Pinning Effects of Exchange and Magnetocrystalline Anisotropies on Skyrmion Lattice

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Reorientation of skyrmion crystal (SkX) with respect to crystallographic axes is believed to be insensitive to anisotropies of fourth order in spin-orbit coupling, for which sixth order terms are considered for explanation. Here, we show that this is wrong due to an oversimplified assumption that SkX possesses hexagonal symmetry. When the deformation of SkX is taken into account, fourth order anisotropies such as exchange anisotropy and magnetocrystalline anisotropy have pinning (in this work, the word ‘pinning’ refers to the reorientation effects of intrinsic anisotropy terms) effects on SkX. In particular, we reproduce some experiments of MnSi and Fe₁₋ₓCoₓSi by considering the effect of fourth order magnetocrystalline anisotropy alone. We reproduce the 30° rotation of SkX in Cu₂OSeO₃ by considering the combined effects of the exchange and magnetocrystalline anisotropies. And we use the exchange anisotropy to explain the reorientation of SkX in VOSe₂O₅.

Keywords: skyrmion crystal, pinning effect, exchange anisotropy, magnetocrystalline anisotropy, helimagnet

1 INTRODUCTION

Helimagnets have attracted extensive interest since the first observation of magnetic skyrmions in 2009 [1]. Magnetic skyrmions in helimagnets are nontrivial spin textures, in which the spins point in all of the directions wrapping a sphere. Their topological protection [2] and facile current driven motion [3, 4] make them possible to be applied in novel spintronic and information storage devices [5, 6].

In helimagnets such as MnSi, Fe₁₋ₓCoₓSi, and Cu₂OSeO₃, the ferromagnetic exchange interaction (for Cu₂OSeO₃, the exchange interaction consists of ferromagnetic and antiferromagnetic interactions, but the field-induced ground state is closer to ferromagnetic than antiferromagnetic [7, 8]) and the Dzyaloshinsky-Moriya interaction (DMI), which arises due to the broken of space inversion symmetry [9, 10], dominate the free energy when studying bulk material free from any external magnetic field. The former favors parallel spin alignment, while the latter favors the twist of the spins. They compete with each other and result in SkX at appropriate magnetic field just below the Curie temperature [1, 11–14]. In experiments, when the magnetic field is along directions with high symmetry, such as the [001], [111] and [110] directions, the wave vectors of SkX are orientated with respect to the crystallographic axes [1, 12, 15, 16]. This indicates the existence of anisotropy energy. The anisotropies of fourth order in spin-orbit coupling, such as the exchange anisotropy and fourth order magnetocrystalline anisotropy, are widely used to explain the pinning of helical phase, the transition from helical to conical phase and the appearance of tilted conical phase [1, 17–20]. However, according to the perturbation theory [1, 12, 16, 21], which treats the anisotropies...
perturbatively and approximates SKX by a triple-Q structure with three equivalent wave vectors forming a regular triangle, they are insensitive to the pinning of SKX. As a consequence, anisotropies with higher order are proposed. In our opinion, ignoring the deformation of SKX is oversimplified, because many experiments show that the structure of SKX is sensitive to anisotropy of the system which destroys its hexagonal symmetry [22–24].

In this work, we study the pinning effects of the exchange anisotropies and the fourth order magnetocrystalline anisotropy on deformable SKX. We apply a rescaled free-energy-density model for T point group and describe Bloch SKX by a three-order Fourier expansion with deformation-related degrees of freedom. Firstly, we study four anisotropies (three types of exchange anisotropies and a fourth order magnetocrystalline anisotropy in helimagnets with T symmetry) separately. It is found that they have different pinning effects on SKX. Then, by plotting the deformation-related parameters as functions of one exchange anisotropy, we figure out that the deformation of SKX is characterized by the change of amplitudes, lengths and azimuth angles of wave vectors. Next, we compare our results with some experiments, the fourth order magnetocrystalline anisotropy may explain the pinning of SKX in MnSi [1, 25, 26] and Fe1â–FeCoSi [12, 27–29]. To reproduce the 30’ rotation of SKX in Cu2OSeO3 [16], we consider both the exchange and magnetocrystalline anisotropy, and find that at certain conditions 30’ rotation of SKX occurs when temperature or magnetic field changes. Lastly, we expand our model so that it is applicable to Cnv helimagnets hosting Néel SKX. It is found that exchange anisotropy has pinning effects on Néel SKX in C4v helimagnets but not in C3v or Cnv helimagnets.

