A theory of skyrmion crystal formation†

Xu-Chong Hu, a,b Hai-Tao Wu a,b and X. R. Wang a,b

A generic theory about skyrmion crystal (SkX) formation in chiral magnetic thin films and its fascinating thermodynamic behaviours is presented. A chiral magnetic film can have many metastable states with an arbitrary skyrmion density up to a maximal value when a parameter $\kappa$, which measures the relative Dzyaloshinskii-Moriya interaction (DMI) strength, is large enough. The lowest energy state of an infinite film is a long zig-zagged ramified stripe skyrmion occupied the whole film in the absence of a magnetic field. Under an intermediate field perpendicular to the film, the lowest energy state has a finite skyrmion density. This is why a chiral magnetic film is often in a stripy state at a low field and a SkX only around an optimal field when $\kappa$ is above a critical value. The lowest energy state is still a stripy helical state no matter with or without a field when $\kappa$ is below the critical value. The multi-metastable states explains the thermodynamic path dependences of various metastable states of a film. Decrease of $\kappa$ value with the temperature explains why SkXs become metastable at low temperature in many skyrmion systems. These findings open a new avenue for SkX manipulation and skyrmion-based applications.

1 Introduction

Skyrmion crystals (SkXs) have attracted enormous attention in the past decade because of their academic interest and potential applications.12 Since they were first unambiguously observed in chiral magnets,3–6 Skyrmions and SkXs are good platform for studying various physics1,4,13–16 such as the emergent electromagnetic fields and topological Hall effect.18–19 Three types of circular skyrmions in chiral magnets,13–15 have been identified, namely, Bloch skyrmions, hedgehog skyrmions, and anti-skyrmions. SkXs have been observed in many magnetic materials with Dzyaloshinskii-Moriya interaction (DMI)3,16–18 or with geometric frustration.17–19

Although many theories predicted SkXs at low or even zero temperatures and in the absence of a magnetic field, it is an experimental fact20,21 that SkXs form thermodynamically under the assistance of an optimal magnetic field and near the Curie temperature to date. Once formed, however, SkXs can be metastable in very large temperature-field regions. For example, a SkX can exist at zero magnetic field and a field much higher than an optimal value by cooling a SkX first in the optimal field to a low temperature followed by removal or rise of the magnetic field. A SkX disappears during the zero field warming and high field warming.12,27 A SkX is a thermal equilibrium state only in a narrow magnetic-field range and near the Curie temperature. The phase region for stable SkXs is normally larger in a thinner film20,21 than that of a thicker film or a bulk material.3,26 Outside the range, SkXs can only be metastable states.20,21 At zero field and a temperature much lower than the Curie temperature, the thermal equilibrium phase becomes a collection of stripe spin textures known as a helical state.20,21 Existing theories have not provided a satisfactory answer to the field effects and the thermodynamic path dependences of various experimentally observed phases.

The thermal equilibrium state of a system results from the competition between the internal energy and entropy. Entropy dominates the higher temperature phases while the internal energy determines the lower-temperature ones. If one views a long flexible stripe as a long flexible polymer, then stripe entropy, according to de Gennes theory,22 is proportional to the logarithm of stripe length. The end-end distribution function of a flexible polymer of $N$ monomers is $p(\vec{x}) \sim [3/(2\pi Na^2)]^{3/2} \exp[-3\vec{x}^2/(2Na^2)]$, where $a$ is monomer size. In information theory, it is well known that the Gaussian probability distribution with a fixed mean and variance gives the maximal entropy. The polymer entropy is $S = -\int p\log(p)d^3\vec{x} \approx \frac{3}{2}\log(N)$. Previous study3 under the assumption of infinite long rigid stripes, argued that the translational entropy of helical states is much smaller than SkX entropy that is proportional to the logarithm of number of sites in one unit cell of the SkX. Obviously, real stripes are flexible as shown in both micromagnetic simulations and experiments and rigid assumption is incorrect. In reality, translational entropy cannot
compete with the entropy of very long stripes in helical states mentioned. This view is consistent with experimental facts that numerous stripe structures, including ramified stripes and maze, were observed while few variations of SkX structures are possible. One interesting mystery about low entropy SKX is its metastability at lower temperatures even when a SkX is a thermal equilibrium state at higher temperatures. This seemingly contradicts the general principle that a higher temperature prefers a higher entropy state. Thus, a proper understanding is needed.

In this paper, the roles of magnetic field in skyrmion crystal (SKX) formation is revealed. We show that a chiral magnetic thin film of a given size with an arbitrary number of skyrmions up to a critical value is metastable when skyrmion formation energy is negative and skyrmions are stripes that are usually called helical states. The energy and morphology of theses metastable states depend on the skyrmion density and the magnetic field perpendicular to the film. At zero field, the energy increases with skyrmion number or skyrmion density. Thus, the film at thermal equilibrium below the Curie temperature and at zero field should be helical states consisting of a few stripe skyrmions. At non-zero field, the film with Qm skyrmions has the lowest energy. Qm first increases with the field up to an optimal value then decreases with the field. A parameter called κ that measures the relative DMI strength to the exchange stiffness and magnetic anisotropy plays a crucial role. For κ above a critical value, Qm near the optimal field is large enough such that the average distance between two neighboring skyrmions is comparable with skyrmion width, and skyrmions form a SKX. For κ below the critical value, the system prefers a strip phase or a mixing phase consisting of stripes and circular skyrmions even in the optimal fields.

