Chiral solitons in monoaxial chiral magnets in tilted magnetic field

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We show that the stability (existence/absence) and interaction (repulsion/attraction) of chiral solitons in monoaxial chiral magnets can be varied by tilting the direction of magnetic field. We thereby, elucidate that the condensation of attractive chiral solitons causes the discontinuous phase transition predicted by a mean field calculation. Furthermore we theoretically demonstrate that the metastable field-polarized-state destabilizes through the surface instability, which is equivalent to the vanishing surface barrier for penetration of the solitons. We experimentally measure the magnetoresistance (MR) of micrometer-sized samples in the tilted fields in demagnetization-free configuration. We corroborate the scenario that hysteresis in MR is a sign for existence of the solitons, through agreement between our theory and experiments.

Introduction. Dzyaloshinskii–Moriya interactions (DMIs) can exist in non-centrosymmetric magnets, where the competition between DMI and exchange interaction induces modulated spin structures such as conical, cycloidal and magnetic skyrmion states. Among those states, noteworthy are magnetic skyrmion lattice (SkL) in cubic chiral magnets and chiral soliton lattice (CSL) in monoaxial chiral magnets; those two states consist of topological objects: single skyrmion in SkL and single discommensuration (called chiral soliton in this paper) in CSL, respectively. Topological stability of those objects allows us to regard them as emergent particles. Their stability and interaction properties can be varied by elevating temperatures. This controllability gives them an advantage in devise application in future spintronics. It is thus important to find more efficient way to control the physical characters of skyrmions and chiral solitons. In this paper, we show that tilting of the direction of magnetic field can change the interacting properties between repulsion and attraction, and stability/instability of chiral solitons in monoaxial chiral helimagnets, without utilizing temperature effects. We also show that interaction properties and stability/instability of chiral soliton account for the structure of the phase diagram at zero temperature found in an early mean field theory. Further we conduct magnetoresistance (MR) experiments for micrometer-sized samples of Cr\(_{1/3}\)NbS\(_2\) in demagnetization-free configuration. We corroborate our theory on stability/instability of chiral solitons, through agreement between our theory and experiments.

Monoaxial chiral magnets. \(\text{Cr}_{1/3}\text{NbS}_2\) is a monoaxial chiral magnet. It shows a helical state with its pitch of 48 nm along the c-axis, which we call the helical axis, in the absence of magnetic field. The helical structure consisting of spins rotating in the \(ab\)-plane is robust because of the strong hard-axis anisotropy along the helical axis. The magnetic field perpendicular to the helical axis induces an ideal chiral soliton lattice, and leads to a continuous phase transition (CPT) to the uniform state. Properties of \(\text{Cr}_{1/3}\text{NbS}_2\) in equilibrium and metastable states have been quantitatively explained by Refs. and respectively, with use of the chiral sine-Gordon model. Thus \(\text{Cr}_{1/3}\text{NbS}_2\) is regarded as a model material of monoaxial chiral magnets. The field parallel to the helical axis induces a CPT from chiral conical state to the uniform state. Recently, Laliena et al. have found three types of field-induced phase transitions, which depend on the direction of magnetic field in monoaxial chiral magnets: CPT for fields with angle \(\theta_H\) (with respect to the \(ab\)-plane) larger than 88.5°, discontinuous phase transition (DPT) for \(81.5° \leq \theta_H \leq 88.5°\), another CPT for \(0 \leq \theta_H \leq 81.5°\), and two multicritical points. In a subsequent paper, they identified the former CPT as the instability-type and the latter CPT as the nucleation-type, following de Gennes’s classification. The period of modulation in the ordered phase diverges in nucleation-
We start with the following energy functional for the classical spins defined on a one dimensional lattice along the helical axis at zero temperature:

\[
E(\{\vec{M}_l\}) = -\sum_l \left[ J_l \vec{M}_l \cdot \vec{M}_{l+1} + D \left( \vec{M}_l \times \vec{M}_{l+1} \right)^2 - \frac{K}{2} \left( \vec{M}_l^2 \right)^2 + H_{\text{ex}} \cdot \vec{M}_l \right].
\]  

(1)