2 MODEL

Based on the continuum spin model established by Bak and Jensen [17], we write the rescaled free-energy density [30] for helimagnets with the symmetry of T point group in the following form:

\[
\omega(\mathbf{m}) = \frac{1}{2} \sum_{i=1}^{3} \left( \frac{\partial \mathbf{m}}{\partial r_i} \right)^2 + 2 \mathbf{m} \cdot (\nabla \times \mathbf{m}) - 2 \mathbf{b} \cdot \mathbf{m} + \omega_L(\mathbf{m}) + \omega_a(\mathbf{m}).
\]  

(1)

Here, \( \mathbf{m} \) is the rescaled magnetization. The first two terms in Eq. (1) represent the ferromagnetic exchange interaction and the DMI, respectively. The third term is the Zeeman energy under the rescaled magnetic field \( \mathbf{b} \). \( \omega_L = \mathbf{r}^2 + \mathbf{m}^2 \) is the Landau expansion with the rescaled temperature \( t \), it consists of the second and the fourth order terms. The last term \( \omega_a \) is the anisotropy energy. In this work, we consider only the exchange anisotropy and the fourth order magnetocrystalline anisotropy, and we express \( \omega_a \) as

\[
\omega_a = a_{e1} \sum_{i=1}^{3} \left( \frac{\partial m_i}{\partial r_i} \right)^2 + a_{e2} \sum_{i=1}^{3} \left( \frac{\partial m_i}{\partial r_{i+1}} \right)^2 + a_{e3} \sum_{i=1}^{3} \left( \frac{\partial m_i}{\partial r_{i-1}} \right)^2 + a_m \sum_{i=1}^{3} m_i^4,
\]  

(2)

where \( a_{e1}, a_{e2} \) and \( a_{e3} \) are the coefficients of exchange anisotropy, \( a_m \) is the coefficient of magnetocrystalline anisotropy, \( r_{i+1} \) and \( r_{i-1} \) represent \( r_1 \) and \( r_3 \), respectively.

For bulk B20 materials, the skyrmion plane rotates with respect to the applied magnetic field. To describe the configuration of SKX under magnetic field with different direction, we should choose an appropriate cartesian coordinates system \( O-r_1'r_2'r_3' \) in which the magnetic field is along the \( r_3' \) axis. Let the azimuthal and polar angles that characterize the magnetic field be \( \theta \) and \( \psi \), respectively. We rotate \( O-r_1'r_2'r_3' \) counterclockwise about the \( r_3' \) axis by angle \( \theta \), and get a new cartesian coordinates system \( O-r_1'r_2'r_3' \). We then perform a second rotation, this time about the \( r_2' \) axis by angle \( \psi \), and we get the final cartesian coordinates system \( O-r_1'r_2'r_3' \). In terms of \( 3 \times 3 \) orthogonal matrices, the product of the two operations can be written as

\[
R(\theta, \psi) = R_{r_2'}(\psi)R_{r_1'}(\theta).
\]

Due to the relation

\[
R_{r_2'}(\psi) = R_{\psi}^T(\theta) R_{r_2'}(\psi) R_{\psi}^T(\theta),
\]

we have

\[
R(\theta, \psi) = R_{\psi}^T(\theta) R_{\psi}(\psi).
\]  

(3)

In the cartesian coordinates system \( O-r_1'r_2'r_3' \), we apply the \( n \)-order Fourier decomposition to describe the magnetization texture of SKX [31],

\[
\mathbf{m}^* = \mathbf{m}_0 + \sum_{q \neq 0} m_{iq} e^{i\mathbf{q} \cdot \mathbf{r}}.
\]  

(4)