2 Model and methodology

We consider a chiral magnetic thin film of thickness d in xy-plane. The magnetic energy of a spin structure ̂m with an interfacial (if.) DMI is

\[ E = d \int \left[ A |\nabla |^2 + D |(\nabla \cdot \hat{m}) m_z - m_z \nabla \cdot \hat{m}|^2 \right] + K_u (1 - m_z^2) - \mu_0 M_s \hat{H}_\parallel \cdot \hat{m} + \mu_0 H M_s (1 - m_z^2) \right] dS, \]  

and energy with a bulk DMI is

\[ E = d \int \left[ A |\nabla |^2 + D |(\nabla \cdot \hat{m}) m_z - m_z \nabla \cdot \hat{m}|^2 \right] + K_u (1 - m_z^2) - \mu_0 M_s \hat{H}_\parallel \cdot \hat{m} + \mu_0 H M_s (1 - m_z^2) \right] dS, \]  

where A, D, K_u, H, M_s, \hat{H}_\parallel and \mu_0 are exchange stiffness constant, DMI coefficient, the magneto-crystalline anisotropy, perpendicular magnetic field, the saturation magnetization, the demagnetizing field and the vacuum permeability, respectively. Ferromagnetic state of m_z = 1 is set as zero energy E = 0. For an ultra thin film, demagnetization effect can be included in the effective anisotropy K = K_u - \mu_0 M_s^2 / 2. This is a good approximation when the film thickness d is much smaller than the exchange length. It is known that isolated circular skyrmions are metastable state of energy 8πAd\sqrt{1-κ} when κ = π^2D^2/(16AK) < 1 [25].

Spin dynamics in a magnetic field is governed by the Landau-Lifshitz-Gilbert (LLG) equation,

\[ \frac{\partial \hat{m}}{\partial t} = -\gamma \hat{m} \times \hat{H}_{\text{eff}} + \alpha \hat{m} \times \frac{\partial \hat{m}}{\partial t}, \]  

where γ and α are respectively gyromagnetic ratio and Gilbert damping constant. \hat{H}_{\text{eff}} = \frac{2A}{M_s} \nabla^2 \hat{m} + \frac{2K_u}{M_s} m_z \hat{m} + H \hat{m} + \hat{H}_A + \hat{H}_{\text{DM}} + \hat{h}

is the effective field including the exchange field, the anisotropy field, the external magnetic field along $\hat{z}$, the demagnetizing field, the DMI field $\hat{H}_{\text{DM}}$, and a temperature-induced random magnetic field of magnitude $h = \sqrt{2\kappa k_B T} / (M_s \mu_0 V \Delta t)$, where $\Delta t$, $\Delta t$, and $T$ are the cell volume, time step, and the temperature, respectively.

In the absence of energy sources such as an electric current and the heat bath, the LLG equation describes a dissipative system whose energy can only decrease [26][29]. Thus, solving the LLG equation is an efficient way to find the stable spin textures of Eqs. (1) and (2). The typical required time for a sample of 200nm is about 1 ns that can be estimated as follows. The spin wave speed is over 1000m/s, thus two spins 200nm apart can communicate with each other within a time of 0.2ns. If two spins can relax after 5 times of information-exchange, it gives a relaxation time of 1 ns that is much longer than the individual dynamic time $h/(k_B T_c) \approx 160$ ps if we use $T_c \approx 30$K.

This method is much faster and reliable than Monte Carlo simulations and direct optimizations of predefined spin textures [20][24]. People [25] have used predefined spin structures to determine whether helical states or SKX is more stable than the other. Such an approach requires a superb ability of guessing solutions of a complicated nonlinear differential equation in order to obtain correct physics, a formidable task. Very often, this approach ends with an inaccurate trial solution. The inaccuracy can easily be demonstrated by using the trial solution as the input of a simulator such as MuMax3 and seeing how far it ends at a steady state. We do not know any work used a reasonably good profile for stripe skyrmions in this approach. This may be why people using this approach did not carry out a self-consistent check because it is doomed to fail.

Interestingly and unexpectedly, for a magnetic film described by energy (1) or (2) with 5 parameters, stable/metastable structures are determined by only $κ = \frac{π^2D^2}{16AK}$ and $κ' = \frac{π^2D^2}{8κ_0AM_s}$. Let us use the bulk DMI as an example to prove this assertion,

\[ E = \int \left\{ A |\nabla \hat{m}|^2 + D |(\nabla \cdot \hat{m}) m_z - m_z \nabla \cdot \hat{m}|^2 \right\} - \frac{π^2D^2}{16A} \int \left\{ \left\{ \frac{4A}{πD} |\nabla \hat{m}| \right\}^2 + \frac{4}{π} \left\{ \hat{m} \cdot \left\{ \frac{4A}{πD} \nabla \cdot \hat{m} \right\} \right\} \right\} - \frac{1}{κ} (m_z)^2 - \frac{2}{κ'} (m_z)^2 \right\} dV. \]  

Replace $x$ by dimensionless variable $\bar{x} = \frac{πD}{16A} x$ and then relabel $\bar{x}'$ back to $x$, the steady spin structures satisfy equation

\[ \nabla^2 \hat{m} + \frac{4}{π} (\nabla \cdot \hat{m}) + \frac{1}{κ} m_z \hat{z} + \frac{1}{κ'} \hat{z} = 0. \]
A/D is a natural length of the system. Similarly, for the case of interfacial DMI, the equation is

\[ \nabla^2 \hat{m} + \frac{4}{\pi} \left[ (\nabla \cdot \hat{m}) \hat{z} - \nabla m_\perp \right] + \frac{1}{\kappa} m_\perp \hat{z} + \frac{1}{\kappa'} \hat{z} = 0. \] (6)

Both equations depend only on \( \kappa = \frac{\kappa_0 D^B}{10^7} \) and \( \kappa' = \frac{\kappa_0 D^D}{10^7} \). To shed some light on the meaning of these two parameters, we note that exchange interaction (A), anisotropy (K) and external magnetic field (H) prefers spins aligning along the same direction while DMI (D) prefers spin curling. \( K \) and \( H \) are two different anisotropy parameters. We can further see the similarity of \( \kappa \) and \( \kappa' \) by examine the equations for static stripe skyrmions whose profile is \( \hat{m} = (\cos \gamma \sin \Theta, \sin \gamma \sin \Theta, \cos \Theta) \) and \( \gamma = 0 \) for interfacial DMI and \( \gamma = \pi/2 \) for bulk DMI [44,45]. From Eqs. (3) and (6), equation for \( \Theta \) is \( \frac{\partial \Theta}{\partial t} = \frac{1}{2 \kappa} \sin 2\Theta - \frac{1}{2} \sin \Theta = 0 \) with boundary conditions of \( \Theta(x = 0) = 0 \) and \( \Theta(x = 2L) = 2\pi \), here 2L being the stripe width.\(^ {44} \) In the absence of a magnetic field, it is known that \( \kappa = 1 \) separates an isolated circular skyrmion from condensed stripe skyrmions \(^ {44,45} \). We expect then that, in the absence of anisotropic constant \( K = 0, \kappa' = 4 \) separates an isolated circular skyrmion from condensed stripe skyrmions. Thus both parameters describe the competition between collinear order and curling (spiral) order. With that being said, two anisotropies are also fundamentally different. \( H \) is obviously unidirectional, i.e. spins parallel and anti-parallel to the field are not same, in contrast to the bidirectional nature of \( K \). We will also see below how \( H \) can make a SkX to be the lowest energy state while \( K \) does not have such an effect.