The local magnetic moment at site \( l \) on the chain is given by \( \vec{M}_l \), and the magnitude of each moment, \(|\vec{M}_l|^2\), is 1. The first and second terms are the Heisenberg exchange, and Dzyaloshinskii–Moriya interactions on the nearest neighbor pairs, respectively. The third term stands for the hard axis anisotropy for positive \( K \). The last term is the Zeeman energy due to tilted magnetic field, \( H_{\text{ex}} \), which has \( x \)- and \( z \)-components. As realistic parameters, we set \( D = 0.16J_|| \) and \( K = 5.68H_d \) with \( H_d = 2(J_||^2 + D^2)^{1/2} - J_\parallel \). The stationary condition is given by \( \vec{M}_l \times \vec{H}_l^{\text{eff}} = 0 \) with

\[
\vec{M}_l = \vec{H}_l^{\text{eff}} = \vec{H}_l^{1/2}/|\vec{H}_l^{1/2}|, 
\]

(2)

\[
\vec{H}_l^{\text{eff}} = J_1(\vec{M}_{l-1} + \vec{M}_{l+1}) + D\hat{z} \times (\vec{M}_{l-1} - \vec{M}_{l+1}) - K\vec{M}_l^2 \hat{z} + H_{\text{ex}}. 
\]

(3)

Properties of chiral solitons. We classify the region in the \( H_{\text{ex}}^2-H_{\text{ex}}^z \) phase diagram, according to existence/absence and interaction properties of chiral soliton, following the method used in Ref. [10]. Let us consider an isolated soliton with its center at \( l = 0 \) and assume the following asymptotic form of the magnetic moment at \( l \gg 1 \):

\[
\vec{M}_l = \vec{M}_u + \text{Re}(\vec{A}\exp(-\kappa x_l)) \text{ with } x_l = la,
\]

(4)

where \( \vec{M}_u \) is the uniform solution without boundaries and \( a \) is a lattice constant. A positive real part of \( \kappa \) describes the soliton tail and corresponds to the inverse of the soliton size. On the other hand, a pure imaginary (PI) \( \kappa = iq \), describes a distorted conical order with a fundamental wave number \( q \) rather than an isolated soliton. In this case, the form (4) is available for all \( l \) when \( \vec{A} \) is regarded as vanishingly small.

Linearization of Eqs. (2) and (3) with respect to the second term of Eq. (1) leads to the linear coupled-equation of \( \vec{A} \) with condition \( \vec{M}_u \cdot \vec{A} = 0 \) deduced from the normalization, and we obtain the quadratic equation in \( \cosh(\kappa a) \) through the condition for the existence of a non-trivial solution of \( \vec{A} \). The values of \( \kappa a \) depend on \( H_{\text{ex}}^2 \) and \( H_{\text{ex}}^z \), and are classified into three cases through the discriminant: (i) real, (ii) complex, and (iii) PI. On the basis of the type of \( \kappa a \), we draw the bold red line ("LA line") in Fig. 2 which separates the phase diagram into the three regions (i), (ii), and (iii). A necessary condition for the existence of an isolated soliton is that \( \kappa a \) belongs to (i) or (ii), and actually there are instability lines of an isolated soliton in this region, which give the sufficient condition.

Figure 2. Phase diagram in the tilted magnetic field using realistic parameters shown in the text. The solid red line is obtained using the linear analysis (LA line), which divides the phase diagram into three regions (i)–(iii). There are, correspondingly, three kinds of phase transition: nucleation-type CPT, the DPT, and the instability-type CPT denoted by the solid purple line, the dotted light-blue line, and the dashed pink line, respectively. Two yellow squares “M” and “T” represent multicritical and tricritical points respectively. The low-field (high-field) side of the phase boundary is the ordered (disordered) phase. The phase boundary is obtained by minimizing the energy functional and basically the same as in Ref. [10]. The black solid lines with values of \( \theta_H \equiv \tan^{-1}(H_{\text{ex}}/H_d) \) are guides to see the field angle. Solid circles labeled “\( H_0 \)” and solid triangles labeled “\( H_1 \)”, respectively, stand for the barrier field and the nucleation field defined in the text.
Following Refs. [16,30], we summarize the interaction properties in the asymptotic region. In the region (i), the interaction is repulsive for any inter-soliton distance. On the other hand, in the region (ii), the interaction energy oscillates as a function of the distance and can be attractive for some values of the distance. In Fig. 2 we see that the interaction between solitons changes from repulsive to attractive in increasing $H_{ex}^z$, the parallel component of the magnetic field.

