent with previous studies [80]. Below, we briefly describe the spin and scalar spin chirality properties associated with each of these three phases.

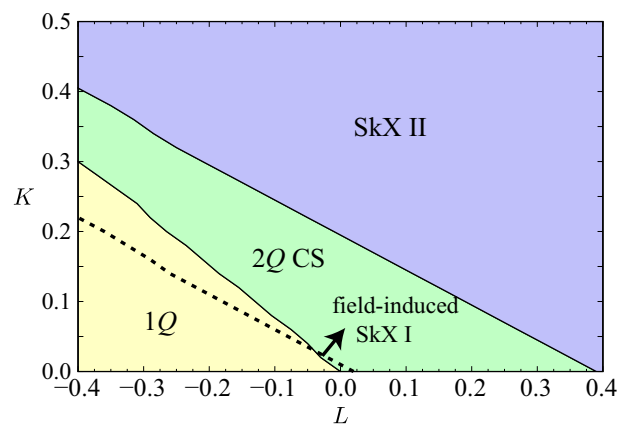


**Figure 1.** Ground-state phase diagram in the plane of the bicubic interaction  $L$  and the biquadratic interaction  $K$ . The phase diagram consists of 1 $Q$ , 2 $Q$  CS, and SkX II, which denote the single- $Q$  spiral state, the double- $Q$  chiral stripe state, and the skyrmion crystal with the skyrmion number of two, respectively. The dashed line stands for the boundary separating the region where the skyrmion crystal with the skyrmion number of one appears or not in an external magnetic field.

The single- $Q$  spiral state, stabilized at  $K = L = 0$ , exhibits a coplanar spiral spin configuration, as illustrated in Figure 2a; the spin configuration can be described by  $S_i = (\cos Q_1 \cdot r_i, 0, \sin Q_1 \cdot r_i)$ , although the orientation of the spiral plane remains arbitrary due to the spin rotational symmetry. In momentum space, the magnetic moments have only the  $Q_1$  component, as shown by the spin structure factor in Figure 2d. This corresponds to  $(m_{Q_1})^2 = 0.5$  and  $(m_{Q_2})^2 = (m_{Q_3})^2 = 0$ , as plotted in Figure 3b. Consistent with the coplanar spin texture, the scalar spin chirality vanishes in this phase, as shown in Figure 3a, indicating the absence of topological character.

From an energetic viewpoint, the appearance of the single- $Q$  spiral state at  $K = L = 0$  is naturally understood. Given the constraint imposed by the fixed local spin length, the spin amplitude must satisfy the momentum-space normalization condition  $\sum_q (m_q)^2 = 1$ . In order to maximize the energy gain from the bilinear exchange interaction  $J$ , the optimal spin configuration is achieved when  $(m_{Q_1})^2 = (m_{-Q_1})^2 = 0.5$ , which corresponds to the single- $Q$  spiral state. In contrast, constructing multiple- $Q$  superpositions while maintaining the local spin-length constraint is generally difficult. Such superpositions typically induce contributions at high-harmonic wave vectors, leading to a reduction in the exchange energy compared to the single- $Q$  spiral state. This energetic disadvantage persists when we

consider the interactions at additional ordering wave vectors other than  $Q_1$ – $Q_3$  in the present effective spin model Hamiltonian; multiple- $Q$  superposition always leads to the energy loss compared to the single- $Q$  spiral state once the isotropic bilinear exchange interaction is considered.