3 Results and discussions

Several sets of very different model parameters relevant to various bulk chiral magnets are used to illustrate our main findings. These sets are listed in Table 1 as samples A, B, and C. Since DMI in films is mostly interfacial, we replace the bulk DMI by the interfacial DMI in most of our studies. However, the results are essentially the same no matter which DMI is used. The verification of this assertion is given in section 3.7.

Stripe skyrmions exist in all three samples with \( \kappa > 1 \).\(^ {44,45} \) The typical spin precession time is order of THz, and spin relaxation time across the sample is order of nano-seconds as demonstrated in both \( E(t) \) and \( Q(t) \). It should be pointed out that all results reported here are similar although very different sets of parameters are used. These parameters mimic very different materials whose Curie temperature differ by more than 10 times. Periodic boundary conditions are used to eliminate boundary effects and the MuMax3 package\(^ {22} \) is employed to numerically solve the LLG equation for films of 200 nm \( \times \) 200 nm \( \times \) 8 nm. The static magnetic interactions are fully included in all of our simulations. The mesh size is of 1 nm \( \times \) 1 nm \( \times \) 1 nm unless otherwise stated. The number of stable states and their structures should not depend on the Gilbert damping constant. We use a large \( a = 0.25 \) to speed up our simulations.

3.1 Metastable spin structures and density dependence of skyrmion morphology

Figure 1 shows typical stable structures of 10, 50, and 150 skyrmions in a 200 nm \( \times \) 200 nm \( \times \) 8 nm film under various perpendicular magnetic fields of \( \mu_0 H = 0, 0.1, \) and 0.4T for sample A with \( \kappa = \frac{\kappa_0 D^B}{10^7} = 8.3 \). They are steady-state solutions of the LLG equation at zero temperature with initial configurations of 10, 50, and 150 nucleation domains of \( m_z = -1 \) in the background of \( m_z = 1 \). Each domain is 5 nm in diameter, and domains are initially arranged in a square lattice. Figs. 1(a1-a3) are zero field structures of 10 (a1), 50 (a2), and 150 (a3) skyrmions characterized by skyrmion number \( Q \) defined as \( Q = \int \rho \, dV \), where \( \rho = \frac{1}{2 \mu_0} \hat{m} \cdot (\partial \hat{m} \times \partial \hat{m}) \) is the skyrmion charge density. The colour encodes the skyrmion charge distribution. Each small domain of initial zero skyrmion number becomes a skyrmion of \( Q = 1 \) after a few picoseconds. Interestingly, both positive and negative charges appear in a stripe skyrmion while only positive charges exist in a circular skyrmion. Figures 1(d1-d3) show how total \( Q \) (the left y-axis and the red curves) and energy \( E \) (the right y-axis and the blue curves) change with time. \( Q \) reaches its final stable values within picoseconds while \( E \) decreases to it minimal values in nanoseconds. The negative skyrmion formation energy explains well the stripe skyrmion morphology that tries to fill up the whole film in order to lower its energy, in contrast to circular skyrmions for positive formation energy.\(^ {19} \) Unexpectedly, the film can host an arbitrary number of skyrmions up to a large value (see Fig. 4 below). At a low skyrmion number of \( Q = 10 \), the film is in a helical state consisting of ramified stripe skyrmions of well-defined width of 8.1 nm. Spin profile of stripes are well characterized by width \( L \) and wall thickness \( w \). Stripe width depends on materials parameters as \( L = a(\kappa) A/D, \) where \( a = 2 \pi \) for \( \kappa >> \).\(^ {1\circ} \) The film with \( Q = 50 \) is in a helical state consisting of rectangular stripe skyrmions while it is a SkX of triangular lattice with \( Q = 150 \) skyrmions. Skyrmion density for \( Q = 150 \) is so high that the distance between two neighbouring skyrmions is comparable to the stripe width, and skyrmion-skyrmion repulsion compress each skyrmion into a circular object. Skyrmions at high skyrmion density prefer a triangular lattice as shown in Fig. 1(a3) instead of the initial square lattice.

Figures 1(b1-b3) and 1(c1-c3) plot the metastable structures under fields \( \mu_0 H = 0.1 \)T (b1-b3) and \( \mu_0 H = 0.4 \)T (c1-c3) with the same initial states as those in (a1-a3). Stripes of \( m_z > 0 \) (the grey regions) parallel to \( H \) expand while the white regions of \( m_z < 0 \), anti-parallel to \( H \), shrink. This is showed in Figs. 1(b1, b2, c1, and c2). Moreover, the amount of increase and decrease of white and grey stripes are not symmetric such that skyrmion-skyrmion repulsion is enhanced by the field, and SkXs tend to occur at lower skyrmion density. This trend can be clearly seen in Figs. 1(c2) and (c3). Figures 1(d2) and (d3) show similar behaviour for \( \mu_0 H = 0.1 \)T (d2) and 0.4T (d3) as their counterparts (d1) at zero field: Skyrmion number \( Q \) grows to its final values rapidly in picoseconds and \( E \) monotonically decreases to its minimum in nanoseconds. Figure 1 shows unexpectedly that the nature shape of skyrmions are various types of stripes when \( \kappa > 1 \) and at low skyrmion density, in contrast to the current belief that all
Table 1 Three sets of parameters

| Sample | \( A \) (pJ/m) | \( D \) (mJ/m²) | \( K_u \) (mJ/m³) | \( M_s \) (MA/m) | \( H \) (T) | DMI type | Materials |
|--------|--------------|--------------|----------------|-------------|---------|----------|-----------|
| A      | 0.4          | 0.33         | 0.036          | 0.15        | variable | itf. & bulk | MnSi      |
| B      | 0.39         | 0.0544       | 0.00103        | 0.03        | 0.014   | bulk     | Fe₀.5Co₀.5Si |
| C      | 5            | 2            | 0.063          | 0.15        | 0.3     | bulk     | Co₀.3Mn₀.7Zn |