Comparison with the ground state phase diagram. In Fig. 2, we have also drawn the phase boundaries given by the three kinds of phase transitions. We see that the multicritical point $M$ connecting the DPT line to the nucleation-type CPT line is located on the boundary between (i) and (ii). In the region (i), the repulsion leads to a logarithmically diverging period near the transition. This explains the reason the phase transition in (i) is identified as nucleation-type CPT. On the other hand, in the region (ii), the attraction favors the periodic structure of solitons with a finite distance even at the transition point and leads to the DPT.

The instability-type CPT around $(H_{ex}^x,H_{ex}^z) \approx (0,H_{ex}^z)$ can be described as the development of a conical order with distortion owing to finite $H_{ex}^z$, which is equivalent to the existence of vanishingly small $\vec{A}$ in the region (iii). A part of the LA line where (ii) and (iii) meet is the instability-type CPT line. The tricritical point $T$ is located at the point where the LA line deviates from the phase boundary.

Consistency between properties of chiral soliton (the existence/absence and repulsion/attraction) and the types of phase transitions in the ground state phase diagram has two-fold implications: It explains the mechanisms of the phase transitions, and it endorses our arguments on properties of chiral solitons.

Recently, attractively interacting skyrmions in the conical phase, which result from a different mechanism, were theoretically studied [31] and experimentally confirmed by observing their clusters [32]. Attractive interaction between chiral solitons can be confirmed, in a similar way, i.e. by observation of the cluster formation of solitons in the uniform state.

Surface instability, surface barrier and hysteresis. So far we have seen that chiral solitons exist in the wide region of the phase diagram. Next we consider hysteresis observed in experiments for micrometer samples [27,29]. Particularly, the reproducible large jump in decreasing field is discussed in connection with surface instability and surface barrier for penetration of chiral solitons.

First we perform the mode analysis in a way similar to that in Ref. [21]. The detail is written in the supplemental material. Let us consider the field polarized state with surface twist $\{M_{s,l}\}$ as a static configuration $(\vec{M}_{s,l})$. Its structure is schematically shown in Fig. 1(c). The system is defined for $l \geq 0$ with the free boundary condition $\vec{M}_{l=-1} = \vec{0}$, and thereby a twisted structure appears around the boundary. We obtain the excitation spectra from the equation of motion based on the bilinear form of energy $\mathcal{H}$ with respect to the normal modes for $\{M_{s,l}\}$. The spectra are shown for $H_{ex}^x = 5H_d$ in Fig. 3(a). The low energy state appears from the continuum spectra in decreasing field, as shown in Fig. 3(b). This excitation is bound to the surface, leading to the penetration of the soliton. The energy becomes zero at $H_{ex}^x \approx 0.1474H_d$, which is a surface-instability field [33]. Note that such a localized state is not always the destabilizing mode. Near the PI region, the lowest energy excitation leads to an instability of a conical order [42].

Then we confirm that this instability field coincides with the field in which the surface barrier vanishes [29]. Figure 3(c) shows that the energy landscapes of the isolated chiral soliton as a function of the soliton center, $l$, for several values of $H_{ex}^x$ and $H_{ex}^z = 5H_d$. Here the single soliton energy $E_1$ is measured from the uniformly polarized state [43]. This kind of energy landscape has been presented in Refs. [22,44] for a superconducting vortex, and in Refs. [27,30] for a chiral soliton. As is known in Ref. [26], there exist the characteristic local maximum and minimum structures inside and outside the system, respectively, for $H_{ex}^x > H_b^x$. The surface barrier is described by the local maximum [36] while the surface twist is by the spin structure of an isolated soliton at the point of the local minimum [34,41]. They merge at $H_{ex}^x = H_b^x$, i.e., the soliton outside the system for $H_{ex}^x > H_b^x$ comes...
to the surface at $H^\infty_{\text{ex}} = H^\infty_0$, and the surface barrier vanishes. Figure 3 shows that $H^\infty_0$ is consistent with the instability field.