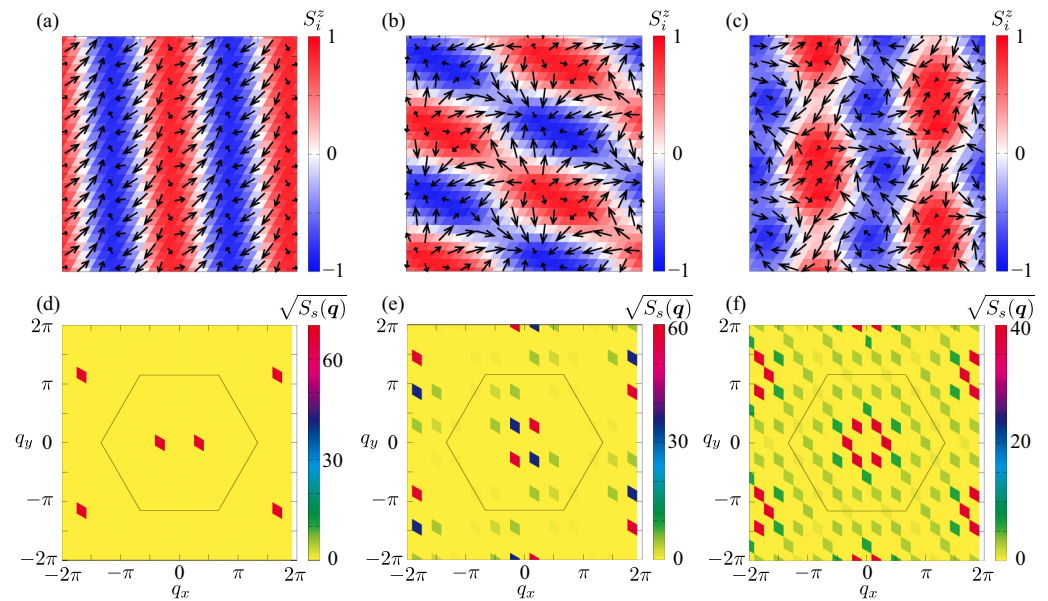


**Figure 2.** (a–c) Real-space spin configurations of (a) the single- $Q$  spiral state at  $K = 0.1$  and  $L = -0.4$ , (b) the double- $Q$  chiral stripe state at  $K = 0.1$  and  $L = 0.1$ , and (c) the  $n_{sk} = 2$  SkX at  $K = 0.1$  and  $L = 0.4$ . The arrows represent the direction of spin moments, and the contour shows the  $z$ -spin component. (d–f) Square root of the spin structure factor corresponding to (a–c). The black hexagon stands for the first Brillouin zone.

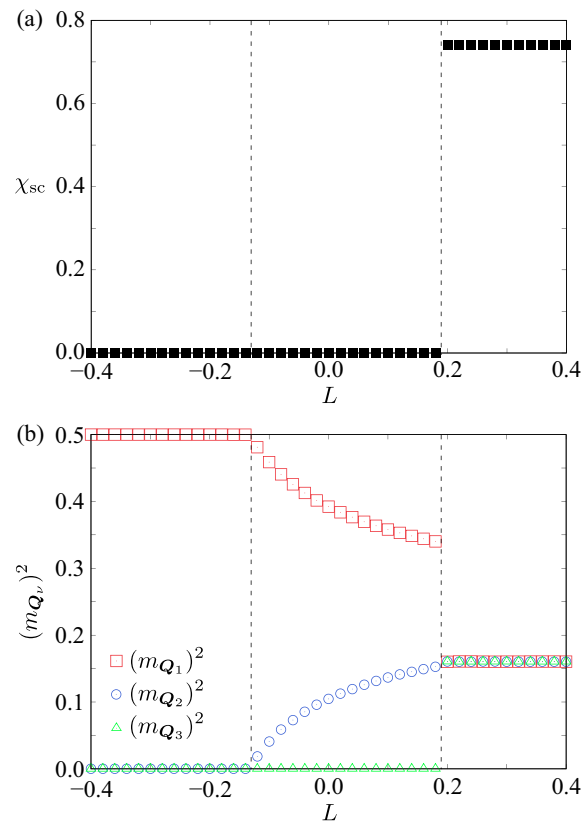


**Figure 3.**  $L$  dependence of (a) the scalar spin chirality and (b) the squared magnetic moment at  $Q_v$  for  $K = 0.1$ . The vertical dashed line represents the phase boundaries.