Fig. 1 Metastable structures of 10 (a1-c1), 50 (a2-c2) and 150 (a3-c3) skyrmions in a 200nm × 200nm × 8nm film at zero temperature for \( \mu_0H = 0 \) T (a1-a3), 0.1 T (b1-b3) and 0.4 T (c1-c3), respectively, from MuMax3. The insets of (a1), (c1), (c3) are the spin profiles along the red lines. The enlarged figures and descriptions are in Fig. 2. (d1-d3) \( Q \) (the left \( y \)-axis and the red curves) and \( E \) (the right \( y \)-axis and the blue curves) of 10 (the dash lines), 50 (the solid lines), and 150 (the dot-dash lines) skyrmions as functions of the time (in the logarithmic scale) for \( \mu_0H = 0 \) T (d1), 0.1 T (d2) and 0.4 T (d3). The initial configurations are 10, 50 and 150 nucleation domains of zero skyrmion number and of 5nm in diameter each arranged in a square lattice.
skyrmions are circular.

Spin profiles of stripe skyrmions in the presence of a magnetic field are described by \( \Theta(x) = 2\arctan \left( \frac{\sinh(|x|/w_1)}{\sinh(L_z/2w_1)} \right) \) \((m_2 < 0)\) and \( \Theta(x) = 2\arctan \left( \frac{\tan \left( \frac{|x|}{w_2} \right)}{\tan \left( \frac{L_z}{2w_2} \right)} \right) \) \((m_2 > 0)\), respectively, with \(|x| \leq L_z/2\). \( \Theta \) is the polar angle of the magnetization at position \( x \) and \( x = 0 \) is the center of a stripe where \( m_2 = -1.1 \) respectively. \( L_z \) and \( w_2 \) \((i = 1, 2)\) are the width and wall thickness of stripes. The spin profile of skyrmions in a SkX can be well described by \( \Theta(r) = 2\arctan \left( \frac{\tan \left( \frac{r}{w_1} \right)}{\tan \left( \frac{L_z}{2w_2} \right)} \right) \) along a line connecting the centers of two neighbouring skyrmions, where \( r \) labels the points on the line with \( r = 0 \) being the center of a chosen skyrmion. \( R \) and \( L \) are respectively the skyrmion size (the radius of \( m_2 = 0 \) contour) and the lattice constant. \( L \) relates to skyrmion number density \( n \) of a SkX in a triangular lattice as \( L \approx 1.07/\sqrt{n} \). Figure 2(a) demonstrates the excellence of this approximate spin profile for thosestripe skyrmions labelled by \( \Theta \) in Figs. 1(a3-c3) (with model parameters of sample A in Tab. 1). The \( y \)-axis is \( m_x \) and \( x = 0 \) is the stripe center where \( m_x = -1 \). Symbols are numerical data and the solid curve is the fit of \( \cos \Theta(x) = -\frac{\sinh(L_z/2w_1) - \sinh(|x|/w_1)}{\sinh(L_z/2w_1) + \sinh(|x|/w_1)} \) for \( -L_z/2 < x < L_z/2 \) and \( \cos \Theta(x) = -\frac{\sinh(L_z/2w_1) - \sinh(|x - L_z/2 - L_z/2|/w_1)}{\sinh(L_z/2w_1) + \sinh(|x - L_z/2 - L_z/2|/w_1)} \) for \( L_z/2 < x < L_z/2 + L_2 \) with \( L_z = L_2 = 8.1 \) nm and \( w_1 = w_2 = 2.7 \) nm. All data from different stripes fall on the same curve and demonstrate that stripes, building blocks of the structure, are identical. Figure 2(b) shows the nice fit of numerical data of all stripes under a magnetic field of 0.4T to the approximate spin profile with \( L_z = 6.05 \) nm, \( L_2 = 12.10 \) nm, and \( w_1 = w_2 = 2.63 \) nm. Figure 1(c) shows that spins along a line connecting the centers of two neighbouring skyrmions fall on our proposed profile with \( L = 16.88 \) nm, and \( R = 4.35 \) nm. The spin profile has been used to find stripe width formula \( L = f(\kappa)A/D \). The value of \( f(\kappa) \) is \( 2\pi \) when \( \kappa \gg 1 \).

3.2 Stable/metastable spin structures defined by \( \kappa \) and \( \kappa' = \frac{g(\kappa)}{\mu_0 H} \)

We have analytically proved in the previous section that stable/metastable structures are fully determined by \( \kappa \) and \( \kappa' \) out of five model parameters, and \( A/D \) defines the length scale. In another word, the spin structures of the same \( \kappa \) and \( \kappa' \) are scale invariant. To verify the results, we use samples B and C listed in Table 1 to simulate metastable structures of 10 skyrmions in a film of 600 nm x 600 nm x 1 nm as what was done in Fig. 1. Both sample B and sample C have the same \( \kappa = 10 \) and \( \kappa' = 22 \) although their \( A \) and \( D \) differ by more than ten times. The stable structures are similar as shown in Figs. 3(a) for sample B and 3(b) for sample C. To further verify that the two structures are scaled by \( A/D \), the spin profiles along the red and blue lines in Figs. 3(a) and (b) are presented in Fig. 3(c). When we scale the x-axis coordinate by \( A/D \) for sample B (the red curve and bottom x-axis) and sample C (the blue curve and top x-axis), two curves are overlapped that verifies our theoretical predictions.