For these values of the tilted field, $\kappa a$ is complex, and correspondingly the interaction between solitons can be attractive in contrast to Refs. [20, 44]. In this case, solitons are possibly attributed to the surface. Inside the system as well as outside, the energy landscape has local minima coming from the oscillation of the soliton profile. Particularly, the local minimum closest to the surface gives the global minimum inside the system. A sufficiently small field step allows a few solitons to penetrate and be bound to local minima near the surface. This state might be observed by local measurements.

We calculate the barrier field, $H_0$, in the region where solitons exist, as shown by blue solid circles in Fig. 2 and directly compare the calculated values of $H_0$ with experimentally observed jump fields below. In the PI region of $\kappa a$, solitons do not exist, and the hysteresis is hardly observed in a magnetization process passing through this region.

Magnetoresistance measurements. For quantitative comparison, we have to take account of the demagnetizing effects, which give difference between the internal and external fields. We, thus, performed MR measurements in the configuration so as to avoid the demagnetizing effects. Dimensions of samples 1 and 2 are (11.25 $\mu m \times 0.7 \mu m \times 17.5 \mu m$), and (8.5 $\mu m \times 0.5 \mu m \times 21 \mu m$), respectively, where the order of the directions is $x \times y \times z (c$-axis). We define the tilted angle of the field $\theta_H = \tan^{-1} \frac{H^\infty_{\text{ex}}}{H^\infty_{\text{sat}}}$. For samples 1 and 2, the field is in the plane of the film for any $\theta_H$, and demagnetizing effects on the field polarized states are small. The data taken from Ref. [29], in which the sample dimension is (0.7 $\mu m \times 10 \mu m \times 17.5 \mu m$) and it has large demagnetizing effects for $\theta_H \sim 0^\circ$, is shown for reference.

We performed two different sequences for sample 1, and label them sample 1 and sample 1’. The robustness of the hysteresis loops is confirmed through the multiple field-sweeps, where one sweep stands for a set of increasing and decreasing field processes. Actually five-time sweeps are done at $\theta_H = 30^\circ$, $60^\circ$, and $80^\circ$ for sample 1, and three-time sweeps are done at $\theta_H = 0^\circ$ for sample 1’, though only one sweep is done in the other cases [46].

There are experimentally important two fields: the saturation field $H_{\text{sat}}$, where the hysteresis of MR closes in increasing field and the jump field $H_{\text{jump}}$, where MR shows the sharp jump in decreasing field [25, 24]. We identify $H_{\text{sat}}$ and $H_{\text{jump}}$ as the theoretically important two fields, $H_0$ and $H_{\text{sat}}$, respectively. Note that we use $H_{\text{sat}}$, which is the nucleation field and defined so that the single soliton energy is zero, instead of $H_c$. For the nucleation-type CPT, $H_{\text{sat}}$ is the same as $H_c$, while for the DPT, $H_{\text{sat}}$ is slightly lower than $H_c$, but the difference is negligible as inferred from Fig. 2.

We compare $H_{\text{jump}}$ with $H_0$ in Fig. 4(a) and do $H_{\text{sat}}$ with $H_{\text{sat}}$ in Fig. 4(b). $H_0$ and $H_{\text{sat}}$ are normalized by 1.8 kOe, which is the thermodynamic critical field at $\theta_H = 0^\circ$ obtained in an experiment [29]. The value of the anisotropy is taken so that the critical field at $\theta_H = 90^\circ$ is 19.5 kOe.

The angle dependences of $H_0$ and $H_{\text{sat}}$ agree well with those of $H_{\text{jump}}$ and $H_{\text{sat}}$, respectively, as shown in Figs. 4(a) and (b), except for the data of Ref. [29], in which disagreement is caused by large demagnetizing effects. This consistency for the whole range of the phase diagram strongly supports the scenario for the clear hysteresis. The hysteresis due to the surface effects does not conflict with the type of phase transitions discussed in the ground state phase diagram. Agreement can be improved by taking into account the demagnetizing effects, but our approach sufficiently explains the physical origin of the characteristic hysteresis as a starting point.