When the biquadratic interaction  $K$  is introduced, the ground-state spin configuration undergoes a change and becomes characterized by a double- $Q$  modulation. This corresponds to the emergence of the double- $Q$  chiral stripe state, whose spin configuration is given by

$$\mathbf{S}_i = \begin{pmatrix} \sqrt{1 - b^2 + b^2 \cos \mathbf{Q}_2 \cdot \mathbf{r}_i} \cos \mathbf{Q}_1 \cdot \mathbf{r}_i \\ b \sin \mathbf{Q}_2 \cdot \mathbf{r}_i \\ \sqrt{1 - b^2 + b^2 \cos \mathbf{Q}_2 \cdot \mathbf{r}_i} \sin \mathbf{Q}_1 \cdot \mathbf{r}_i \end{pmatrix}^T, \quad (14)$$

where the parameter  $b$  quantifies the degree of double- $Q$  modulation; the spin configuration reduces to the single- $Q$  spiral state in the limit of  $b \rightarrow 0$ . Thus, the double- $Q$  chiral stripe state can be interpreted as a linear combination of the spiral wave at  $\mathbf{Q}_1$  and the sinusoidal wave at  $\mathbf{Q}_2$ , with their respective spin components tending to be orthogonal. A representative real-space spin configuration is shown in Figure 2b, and the corresponding spin structure factor in Figure 2e, where  $\mathbf{Q}_3$  and  $\mathbf{Q}_2$  serve as the dominant and subdominant ordering wave vectors, respectively. The double- $Q$  chiral stripe state exhibits noncoplanar spin textures and hence supports finite local scalar spin chirality, although the net scalar spin chirality averaged over the entire system vanishes due to its spatial modulation. Such a state has been observed at zero field in GdRu<sub>2</sub>Si<sub>2</sub> [56].

The emergence of the double- $Q$  chiral stripe state is attributed to the biquadratic interaction  $K$ . A positive  $K$  penalizes the single- $Q$  spiral state energetically, thereby favoring the formation of a double- $Q$  superposition. This tendency can be understood analytically by evaluating the energy gain and loss associated with introducing a small double- $Q$  component characterized by a finite parameter  $b$ . Specifically, the energy gain due to the biquadratic term scales as  $Kb^2$ , while the corresponding exchange energy loss scales as  $Jb^4$  [23]. As a result, even an infinitesimal biquadratic interaction  $K$  leads to a nonzero optimal  $b$ , destabilizing the single- $Q$  spiral state and giving rise to the double- $Q$  chiral stripe state. This analytic insight is consistent with the numerical phase boundary shown in Figure 1.

For  $K \geq 0.19$ , the system transitions from the double- $Q$  chiral stripe state to another multiple- $Q$  state corresponding to the  $n_{\text{sk}} = 2$  SkX, denoted as SkX II in Figure 1. The spin configuration of this state is characterized by equal-amplitude spin modulations at all three wave vectors,  $\mathbf{Q}_1$ ,  $\mathbf{Q}_2$ , and  $\mathbf{Q}_3$ , as evidenced by the triple- $Q$  peaks in the spin structure factor shown in Figure 2f. The corresponding real-space spin configuration, shown in Figure 2c, reveals a vortex-like arrangement. Both the scalar spin chirality and the skyrmion number are finite in this phase, indicating its topologically nontrivial nature. The scalar spin chirality is plotted in Figure 3a. Especially, the skyrmion number  $n_{\text{sk}}$  is given by  $-2$  in the case of Figure 2c. However, spin configurations with  $n_{\text{sk}} = +2$  can also be obtained via simulated annealing depending on the initial spin configurations, reflecting the intrinsic degeneracy between the two configurations. This degeneracy can be lifted by including anisotropic interactions, such as bond-dependent interactions [107,108]. The emergence of the  $n_{\text{sk}} = 2$  SkX at zero magnetic field has been previously reported in the Kondo lattice model [109,110], where the biquadratic interaction was shown to play a crucial role [23]. It is also worth noting that such SkXs with high skyrmion numbers can also be found even in the absence of the biquadratic interaction when the magnetic anisotropic interactions originating from the spin-orbit coupling are present [111–115].