3.3 The role of magnetic field in SkX formation

We compute below the skyrmion-number-dependence of energy for various magnetic field in order to understand the role of a field in SkX formation. We limit this study to the Q's far below its maximal value of more than 500. Physics around the maximal Q is very interesting by itself, but not our concern here, and it will be investigated in the future. Q nucleation domains of 5 nm in diameter each arranged in a square lattice is used as the initial configuration to generate a stable structure of Q skyrmions with energy E. The left panel of Fig. 4(a) is the Q-dependence of E from MuMax3 simulations for \( \mu_0 H = 0, 0.1, 0.2, 0.3, 0.39, 0.5, 0.6 \) and 0.7T denoted by \( 1-7 \). At zero field, E increases monotonically with skyrmion number Q. States with few skyrmions or low skyrmion density are preferred. Since long stripe skyrmions have more way to deform than circular skyrmions, the entropy of a helical phase is larger than a SkX (see numerical evidences according to the approach in Ref[22] below). Thus, helical states should always be the thermal equilibrium phase below the Curie temperature, and a SkX can be a metastable state at most. E of helical states at zero field is not very sensitive to Q. Thus the thermal equilibrium helical states can have different number of irregular skyrmions with many different forms or morphologies. This understanding agrees with experimental facts of rich stripe morphologies[23]. Things are different when a magnetic field is applied. Firstly, E of fixed Q increases with \( H \). Secondly, E is minimal at \( Q_m \) for a fixed field below a critical value. \( Q_m \) first increases with \( H \) up to an optimal field of around \( \mu_0 H = 0.3 \)T in our case and then decreases with \( H \). Above \( \mu_0 H > 0.7 \)T (the brown stars), positive E means that ferromagnetic state of \( m_x = 1 \) has a lower energy of E = 0. Thus, \( m_x = 1 \) is the thermal equilibrium state below the Curie temperature when \( \mu_0 H > 0.7 \)T. \( Q_m \), at the optimal field of \( \mu_0 H = 0.3 \)T, can be as large as more than 141 or a skyrmion density more than 3500/µm² at which two nearby skyrmions are in contact. All skyrmions are compressed into circular objects and form a SkX. Strictly speaking, they are not circular, as evident from our simulations. Our results agree qualitatively with experiments[16][23][33][54].

To substantiate our claim that there is a maximal skyrmion density for a given film, we consider a 200 nm x 200 nm x 8 nm film for sample A. To find the approximate maximal number of skyrmions that can maintain its metastability, we simply add more 2 nm-domains in square lattices as the initial configurations. The system always settles to a metastable state in which the number of skyrmions is the same as the number of initial nucleation domains as long as the the number is less than 500. If the number is bigger than 700, the final number of skyrmions in stable states would be less than the initial number of nucleation domains. Occasionally, we obtain metastable states containing about 700 skyrmions in a triangular lattice, corresponding to skyrmion density of 17,500/µm². Figure 5 shows SkX of 567 skyrmions when the initial configuration contains 625 nucleation domains of 2 nm radius each arranged in a square lattice. The details of how the final metastable states depends on the initial number of nucleation domains and their arrangement, as well as the boundary conditions, deserves a further study.
Fig. 2 The red and blue points in (a), (b) and (c) are the $z$-component of magnetization along the red lines in Figs. 1(a1), (c1) and (c3). The green lines are the fit to our approximate profile. The cyan arrows indicate the widths of stripes of $m_z < 0$ or $m_z > 0$ and skyrmion diameter.

Fig. 3 Metastable structures of 10 skyrmions in a 600 nm × 600 nm × 1 nm film of $\kappa = 10$ and $\kappa' = 22$ for samples B (a) and C (b) in Table 1. (c) Spin profiles along the red and blue lines in (a) and (b). The x-axis is scaled by $A/D$. The red curve and the bottom x-axis are for sample B while the blue curve and the top x-axis are for sample C.

Fig. 4 Skyrmion-number dependence of energy at various magnetic fields of $\mu_0 H = 0$ T (the squares and the black curve), 0.1 T (the circles and the red curve), 0.2 T (the up-triangles and the blue curve), 0.3 T (the down-triangles and the green curve), 0.39 T (the diamonds and the violet curve), 0.5 T (left-triangles and the yellow curve), 0.6 T (right-triangles and the cyan curve), and 0.7 T (the stars and the brown curve) (the left panel). The film size is the same as in Fig. 1. Arrows denote the $Q_m$ for $\mu_0 H = 0$, 0.1, 0.2, 0.3, 0.39, 0.5 and 0.6 T, respectively. Corresponding structures are shown in the right panel. The structures varies from a highly ramified stripe of $Q_m = 1$ for $\mu_0 H = 0$ T, to the mixture of helical order and SkX of $Q_m = 31$ for $\mu_0 H = 0.2$ T, and to beautiful SkX of $Q_m = 141$ for $\mu_0 H = 0.3$ T. Equally nice SkX for $\mu_0 H = 0.5$, 0.6 T with a smaller $Q_m$ demonstrates importance of a field in SkX formation.
3.4 Condensed skyrmion states in various thermodynamic process

To substantiate our assertion that topology prevents a metastable helical state from transforming into a thermal equilibrium SkX state at $\mu_0H = 0.3T$ and far below the Curie temperature, we study stochastic LLG at a finite temperature starting from the structure of Fig. 1(c1) with 10 skyrmions. We run the MuMax3 at 20K and 29K, respectively below and near the Curie temperature of $T_c = 33K$. Figures 6(a) and (b) show the structures after 30ns evolution. Thermal fluctuations at 20K are not strong enough to neither create enough nucleation centers nor cut a short stripe into two that are the process of destroying the conservation of skyrmion number, at least within tens of nanoseconds. The system is still in a helical state with $Q = 11$ skyrmions after 30 nanoseconds, one more than the initial value (see Movie a in the Supporting Information). However, at 29K, a helical state transforms to the thermal equilibrium SkX state with $Q = 132$ skyrmions through cutting stripes into smaller pieces and creating nucleation centers to generate more skyrmions thermodynamically (see Movie b in the Supporting Information). For a comparison, we have also started from the SkX shown in Fig. 1(c3) with 150 skyrmions at $\mu_0H = 0.3T$. The SkX under 29K becomes another SkX with $Q = 136$ skyrmions as shown in Fig. 6(c) after 10ns evolution (see Movie c in the Supporting Information), 14 less than the starting value. This is expected because the average skyrmion number at the thermal equilibrium state should be smaller than $Q_m = 141$ according to the energy curve of $\mu_0H = 0.3T$ in Fig. 4.