Discussion. Earlier studies [16, 111, 47] have discussed attractive interaction between solitons/skyrmions due to “soft modulus effects” [47], i.e. effects due to spatial variation of modulus of local magnetic moment. This effect becomes important at finite temperatures, although they have not been experimentally confirmed yet. Our study demonstrates that soft modulus effects exist even at zero temperature by tilting magnetic field; Reduction of the in-plane moduli of local magnetic moments can change spin profiles, interaction properties and stability of chiral solitons. At zero temperature, whether soft modulus effects are possible depends on the manifold of topological defects. The soliton is a defect of in-plane components (XY spins) and has an extra direction for softening of skyrmion defects. The soliton is a defect of in-plane components (XY spins) and has an extra direction for softening of skyrmion defects.
sibility to control the physical properties of chiral solitons in chiral magnets.

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file obtained by arranging the single soliton solution to Eqs. 2 and 3 under the periodic boundary condition of a finite-size chain with its center at $l_c \in \mathbb{Z}$. Note that the surface modulation is necessary for the genuine solution when the surface exists and the free boundary condition is imposed.

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Supplemental Materials: Chiral soliton in monoaxial chiral magnets under tilted magnetic field

EXPERIMENTAL DETAIL

Figure S1. (a) Schematic of the specimen and magnetic field configuration. (b)-(e) Magnetoresistance data in increasing and decreasing field processes for the sample 1 at 10 K. All data for five times field cycles are plotted in each panel except for 85 degree.

Bulk single crystals of CrNb₃S₁₆ were grown by chemical vapour transport method as described elsewhere [S1]. Micrometer-sized platelet specimens were cut from the bulk single crystal used in Ref. [S2] by using a focused ion beam (FIB) machine. Gold electrodes were prepared on the specimens for four-terminal resistance measurements by means of electron beam lithography (EBL) and lift-off techniques. The specimen dimensions are given in the main text. The resistance measurements were performed using a four-terminal method with ac current whose amplitude was 1.0 mA and frequency was 137 Hz. Magnetic field direction was rotated in the specimen plane to minimize the contribution of demagnetizing effect as schematically drawn in Fig. S1(a). The angle is defined as 0 degree when \( H \) is perpendicular to the \( c \) axis of the specimen, while 90 degree in the configuration with \( H \) parallel to the \( c \) axis. Figures S1(b) to S1(e) present the magnetoresistance data of the sample 1 at 10 K at 30, 60, 80, and 85 degrees, respectively. The measurements were performed five times except for the data at 85 degree. The magnetic field intervals are 50 Oe for the data taken at 30 and 60 degrees, and 100 Oe for at 80 and 85 degrees.
DETAIL OF MODE ANALYSIS

Formulation

We summarize the eigenequation for normal modes in the presence of the modulated structure as a static solution. We start with the following Hamiltonian of the monoaxial chiral magnets

$$\mathcal{H} = - \sum_j \left[ J_z \hat{M}_j \cdot \hat{M}_{j+z} + D \vec{e}^z \cdot \left( \hat{M}_j \times \hat{M}_{j+z} \right) + \vec{H}_{\text{ex}} \cdot \hat{M}_j - \frac{K}{2} \left( \hat{M}_j \cdot \vec{e}^z \right)^2 \right] + \sum_j \mathcal{H}_{\perp,j}, \quad (S1)$$

$$\mathcal{H}_{\perp,j} = - \sum_{\mu=x,y,z} \left[ J_\mu \hat{M}_j \cdot \hat{M}_{j+\mu} + D_\mu \vec{e}^\mu \cdot \left( \hat{M}_j \times \hat{M}_{j+\mu} \right) \right]. \quad (S2)$$