Here, \( \mathbf{m}_0 = \left[ m_{01}, m_{02}, m_{03} \right]^T \) is the average magnetization over the entire SKX, and \( n \) is the number of \( n \)th order waves. The \( n \)th order waves are characterized by their wave vectors \( \mathbf{q}_n \) and polarizations \( \mathbf{m}_{iq} \). In the presence of anisotropy energy, SKX with hexagonal symmetry will go through deformation, and the deformation-related parameters are introduced through the following equation

\[
\mathbf{q}_n = \begin{bmatrix} 1 + \epsilon_{11}^{n} & \epsilon_{12}^{n} + \omega_{12} & \epsilon_{13}^{n} + \omega_{13} \\ \epsilon_{12}^{n} + \omega_{12} & 1 + \epsilon_{22}^{n} & \omega_{23} \\ \epsilon_{13}^{n} + \omega_{13} & \omega_{23} & 1 + \epsilon_{33}^{n} \end{bmatrix} \mathbf{q}_0.
\]  

(5)

In reciprocal space, \( \epsilon_{11}^{n} \) and \( \epsilon_{22}^{n} \) are the normal strains; \( \epsilon_{12}^{n} \) and \( \omega_{12} \) reflect the shear deformation and rotation of the plane spanned by \( \mathbf{q}_0 \), respectively. \( \mathbf{q}_0 \) are the undeformed wave vectors, they all can be expressed as a linear combination of \( \mathbf{q}_{11} \) and \( \mathbf{q}_{12} \) (without loss of generality, for hexagonal SKX, we set \( \mathbf{q}_{11} = [0, 1]^T \), \( \mathbf{q}_{12} = \left[ -\frac{\sqrt{3}}{2}, -\frac{1}{2} \right]^T \)). As to \( \mathbf{m}_{iq} \), we decompose them along the basis vectors \( \mathbf{P}_{ij} = \frac{1}{\sqrt{2}} \mathbf{q}_{ij}, [i\mathbf{q}_{ij}, q_{ij}, 0]^T \), and \( \mathbf{P}_{ij} = \frac{1}{\sqrt{2}} \mathbf{q}_{ij}, [i\mathbf{q}_{ij}, -i\mathbf{q}_{ij}, |\mathbf{q}_{ij}|]^T \) (for the chosen of the orthogonal basis, see Ref. [32]), and we have

\[
\mathbf{m}_{iq} = \sum_{k=1}^{3} c_{ijk} \mathbf{P}_{ijk},
\]  

(6)

where \( c_{ijk} = \epsilon_{ij}^{k} + i \omega_{ijk} \) (\( k = 1, 2, 3 \)) are the complex coefficients.
According to Eqs. (1) and (2), the free energy density is a functional of $m_i$ and $m_{jk} (m_{jk} \text{ denotes } \frac{d m_j}{d r_i})$, i.e., $\omega = \omega (m_i, m_{jk})$. Applying the following coordinate transformation

\[ m_i = \sum_{i=1}^{3} R(\theta, \psi)_{i,i} m_i', \tag{7} \]

\[ m_{jk} = \sum_{j'k'=1}^{3} R(\theta, \psi)_{j,j'} R(\theta, \psi)_{k,k'} m_{j'k'}, \tag{8} \]

the free-energy density can be rewritten as, after averaging over a magnetic unit cell

\[ \omega = \omega (\epsilon_{11}, \epsilon_{22}, \epsilon_{12}, \alpha_{11}, m_{01}, m_{02}, m_{03}, \epsilon_{jk}, \alpha_{jk}). \tag{9} \]

At certain temperature $t$, magnetic field $b$, rotation angles $\theta$ and $\psi$, exchange and magnetocrystalline anisotropies $a_{c1}, a_{c2}, a_{c3}$ and $a_m$, the parameters describing SkX are calculated via minimization of Eq. (9). In this work, we set the order of Fourier expansion $n = 3$.

Our analytical method can only deal with periodic magnetization structure. For the cases where the periodicity of skyrmions is broken, e.g., the thermal-induced disorder or the pinning from impurities, the review [33] and references therein are good to refer to.