We demonstrate below that a SkX at zero field is not a thermal equilibrium state by showing the disappearance of a SkX in both zero field cooling and warming. Starting from the SkX in Fig. 1(c3) and gradually increasing (decreasing) the temperature from 0K (30K) to 30K (0K) at sweep rate of 1K/ns at $\mu_0H = 0T$ (see Movies d and e in the Supporting Information), final structures shown in Fig. 6(d) (zero field cooling) and (e) (zero field warming) are helical state consisting of stripe skyrmions. In contrast, field cooling at the optimal field of $\mu_0H = 0.3T$ from 30K to 0K at same sweep rate does not change the nature of the SkX. This is consistent with our assertion that SkXs are the thermal equilibrium states at $\mu_0H = 0.3T$ below the Curie temperature.

The nucleation centers can also be thermally generated near the Curie temperature such that skyrmions can develop from these thermally generated nucleation centers, rather than from artificially created nucleation domains. To substantiate this claim, we carried out a MuMax3 simulation at 29K under the perpendicular field of $\mu_0H = 0.3T$, Fig. 6(f) is a snapshot of spin structure of the thermal equilibrium state for the same film size and with the same model parameters as those for Figs. 6(a-e). A SkX with 133 skyrmions is observed. The birth of these skyrmions and how the SkX is formed can be seen from Movie f in the Supporting Information.

Small energy gain or loss from the transition between two states of different $Q$‘s makes such a transformation difficult because of the conservation of skyrmion number under continuous spin structure deformation and entanglements among stripes. This study shows that, similar to liquid drop formation, new skyrmions can be generated only from nucleation centers or by splitting a stripe skyrmion into two. These process require external energy sources such as the thermal bath and result in topological protection and energy barrier between states of different $Q$‘s. Although the energy of $Q = 141$ SkX has the lowest energy at $\mu_0H = 0.3T$ at zero temperature (Fig. 4), an initial state with a few stripe skyrmions would not resume its lowest energy state at a low temperature (Fig. 6(f)) within the simulation time. This demonstrates the multi-metastable states of various $Q$ and topological protection to prevent SkXs and helical states from relaxing to the thermal equilibrium phases. People have studied the thermal effects on stripes and SkXs, but early studies did not reveal the role of $\kappa$ and magnetic field and cannot come up with a unified picture rich observations. The present new understanding can perfectly explain those fascinating appearance and disappearance of SkX and helical states along different thermodynamic paths. For example, the disappearance of a SkX in zero field warming and high field warming is because helical state is the thermal equilibrium phase. At a high enough temperature below the Curie temperature, thermal fluctuations can spontaneously generate enough nucleation centers such that the system can change its skyrmion number and reach its thermal equilibrium phase of either helical state of low skyrmion density or SkX state of high skyrmion density.

3.5 $\kappa$ and SkX formation

All results presented so far are for relative large $\kappa > 8$. It is not clear how the results change with other $\kappa$‘s. It should be emphasized that results above are based on the assumptions that all model parameters do not change with the temperature. Ob-
Fig. 6 (a, b) Structures after 30 ns evolution at $\mu_0H = 0.3$ T, and $T = 20$K (a) and 29 K (b), starting from the initial configuration of Fig. 1(c1). (c) Structures after 10 ns evolution at $\mu_0H = 0.3$ T and $T = 29$ K, starting from the initial configuration of Fig. 1(c3). (d, e) Structures after 30 ns zero field cooling (d) and warming (e), starting from the SkX in Fig. 1(c3). (f) A SkX of 133 skyrmions at $\mu_0H = 0.3$ T and 29 K comes from the thermally generated nucleation centers.
viously, this assumption does not apply to real materials. Exchange stiffness $A$ and magnetic anisotropy $K$ depend on temperature, and behave as a power law of $M(T)$ \cite{51} For example, it is known magnetic anisotropy in many materials can change appreciably with temperature. Magnetic anisotropy of 1.2 nm CoFeB film could reduce by 50% as temperature increases from 300K to 400K \cite{52}. More importantly, it is known experimentally \cite{53} that SkXs are not stable at low temperature in almost all systems so far. Thus, a proper explanation of metastability of SkXs at low temperatures and at the optimal magnetic field is required. $\kappa = 1$ separates condensed skyrmion states from isolated skyrmions \cite{54}. In general, decreases as the temperature is lowered since both $A$ and $K$ increases as the temperature decreases. Thus, it is interesting to find out how results in Fig. 7 are modified when $\kappa = 8.3$ for sample A becomes not too far from $\kappa = 1$ in order to mimic the effect of lowering the temperature far from the Curie temperature. We repeat the same calculations as those in Fig. 4 by changing crystalline magnetic anisotropy in our model from 0.036 MJ/m$^3$ to 0.165 MJ/m$^3$, 0.131 MJ/m$^3$, 0.098 MJ/m$^3$, 0.064 MJ/m$^3$, and 0.030 MJ/m$^3$ without changing other parameters. This corresponds to change $\kappa$ from 8.3 for Fig. 4 to 1.11, 1.42, 2, 3.3, and 10. The results are shown in Fig. 7. It is interesting to notice $Q_m \approx 0$ when $\kappa = 1.11, 1.42$ such that this phase is thermal equilibrium state ($E > 0$ means ferromagnetic state more stable). $Q_m \approx 132, 142$ and the optimal field is $\mu_0 H = 0.3 T$ for $\kappa = 3.3, 10$, respectively. $Q_m \approx 92$ at the optimal field of $\mu_0 H = 0.15 T$ for $\kappa = 2$, and the corresponding spin texture is a mixture of stripe skyrmions and circular skyrmions as shown in the insets of the figure. Our numerical simulations on two very different sets of model parameters suggest that $Q_m$ will not be large enough to support an thermal equilibrium SkX state whenever $\kappa < 1.6$.