Interactions between in-plane spins, $J_\mu$ and $D_\mu$ are independent of the direction $\mu = x, y$ in the case of the monoaxial magnet, and we can write them as $J_x = J_y = J_\perp$ and $D_x = D_y = D_\perp$. Here we specify a site on a cubic lattice as $j = j_\perp + t \hat{z} = j \hat{x} + k \hat{y} + l \hat{z}$. Let us consider the modulated structure in $z$-direction given by

$$\hat{M}_{n,j} = \left( \cos \varphi_{s,l} \sin \theta_{s,l}, \sin \varphi_{s,l} \sin \theta_{s,l}, \cos \theta_{s,l} \right), \quad (S3)$$

and new spin coordinate system given by

$$\hat{M}_j = \hat{U}_l \hat{M}_j, \quad \hat{U}_l = \begin{pmatrix} -\sin \varphi_{s,l} & -\cos \varphi_{s,l} \cos \theta_{s,l} & \cos \varphi_{s,l} \sin \theta_{s,l} \\ \cos \varphi_{s,l} & -\sin \varphi_{s,l} \cos \theta_{s,l} & \sin \varphi_{s,l} \sin \theta_{s,l} \\ 0 & \sin \theta_{s,l} & \cos \theta_{s,l} \end{pmatrix}. \quad (S4)$$

Subscripts $s$ denote static. We introduce the unit vectors in the tilde frame as

$$\hat{M}_j = \hat{M}_j \hat{e}^x_j + \hat{M}_j \hat{e}^y_j + \hat{M}_j \hat{e}^z_j, \quad (S5)$$

where

$$\hat{U}_l = (\hat{e}^x_l, \hat{e}^y_l, \hat{e}^z_l), \quad \hat{e}^x \cdot \hat{e}^z = 0, \quad \hat{e}^x \cdot \hat{M}_{n,j} = 1. \quad (S6)$$

Introducing the following Fourier transform:

$$\hat{M}_{j,\mu} = \frac{1}{\sqrt{N_{2d}}} \sum_{k_{\perp}} \hat{M}_{k_{\perp},l} e^{i k_{\perp} \cdot j_{\perp}} \quad (S7)$$

and we write down the Hamiltonian up to second order of $\hat{M}_{j,\mu}$ and $\hat{M}_{j,\nu}$ in the form

$$\mathcal{H} = E(\{\varphi_{s,l}\}, \{\theta_{s,l}\}) + \frac{1}{2} \sum_{k_{\perp},l,\mu,\nu} \sum_{m} \sum_{x,y,z} \hat{M}_{k_{\perp},l}^{\mu} \hat{K}_{m,l}^{\nu} \hat{M}_{k_{\perp},l}^{\nu} \quad (S8)$$

It is obvious that $K$ is hermitian in the sense that $(K_{\mu,l,m}(k_{\perp}))^* = K_{\nu,l,m}(k_{\perp})$. The first order terms of $\hat{M}_{j,\mu}$ and $\hat{M}_{j,\nu}$ vanishes owing to the equilibrium condition of $\varphi_{s,l}$ and $\theta_{s,l}$. Note that $\sum_{j_{\perp}} \hat{M}_{j_{\perp}} \approx \sum_{k_{\perp}} 1 - [\hat{M}_{k_{\perp},l} \hat{M}_{k_{\perp},l} + \hat{M}_{k_{\perp},l} \hat{M}_{k_{\perp},l}] / 2$.

Exchange interaction:

For convenience, we use the notations $\cos \theta_{s,l} = c\theta_{s,l}$, $\sin \theta_{s,l} = s\theta_{s,l}$, and $\varphi_{s,l} - \varphi_{s,l+1} =: \Delta \varphi_{s,l}$. The exchange term is transformed using $\hat{M}$ as $\hat{M}_j \cdot \hat{M}_{j+z} = \sum_{\mu,\nu = x,y,z} \hat{M}_j^{\mu} \vec{e}^\nu_{l+1} \cdot \hat{M}_{j+z}^{\mu}$