### 3 RESULTS AND DISCUSSION

We first investigate the pinning effects of anisotropies $a_{c1}, a_{c2}, a_{c3}$ and $a_m$ on Bloch SkX, separately. The value of $\theta$ is $45^\circ$; thus, $\psi = 0^\circ, 55^\circ$ and $90^\circ$ correspond to the directions [001], [111] and [100], respectively. The temperature and the magnetic field are set to be $t = 0.5$ and $b = [0, 0, 0.2]^T$ (in the $O-\mathbf{r_1'}\mathbf{r_2'}\mathbf{r_3'}$ coordinate system) so that SkX exists as a stable or metastable state. The thermodynamic parameters for MnSi [34] and Cu$_2$OSeO$_3$ [7] are available. Using these parameters, we have $(T, B) = (28.0K, 87mT)$ for MnSi and $(T, B) = (58.1K, 4.3mT)$ for Cu$_2$OSeO$_3$, these points are near the skyrmion stable region in the magnetic field-temperature phase diagram. The anisotropy coefficients of helimagnets are hard to get in experiments. We only find the relative exchange anisotropy for GaV$_4$O$_8$, which is about $5\%$ [35]. In this work, the values used for the anisotropy coefficients are $0.005 \sim 0.1$. We think, to some extent, the values are within a realistic range.

We change the rotation-related parameter $\omega^{\theta i}$, minimize the free energy density and then plot $\omega$ as a function of $\psi_{11}$, the angle between the wave vector $\mathbf{q}_{11}^d$ and the $\mathbf{r}_1'$ axis, in Figure 2. Figures 2A,B show the effects of exchange anisotropy $a_{c1}$ on SkX. For $b||[001]$, a negative $a_{c1}$ (Figure 2A) prefers a wave vector along the [100] or [010] direction; while a positive $a_{c1}$ (Figure 2B) prefers a wave vector along the [110] or [101] direction. For $b||[111]$ and [110], $\omega$ reaches its minimum at $\psi_{11} = 90^\circ$ and $\psi_{11} = 60^\circ$, respectively (Figures 2A,B), i.e., both negative and positive $a_{c1}$ prefer a wave vector along the [110] direction for $b||[111]$ and [010] direction for $b||[001]$. Figures 2C–F show the effects of $a_{c2}$ and $a_{c3}$ on SkX, respectively. For $b||[001]$, a $a_{c2}$, no matter what its sign is, pins a wave vector of SkX along the [010] direction; while a $a_{c3}$ pins a wave vector of SkX along the [100] direction. For $b||[111]$, $\psi_{11}$ is between $45^\circ$ and $60^\circ$ or between $60^\circ$ and $75^\circ$, meaning that no wave vector is along any direction with high symmetry. For $b||[110]$, both $a_{c2}$ and $a_{c3}$ pin a wave vector of SkX along the [001] direction. Figures 2G,H show the effects of $a_m$ on SkX. For $b||[001]$ and [111], $a_m$ has the same the pinning effects on SkX as $a_{c1}$; while for $b||[110]$, $a_m$ is different from $a_{c1}$, it results in a wave vector along the [110] direction.