The inclusion of the bicubic interaction  $L$  systematically alters the stability regions of the three magnetic phases. A positive  $L$  promotes the stability of the  $n_{\text{sk}} = 2$  SkX, while a negative  $L$  favors the single- $Q$  spiral state. This trend can be qualitatively understood from the form of the bicubic interaction term  $(\mathbf{S}_{\mathbf{Q}_v} \cdot \mathbf{S}_{-\mathbf{Q}_v})^3$ , which lowers (raises) the energy of

the single- $Q$  spiral spin configuration when  $L$  is negative (positive). Although the same feature appears in the  $n_{\text{sk}} = 2$  SkX, the degree of the energy gain or loss by  $L$  is smaller than that in the single- $Q$  spiral state owing to the large intensities of the spin structure factor at  $Q_\nu$ , i.e.,  $\sum_\nu S_s(Q_\nu)$ , for the latter state. Accordingly, a positive  $L$  promotes instabilities toward multiple- $Q$  states. This tendency is also clearly manifested in the behavior of the double- $Q$  chiral stripe state, as shown in Figure 3b; the multiple- $Q$  character becomes more pronounced as  $L$  increases. Therefore, not only a positive biquadratic interaction  $K$  but also a positive bicubic interaction  $L$  is desired to realize the zero-field SkX with a high skyrmion number.

### 3.2. Finite Magnetic Fields

Next, we turn to the case where the external magnetic field is applied along the out-of-plane direction by setting  $H \neq 0$ . In accordance with previous findings in Ref. [23], the application of the magnetic field induces another topologically nontrivial phase: the  $n_{\text{sk}} = 1$  SkX, which emerges even in the absence of the bicubic interaction  $L$ . To systematically investigate the emergence of this field-induced  $n_{\text{sk}} = 1$  SkX in the presence of the bicubic interaction  $L$ , we vary  $L$  and the magnetic field  $H$  while also scanning the biquadratic interaction  $K$ . Specifically, the parameters  $L$  and  $K$  are discretized at intervals of 0.02 and the magnetic field  $H$  is varied at intervals of 0.05. The resulting phase boundary is indicated by the dashed line in Figure 1; the  $n_{\text{sk}} = 1$  SkX phase appears in the upper-right region relative to this boundary. One finds that the stability region of the  $n_{\text{sk}} = 1$  SkX expands with increasing  $L$ , mirroring the behavior of the  $n_{\text{sk}} = 2$  SkX at zero field. This observation indicates that a positive bicubic interaction  $L$  not only promotes the formation of the zero-field  $n_{\text{sk}} = 2$  SkX but also enhances the robustness of the field-induced  $n_{\text{sk}} = 1$  SkX.

To elucidate the role of the bicubic interaction  $L$ , we plot the magnetic field  $H$  dependence of magnetization and scalar spin chirality for three representative values:  $L = -0.1$ , 0, and 0.1, with fixed  $K = 0.1$ , as shown in Figure 4a and 4b, respectively. As discussed in Section 3.1, the zero-field state for  $L = -0.1$  corresponds to the double- $Q$  chiral stripe, whereas, for  $L = 0$  and 0.1, the system stabilizes into the  $n_{\text{sk}} = 2$  SkX.