We see that $Q_m$ is not large enough to form a SkX even at the optimal field when $\kappa < 1.6$. To see that this is in general true, we vary $\kappa$ by increasing the magneto-crystalline anisotropy for both samples B and C in Table 1 such that $\kappa$ changes from 10 to 3.33, 2, 1.42 and 1.11. The film size of samples B and C is 600 nm x 600 nm x 1 nm. Figure 8 shows $E(Q)$ for various $\mu_0 H$ and for $\kappa = 10$ (a), 3.33 (b), 2. (c), 1.42 (d), 1.11 (e). The red curves are for sample B while the blue ones are for sample C. Clearly, $Q_m$ is small and the helical state has the lowest energy for both sets of parameters differing by more than ten times whenever $\kappa < 1.6$.

In summary, general decreases of $\kappa$ may explain why SkXs in most systems become metastable at low temperatures. Material parameters vary also with film thickness. This may also explain why the SkX formation and SkX stability are very sensitive to the film thickness. This conjecture needs more detail studies.

### 3.6 Entropy difference of SkXs and stripe states at a given temperature

In an isothermal process, the Maxwell relation \( \frac{\partial S}{\partial H} \Big|_{T} = \frac{\partial M}{\partial T} \Big|_{H} \) provides a way of calculating the entropy difference at two different fields for a given temperature from the temperature dependence of magnetization \cite{55}.

\[
\Delta S(T, H) \equiv S(T, H) - S(T, 0) = \int_{0}^{H} \left( \frac{\partial M}{\partial T} \right)_{H} dH. \quad (7)
\]

$\Delta S(T, H_m)$ measures the entropy change from a stripe state at zero field to a SkX at optimum field $H_m$ when $T \approx T_c$. Differ from experiments where the noises are bigger than the signal $\Delta S(T, H_m)$, computations do not suffer from this problem such that one can reliably compute the difference between a SkX entropy and a stripe-state entropy at a fixed temperature. We carried out MuMax3 simulations for $M(T, H)$ of sample A at $T = 28 K$ for various $\mu_0 H$ ranging from 0T to 0.3T. Sample size is 200nm x 200nm x 8 nm as before. Figures 9(a-b) are $M(T, H)$ (a) and $\partial M/\partial T|_{H} < 0$ (b) for various $H$. $M(T < T_c, 0) \approx 0$ as shown in Fig. 9(a) because of our helimagnet model. This is why $\Delta M(\partial T)|_{H=0}$ fluctuates around zero as shown by the red dots in Fig. 9(b) and the one-$\sigma$ region is bounded by two red dash lines. One can confidently conclude that $\Delta M(\partial T)|_{H} < 0$ for $T > 0.1T$ (the yellow dots and curves) at which a stripe state has the lowest energy [see Fig. 4(a)] while one may not be so sure of the sign of $\Delta (\partial M/\partial T)$ in experiments. The simulation results clearly support the claim that SkX entropy is smaller than a stripe-phase entropy.

### 3.7 Insensitivity of spin structures to DMI-types

There are two types of Dzyaloshinskii–Moriya interaction (DMI), namely interfacial DMI and bulk DMI. We have mainly presented results for the interfacial DMI so far. Although the spin orientations in skyrmion walls depends on the type of DMI's, the location of energy minimum in energy-$Q$ curves for a fixed $\mu_0 H$, as well as the thermodynamic properties of the system, are not sensitive to DMI type. When the interfacial DMI is replaced by the bulk DMI for those sets of parameters used above, the results are essentially the same except that the Néel-type stripes (skyrmions) for the interfacial DMI change to the Bloch-type stripes (skyrmions).

To substantiate this claim, we use bulk DMI and repeat those simulations for Figs. 1, 4, and 6. Figure 10 shows the metastable structures with 10 (a1-c1), 50 (a2-c2) and 150 (a3-c3) skyrmions under magnetic field of 0T (a1-a3), 0.1T (b1-b3) and 0.4T (c1-c3). Similar to Fig. 1 the film is in a helical state consisting of ramified stripe skyrmions with a small skyrmion number, and a SkX in a triangular lattice with a large skyrmion number. $Q_m$ are also the same as those in Fig. 4 at various magnetic field, although the energy curves are slightly different as shown in Fig. 11. To further prove that states with $Q_m$ skyrmions are thermal equilibrium states, Figs. 12(a) and 12(b) show the spin structure with 11 and 134 skyrmions after 30ns evolution starting from Fig. 19(c1) at 20K and 33K under magnetic field of 0.3T. Figure 12(c) shows the structure with 132 skyrmions after 10ns evolution starting from Fig. 12(c3) at 33K under a magnetic field of 0.3T. Figures 12(d) and 12(e) show the spin structure with 12 and 2 skyrmions after 33ns zero-field warming and zero-field cooling, respectively, starting from 10(c3). The temperature change rates are the same as those in the main text. Figure 12(f) shows the spin structure with 133 skyrmions after 30ns evolution starting from ferromagnetic state at 33K. Clearly, the formation and disappearance of
Fig. 7 Skyrmion-number dependence of energy at various magnetic fields for $\kappa = 1.11$ (a), 1.42 (b), 2. (c), 3.33 (d) and 10 (e). Corresponding spin textures at $Q_m$ are shown in the insets. All other model parameters are the same as those for Fig. 2.