In Figure 2, we give the values of $\Delta \omega$, difference between the maximum and minimum of free energy. They are much smaller than $\omega$ (about $-0.1$), about $10^{-9}$ for $a_{c2} = \pm 0.05$ and $\psi = 0^\circ$, and about $10^{-7}-10^{-5}$ for other cases. The strength of $a_m$-induced anisotropy in [001] plane is obviously smaller than that in [111] and [110] planes. Comparing the energy curves for $a_{c2}$ and for $a_{c3}$, we find that they have the same $\Delta \omega$ and are symmetric about $\psi_{11} = 60^\circ$. The similarity between $a_{c2}$ and $a_{c3}$ can be inferred from their energy formula in Eq. 2, which are related by the coordinate transformation $r_1 \leftrightarrow r_2$. It should be noticed that for $b||[001]$, the periodicity of $\omega(\psi_{11})$ is $30^\circ$ for $a_{c2}$ and $a_m$, and is $60^\circ$ for $a_{c2}$ and $a_{c3}$. This can be explained by symmetry analysis. The $a_{c2}$ and $a_m$ terms in Eq. (2) have a higher symmetry than $T$ point group, they are invariant with respect to fourfold $C_4$ rotations around the $(001)$ axes. $a_{c2}$ and $a_m$ terms in Eq. (2) have lower symmetry and are invariant with respect of twofold $C_2$ rotations around the $(001)$ axes, meaning the broken of the equivalence between [100] and [010].
SkX is treated as a deformable structure. To reveal how anisotropy energy deforms SkX, we take $a_{e2}$ as an example and plot some deformation-related parameters as functions of $a_{e2}$ in Figure 3. It can be found that for nonzero $a_{e2}$, 1) the wave amplitudes $c_{111}^{\parallel}$ and $q_{12}^{d}$ are not equal to each other (Figure 3A), 2) the wave lengths $q_{11}^{d}$, $q_{13}^{d}$, $q_{21}^{d}$, and $q_{23}^{d}$ are not equal to each other (Figure 3B), and 3) the angle $\phi_{12}$ between $q_{11}^{d}$ and $q_{23}^{d}$ deviates from 120° (Figure 3C). We conclude that anisotropy energy breaks the hexagonal symmetry of SkX by changing the amplitudes of, the lengths of, and the angles between the wave vectors. In many of the small-angle neutron scattering (SANS) experiments (1; 27; 36), the observed Bragg spots have different intensities, this might be explained by our calculation. By energy minimization, we find that the dominant coefficients $c_{ik}$ are $c_{111}^{\parallel}$, $c_{121}^{\parallel}$, and $c_{111}^{\perp}$, which represent the wave amplitudes of the first order waves with vectors $q_{11}^{d}$, $q_{21}^{d}$, and $q_{23}^{d}$. Their ratios reflect the relative intensities of the first-order Bragg spots. In the inset of the

**Figure 3A**, two Bragg spots are brighter than the other four, because $c_{111}^{\parallel} = c_{121}^{\parallel} < c_{111}^{\perp}$.

We now compare our results with some experiments. The SANS experiments of Fe$_{1-x}$Co$_x$Si [12, 27–29] show that for $b||[111]$ and $[100]$ directions, two of the six scattering spots are aligned with the [110] axis; for $b||[001]$, two sets of six scattering spots are observed, one is aligned with one of the [100] direction, the other one the [010] direction. This is compatible with the results shown in Figure 2G. Therefore, a negative $a_{m}$ may explain the pinning of SkX in Fe$_{1-x}$Co$_x$Si. Different from Fe$_{1-x}$Co$_x$Si, MnSi [26] is observed to have a wave vector along the [110] direction for $b||[001]$. This may be explained by a positive $a_{m}$ (Figure 2H). We should point out that at zero magnetic field, a negative (positive) $a_{m}$ prefers the $\langle 100 \rangle$ ($\langle 111 \rangle$) directions for the helical state, which is indeed the case for Fe$_{1-x}$Co$_x$Si (MnSi) [11, 28, 29, 37, 38]. In the work [26], two kinds of sixth order magnetocrystalline anisotropies $m$.
In another published work [41], we explain the electric-field-induced continuous rotation of SkX [42] by extending the present model. Unlike a previous theory [42] which explains the phenomenon by considering both the fourth and sixth order magnetocrystalline anisotropies, we find that a combination of fourth order exchange anisotropies and magnetocrystalline anisotropies dominates the phenomena. This is because the theory used in [42] obtains a positive coefficient of the fourth order magnetocrystalline anisotropy \( a_m \) which is inconsistent with other experiments [20, 39, 40], while our model obtains a negative \( a_m \).

In polar magnets with \( C_{nm} (n = 3, 4, 6) \) symmetry, the DMI and the exchange anisotropy are different from that in Eq. (1). By applying the symmetry analysis, we derive the DMI: \( \omega_{\text{DMI}} = 2m_1m_{31} - 2m_2m_{11} + 2m_3m_{22} - 2m_4m_{22} \) (in this case, the Néel SkX is stabilized, and the basis vectors in Eq. (6) are chosen to be \( P_{ij} = \frac{1}{\sqrt{2}} ( - i q_{i x}, - i q_{i y}, |q_{ij}|)^T \), \( P_{ij} = \frac{1}{\sqrt{2}} ( - i q_{i x}, i q_{i y}, |q_{ij}|)^T \) and \( P_{ij} = \frac{1}{\sqrt{2}} ( i q_{i x}, i q_{i y}, |q_{ij}|)^T \) [32]), and the exchange anisotropy:

\[
\omega_{\text{ex}} = a_{\text{ex}} \left( \frac{\partial m_z}{\partial r_1} \right)^2 + \left( \frac{\partial m_1}{\partial r_2} \right)^2 + a_e \left( \frac{\partial m_2}{\partial r_1} \right)^2 + \left( \frac{\partial m_3}{\partial r_2} \right)^2 \tag{10}
\]