Among the above  $L$  values, let us mainly discuss the result for  $L = -0.1$  since the situation without the zero-field SkX has been reported in real materials, such as  $\text{Gd}_2\text{PdSi}_3$  [47],  $\text{Gd}_3\text{Ru}_4\text{Al}_{12}$  [52]. We show the  $H$  dependence of the momentum-resolved magnetic moments for the in-plane  $xy$  component in Figure 5a and those for the out-of-plane  $z$  component in Figure 5b. Upon applying the magnetic field  $H$ , the double- $Q$  chiral stripe state undergoes a transition into an anisotropic triple- $Q$  state, which we refer to as the triple- $Q$  I state. As shown in Figure 5a, a weak but finite component appears at  $Q_3$  in the in-plane spin structure, indicating that the spin texture acquires a third wave vector. This triple- $Q$  I state is characterized by the anisotropic double- $Q$  peak structures in the in-plane spin structure factor [Figure 6d] and the single- $Q$  peak structure in the out-of-plane spin structure factor [Figure 6g]. These additional modulations slightly deform the real-space spin configuration, as shown in Figure 6a, which deviates from the configuration of the zero-field double- $Q$  chiral stripe state presented in Figure 2a.

As the magnetic field increases, the triple- $Q$  I state transitions into the  $n_{\text{sk}} = 1$  SkX at approximately  $H \simeq 0.4$ . This transition is signaled by discontinuities in the magnetization  $M^z$  in Figure 4a and the scalar spin chirality  $\chi^{\text{sc}}$  in Figure 4b. Similar to the  $n_{\text{sk}} = 2$  SkX, the  $n_{\text{sk}} = 1$  SkX is characterized by a triple- $Q$  spin structure in both  $xy$  and  $z$  spin components, as seen in Figure 6a,b,e,h. In real space, this phase exhibits a periodic lattice of magnetic skyrmions, as illustrated in Figure 6b. It is noted that, in the absence of the spin-orbit coupling, the model does not lift the degeneracy between skyrmions with opposite skyrmion

numbers (scalar spin chirality). Therefore, the energies of the SkX with  $n_{\text{sk}} = +1$  and  $n_{\text{sk}} = -1$  are identical, and the sign of the skyrmion number remains undermined.

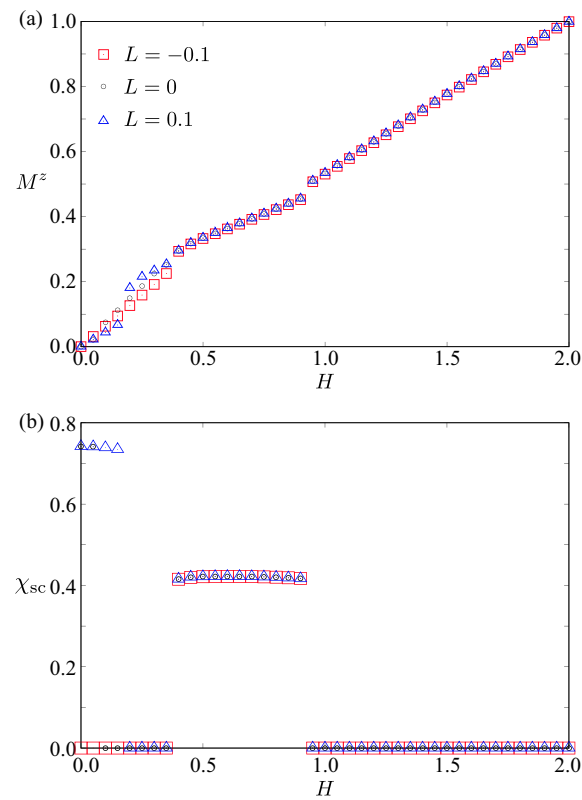


**Figure 4.**  $H$  dependence of (a) the field-induced magnetization and (b) the scalar spin chirality for  $L = -0.1$  (red square), 0 (black circle), and 0.1 (blue triangle) at  $K = 0.1$ .