Fig. 8 $E(Q)$ for sample B (red curves and axes) and sample C (blue curves and axes) for various $\mu_B H$ (given by $\frac{\text{8} \mu_B A M_s}{4 \pi D^2}$) and for $\kappa = 10$ (a), 3.33 (b), 2. (c), 1.45 (d), and 1.11 (e). $A$, $D$ and $M_s$ are fixed as given in Table 1. Corresponding spin textures at $Q_m$ are shown in the insets.
Fig. 9 (a) Temperature dependence of time-averaged $z$-component of magnetization $M_z$ at fixed magnetic fields, ranging from 0 mT to 300 mT with a field interval of 25 mT. Every data point is 2 ns-averaged $z$-component of magnetization in the thermal equilibrium state at a fixed temperature and a fixed magnetic field. Error bars are the standard deviations of magnetization over the time periods. (b) Solid curves are temperature derivatives of magnetization $\partial M_z / \partial T$ at $\mu_0 H = 0, 100, 200,$ and $300$ mT. Dash curves bound 1-sigma regions. Inset shows $\partial M_z / \partial T$ for all field strength in (a). (c) Density plot of entropy change $\Delta S$ from a stripy state at zero field to state at a finite field at fixed temperatures in the $T$-$H$ plane. Clearly, a SKX at $T = 28$ K and $\mu_0 H = 300$ mT has lower entropy than that of a stripy state at $T = 28$ K and $\mu_0 H = 0$ mT.
SkXs are the same as Fig. 6 but the only difference is the skyrmion walls change from a Neel type to a Bloch type.

4 Conclusion and perspectives

In conclusion, both helical states and SkXs are the collections of skyrmions. Stripes, not disks, are the natural shapes of skyrmions when their formation energy is negative. The distinct morphologies of helical and SkX states come from the skyrmion density. Skyrmions become circular objects and in a triangles lattice due to the compression and strong repulsion among highly packed skyrmions. Unexpectedly, there are enormous number of metastable states with an arbitrary skyrmion density. These metastable states result in the thermodynamic path dependence of an actual state that a film is in. The energy of these states depends on the skyrmion number and the magnetic field. The role of a magnetic field in SkX formation is to create the lowest energy state being a skyrmion condensate with a high skyrmion density for \( \kappa \) above a critical value. Below the critical value, the lowest energy state is still a collection of condensed stripes skyrmions even in the optimal field. The possible reason for metastability of SkXs in most chiral magnetic film at low temperature is due to increase of exchange stiffness constant and magnetic anisotropy such that \( \kappa \) is below the critical value at low temperature. It is showed that our findings are robust to parameter variations as long as \( \kappa \) is given. Whether the chiral magnets have the interfacial DMI or bulk DMI does not change the physics. Of course, the stripes of circular skyrmions will change from the Neel type to the Bloch type. We have also numerically shown that the SkX entropy is lower than a collection of stripes. In order to realize truly stable SkXs at extremely low temperature, one may use those chiral magnets whose \( \kappa \) is larger than the critical value in the required energy range. Our findings have profound implications in skyrmion-based applications.

Looking forward, several issues require further studies. According to the current findings, the criteria for SKX formation in a chiral magnetic film are \( \kappa > 2 \) and a proper perpendicular magnetic field. Although all known materials support stable SkXs only in a small pocket in the field-temperature plane near the Curie temperature to date while SkXs are metastable in low temperatures, it should be interesting to search materials that support stable SkXs in low temperatures and metastable near the Curie temperature. In fact, there is no reason why one cannot have a film that supports stable SkX in all temperatures below the Curie temperature under a proper field. The physics around the maximal skyrmion density has not been revealed yet in this study. The critical \( \kappa \) is a purely numerical fact based on three sets of very different materials parameters. No clear understanding of its value and related physics has been obtained here. In fact, even why a chiral magnetic film prefers a state with a finite skyrmion density in a magnetic field prefer is not clear. It again is a purely numerical observation at the moment. Thus, we are still in the infant stage of a true understanding of SkX formation.

Conflicts of interest

There are no conflicts of interest to declare.

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Fig. 10 Metastable structures of 10 (a1-c1), 50 (a2-c2) and 150 (a3-c3) skyrmions in a 200nm × 200nm × 8nm film with the bulk DMI at zero temperature for $\mu_0H = 0$ T (a1-a3), 0.1 T (b1-b3) and 0.4 T (c1-c3), respectively. (d1-d3) show how $Q$ (the left y-axis and the red curves) and $E$ (the right y-axis and the blue curves) of 10 (the dash lines), 50 (the solid lines), and 150 (the dot-dash lines) skyrmions vary with time (in the logarithmic scale) for $\mu_0H = 0$ T (d1), 0.1 T (d2) and 0.4 T (d3). The insets of (a1), (c1), (c3) show the spin profiles of stripe and circular skyrmions. The initial configurations are 10, 50 and 150 nucleation domains arranged in a square lattice. Initially, each domain is 5nm in diameter and has zero skyrmion number.

Fig. 11 Skyrmion-number dependence of energy for various magnetic fields of $\mu_0H = 0$ T (the squares and the black curve), 0.1 T (the circles and the red curve), 0.2 T (the up-triangles and the blue curve), 0.3 T (the down-triangles and the green curve), 0.39 T (the diamonds and the violet curve), 0.5 T (the left-triangles and the yellow curve), 0.6 T (the right-triangles and the cyan curve), and 0.7 T (the stars and the brown curve) (the left panel). The film size is the same as that in Fig. 10. Arrows denote the $Q_m$ for $\mu_0H = 0$, 0.1, 0.2, 0.3, 0.39, 0.5 and 0.6 T, respectively. Corresponding structures are shown in the right panel. The structures varies from highly ramified stripes of $Q_m = 1$ for $\mu_0H = 0$ T, to the mixture of helical order and SkX of $Q_m = 31$ for $\mu_0H = 0.2$ T, and to a beautiful SkX of $Q_m = 141$ for $\mu_0H = 0.3$ T. Equally nice SkX for $\mu_0H = 0.5$, 0.6 T with a smaller $Q_m$ demonstrates importance of a field in SkX formation.
Fig. 12 (a, b) Structures after 30ns evolution at $\mu_0 H = 0.3 \, \text{T}$, and $T = 20 \, \text{K}$ (a) and 33K (b), starting from the initial configuration of Fig. 10(c1). (c) Structures after 10ns evolution at $\mu_0 H = 0.3 \, \text{T}$ and $T = 33 \, \text{K}$, starting from the initial configuration of Fig. 10(c3). (d,e) Structures after 33ns zero field warming (d) and cooling (e), starting from the SkX in Fig. 10(c3). (f) A SkX of 133 skyrmions at $\mu_0 H = 0.3 \, \text{T}$ and 33K comes from the thermally generated nucleation centers.