Here, we have ignored the terms \( (\partial m_2/\partial x)^2 \) due to the fact that in polar magnets, SkX plane is perpendicular to the \( n \)-fold axis no matter what direction the applied magnetic field is along [43, 44]. The term with coefficient \( a_{\text{ex}} \) is rotationally symmetric, and it has no pinning effects on SkX.

For \( C_{3y} \) and \( C_{6v} \) point groups, \( a_{\text{ex}} \) is zero. As a result, the orientation of the wave vector of SkX is insensitive to the exchange anisotropy. In Ref. [44], based on a discrete model and Monte Carlo simulations, the authors attribute the pinning of Néel SkX in \( C_{3y} \) polar magnet \( \text{GaV}_2\text{Se}_6 \) to the Dzyaloshinskii-Moriya vectors. However, according to the continuum model, the DMI possesses rotational symmetry and has no pinning effects on Néel SkX. In our opinion, this contradiction is because the

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**FIGURE 4** Combined effects of the exchange and fourth order magnetocrystalline anisotropies on SkX. The absolute value of exchange anisotropy coefficient is fixed to be 0.005, the value of magnetocrystalline anisotropy coefficient is fixed to be = -0.1 or = -0.05 or = -0.01. The results are calculated at \( \varphi_3 = 90^\circ \), \( b = 0.2 \) and \( t = 0.5 \).
continuum model ignores higher order DMI terms which emerge during the process of transforming the discrete model to the continuum model. These higher order DMI terms possess lower symmetry $C_{4v}$ or $C_{6v}$ and might reorientate Néel SkX.

For $C_{4v}$ point groups, we have $a_{e4} \neq 0$. The term with coefficient $a_{e4}$ will deform and thus reorientate the Néel SkX. Because it possesses $C_{4v}$ symmetry, which is different from the $C_{6v}$ symmetry possessed by the undeformed SkX. To study the pinning effects of $a_{e4}$ on Néel SkX, we plot $\omega$ as a function of $\varphi_{11}$ for 1) positive and 2) negative $a_{e4}$ in Figure 6. It is found that a positive $a_{e4}$ prefers a wave vector along the $[110]$ or $[1\bar{1}0]$ direction, and a negative $a_{e4}$ prefers a wave vector along the $[100]$ or $[0\bar{1}0]$ direction. In experiments, very few $C_{4v}$ helimagnets hosting Néel SkX have been found. VOSe$_2$O$_5$ [45] is one of these $C_{4v}$ helimagnets, in which the Néel SkX is orientated with a wave vector along the $[100]$ or $[00\bar{1}]$ direction. In previous theories, less attention has been paid to the reorientation of Néel SkX in $C_{4v}$ helimagnets. Here, a negative $a_{e4}$ gives a possible explanation for the SkX-reorientation-related phenomena in VOSe$_2$O$_5$.

4 CONCLUSION

In conclusion, the exchange and fourth order magnetocrystalline anisotropies deform SkX by changing the amplitudes of, the lengths of, and the angles between wave vectors and thus show pinning effects on SkX. The results of magnetocrystalline anisotropy [exchange anisotropy] may explain some experiments of MnSi and Fe$_{1-x}$Co$_x$Si [VOSe$_2$O$_5$]. By considering the exchange and magnetocrystalline anisotropies at the same time, the 30° rotation of SkX in Cu$_2$OSeO$_3$ is reproduced.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/Supplementary Material, further inquiries can be directed to the corresponding authors.

AUTHOR CONTRIBUTIONS

XW and YH conceived the idea. XW finished the analytical deduction, and performed all the calculations. XW, YH, ZH, and BW discussed the results for revision and co-wrote the manuscript.

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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