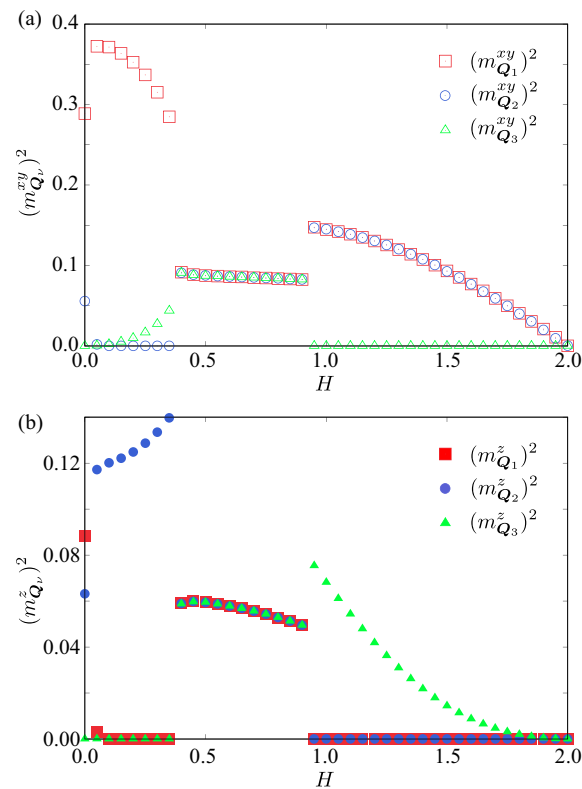


**Figure 5.**  $H$  dependence of (a) the  $xy$  and (b)  $z$  components of the squared magnetic moment at  $Q_v$  for  $Q_1$  (red square),  $Q_2$  (blue circle), and  $Q_3$  (green triangle) at  $K = 0.1$  and  $L = -0.1$ .

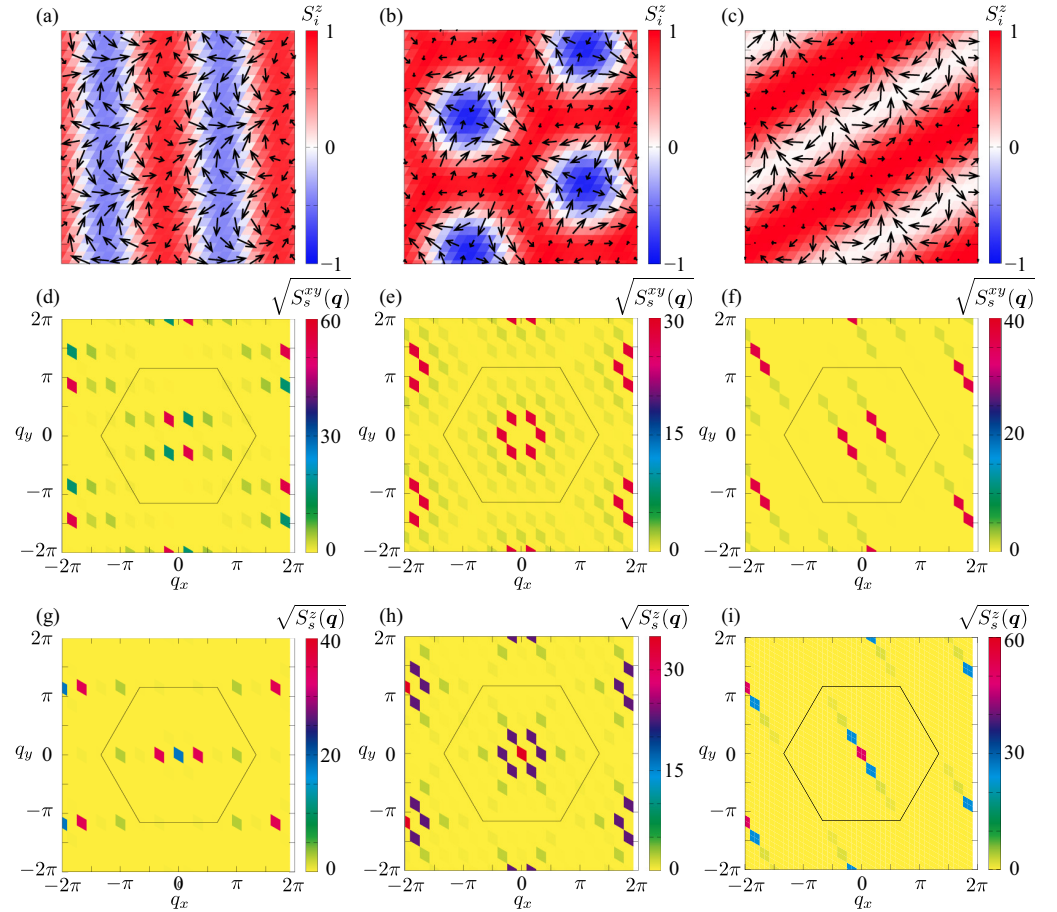


**Figure 6.** (a–c) Real-space spin configurations of (a) the triple-Q I state at  $H = 0.3$ , (b) the  $n_{\text{sk}} = 1$  SkX at  $H = 0.6$ , and (c) the triple-Q II state at  $H = 1$  for  $K = 0.1$  and  $L = -0.1$ . The arrows represent the direction of spin moments, and the contour shows the z-spin component. (d–f) Square root of the in-plane component in the spin structure factor corresponding to (a–c). The black hexagon stands for the first Brillouin zone. (g–i) Square root of the out-of-plane component in the spin structure factor corresponding to (a–c).

As the magnetic field increases further, the  $n_{\text{sk}} = 1$  SkX changes into another anisotropic triple-Q state, referred to as the triple-Q II state, within the field range of  $0.95 \lesssim H \leq 2$ . This state is characterized by the isotropic double-Q peak structures in the in-plane spin structure factor [Figure 6f] and the single-Q peak structure in the out-of-plane spin structure factor [Figure 6i]. Due to the relation of  $(m_{Q_1}^{xy})^2 = (m_{Q_2}^{xy})^2 > (m_{Q_3}^z)^2$ , the oscillation of the z component is suppressed, as illustrated in the real-space spin configuration in Figure 6c. The triple-Q II state does not exhibit a net scalar spin chirality, indicating that it is topologically trivial. Eventually, this state continuously evolves into the fully polarized state for  $H \geq 2$ .

Similar sequences of field-induced phase transitions are observed for  $L = 0$  and  $L = 0.1$ , as shown in Figure 4a,b. The key difference arises in the low-field region: for  $L = 0$  and  $L = 0.1$ , the  $n_{\text{sk}} = 2$  SkX appears, while this phase is absent for  $L = -0.1$ . In the former cases, the  $n_{\text{sk}} = 2$  SkX turns into the triple-Q I state as the magnetic field increases, and subsequent phase transitions follow a similar sequence to that observed for  $L = -0.1$ .

#### 4. Conclusions

In this study, we investigated the stability of SkX phases within an effective spin model that includes bilinear, biquadratic, and bicubic interactions on a centrosymmetric triangular lattice. Using the simulated annealing method, we constructed low-temperature magnetic

phase diagrams under both zero and finite external magnetic fields. At zero magnetic field, we demonstrated that the inclusion of a positive bicubic interaction significantly enlarges the stability region of the SkX phase with a skyrmion number of two,  $n_{\text{sk}} = 2$ , while a negative bicubic interaction favors a single-Q spiral phase. In the presence of the magnetic fields, a skyrmion crystal with skyrmion number of one,  $n_{\text{sk}} = 1$ , emerges over a wide range of parameters, and its stability is further enhanced by the inclusion of a positive bicubic interaction. These findings indicate that higher-order multi-spin interactions, particularly those resolved in momentum space, play a crucial role in stabilizing both zero-field and field-induced topological spin textures. Our results underscore the importance of momentum-resolved multi-spin interactions in realizing complex magnetic orders and provide a theoretical foundation for designing and engineering topological magnetic phases in itinerant magnets.

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