$$\hat{e}^\nu_{l} \cdot \hat{e}^\nu_{l+1} = \begin{pmatrix} \cos \Delta \varphi_{s,l} & c\theta_{s,l} s\theta_{s,l+1} \sin \Delta \varphi_{s,l} & -s\theta_{s,l+1} \sin \Delta \varphi_{s,l} \\ -c\theta_{s,l} \sin \Delta \varphi_{s,l} & c\theta_{s,l} c\theta_{s,l+1} \cos \Delta \varphi_{s,l} + s\theta_{s,l} s\theta_{s,l+1} & -c\theta_{s,l} s\theta_{s,l+1} \cos \Delta \varphi_{s,l} + s\theta_{s,l} c\theta_{s,l+1} \\ s\theta_{s,l} \sin \Delta \varphi_{s,l} & -s\theta_{s,l} c\theta_{s,l+1} \cos \Delta \varphi_{s,l} + c\theta_{s,l} s\theta_{s,l+1} & s\theta_{s,l} c\theta_{s,l+1} \cos \Delta \varphi_{s,l} + c\theta_{s,l} c\theta_{s,l+1} \end{pmatrix}. \quad (S9)$$

Dzyaloshinskii–Moriya interaction:

The second term is written as $\vec{e}^z \cdot (\hat{M}_j \times \hat{M}_{j+z}) = \sum_{\mu,\nu = x,y,z} \hat{M}_j^{\nu} (\vec{e}^\mu \times (\vec{e}^\nu_{l} \times \vec{e}^\nu_{l+1})) \cdot \hat{M}_{j+z}^{\mu}$ and we calculate the matrix element as follows:

$$[\vec{e}^\nu_{l} \times \vec{e}^\nu_{l+1}]^z = \begin{pmatrix} -\sin \Delta \varphi_{s,l} & c\theta_{s,l} \sin \Delta \varphi_{s,l} & s\theta_{s,l} \sin \Delta \varphi_{s,l} \\ -\cos \Delta \varphi_{s,l} & c\theta_{s,l} \cos \Delta \varphi_{s,l} - c\theta_{s,l} s\theta_{s,l+1} \sin \Delta \varphi_{s,l} & c\theta_{s,l} s\theta_{s,l+1} \sin \Delta \varphi_{s,l} \\ s\theta_{s,l} \cos \Delta \varphi_{s,l} & s\theta_{s,l} c\theta_{s,l+1} \sin \Delta \varphi_{s,l} - s\theta_{s,l} s\theta_{s,l+1} \sin \Delta \varphi_{s,l} & -s\theta_{s,l} s\theta_{s,l+1} \sin \Delta \varphi_{s,l} \end{pmatrix} \quad (S10)$$
Zeeman coupling:

The third term is given by \( \hat{H}_{ex} \cdot \hat{M}_j = \sum \mu \hat{H}_{ex} \cdot \vec{e}_\mu \hat{M}_{j\mu} \rightarrow \hat{H}_{ex} \cdot \hat{\vec{e}}_l \hat{M}_{j\mu} \). In the final transformation, we retain the term contributing the equilibrium state energy and the second order expansion.

\[
\hat{H}_{ex} \cdot \hat{\vec{e}}_l^z = H_{ex}^z \cos \phi_{s,l} + H_{ex}^z \epsilon \theta_{s,l}.
\]  

(S11)

Anisotropy:

The fourth term is given by \((\hat{M}_j \cdot \hat{\vec{e}}^z)^2 = \sum_{\mu,\nu=x,y,z} \hat{M}_j^\mu (\hat{\vec{e}}_l^\mu \cdot \hat{\vec{e}}^z) (\hat{\vec{e}}_l^\nu \cdot \hat{\vec{e}}^z) \hat{M}_j^\nu\).

\[
(\hat{\vec{e}}_l^\mu \cdot \hat{\vec{e}}^z) (\hat{\vec{e}}_l^\nu \cdot \hat{\vec{e}}^z) = \begin{pmatrix} 0 & 0 & 0 \\ 0 & s^2 \cos \theta_{s,l} & s \sin \phi_{s,l} \cos \theta_{s,l} \\ 0 & s \sin \phi_{s,l} \cos \theta_{s,l} & \cos^2 \theta_{s,l} \end{pmatrix}.
\]  

(S12)

In-plane interactions:

In-plane exchange and DMIs have dependence on the in-plane wave vector. We consider the in-plane DMIs of the form

\[-\sum \sum_{\mu=x,y} D_{\mu} (\hat{M}_j \times \hat{\vec{e}}_l^\mu) \cdot \hat{\vec{e}}_l^\nu.\]  

(S13)

We transform \( \sum_{j} \sum_{\rho=x,y} \sum_{\mu=x,y,z} \hat{M}_j^\mu K_{int,j,j+\rho}^\mu \hat{M}_j^\nu \), and \( K_{int} \) is given by

\[
K_{int,j,j+\rho} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} - 2D_x \begin{pmatrix} 0 & \cos \phi_{s,l} \cos \theta_{s,l} & \cos \phi_{s,l} \sin \theta_{s,l} \\ -\cos \phi_{s,l} \sin \theta_{s,l} & 0 & -\sin \phi_{s,l} \\ -\cos \phi_{s,l} \cos \theta_{s,l} & \sin \phi_{s,l} & 0 \end{pmatrix} \]  

(S14)

\[
K_{int,j,j+\rho} = -2J_y \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} - 2D_x \begin{pmatrix} 0 & \sin \phi_{s,l} \cos \theta_{s,l} & \sin \phi_{s,l} \sin \theta_{s,l} \\ -\sin \phi_{s,l} \sin \theta_{s,l} & 0 & \cos \phi_{s,l} \\ -\sin \phi_{s,l} \cos \theta_{s,l} & -\cos \phi_{s,l} & 0 \end{pmatrix}.
\]  

(S15)

Using \( \hat{M}_j^z \simeq 1 - \sum_{\mu=x,y} (\hat{M}_j^\mu)^2 / 2 \), \( \sum_{j} \sum_{\rho=x,y} \) is reduced to

\[
\sum_{l, k=\pm} \left\{ -(J_x + J_y) + \frac{1}{2} \left( \hat{M}_{-k,l}^x \hat{M}_{k,l}^y - \hat{M}_{-k,l}^y \hat{M}_{k,l}^x \right) \right\} \sum_{\mu=x,y} \left\{ J_\mu (1 - \cos k_{\mu} a) - iD_\mu M_{s,j}^\mu \sin k_{\mu} a \right\} \left( \hat{M}_{k,l}^x \right) \left( \hat{M}_{k,l}^y \right) \left( \hat{M}_{k,l}^z \right).
\]  

(S16)

We summarize the above expressions. \( J = \sqrt{J_x^2 + D^2} \) and \( \tan \alpha = D / J_x \), we obtain the explicit forms of \( E \) and \( K \) as follows:

\[
E(\{\phi_{s,l}\}, \{\theta_{s,l}\}) = -N_{2d} \sum_{l, \mu=x,y} \left\{ \hat{J}(s\phi_{s,l}s\theta_{s,l+1} + \Delta \phi_{s,l} + \alpha) + J_\mu c\theta_{s,l} c\theta_{s,l+1} \\ + \hat{H}_{ex}^x \cos \phi_{s,l}s\theta_{s,l} + \hat{H}_{ex}^z c\theta_{s,l} - \frac{K}{2} c^2 \theta_{s,l} + (J_x + J_y) \right\}.
\]  

(S17)
and

\[
\begin{align*}
\mathcal{K}^x_{l,l+1} &= -\tilde{J} \cos(\Delta \varphi_{s,l} + \alpha) \\
\mathcal{K}^y_{l,l+1} &= -\tilde{J} \theta_{s,l+1} \sin(\Delta \varphi_{s,l} + \alpha) \\
\mathcal{K}^z_{l,l+1} &= +\tilde{J} \theta_{s,l} \sin(\Delta \varphi_{s,l} + \alpha) \\
\mathcal{K}^z_{l,l+1} &= -\tilde{J} \theta_{s,l} \cos(\Delta \varphi_{s,l} + \alpha) - J_{l} s \theta_{s,l+1} s \theta_{s,l+1} \\
\mathcal{K}^z_{l,l+1} &= \tilde{J} s \theta_{s,l+1} \cos(\Delta \varphi_{s,l} + \alpha) + \theta_{s,l-1} \cos(\Delta \varphi_{s,l-1} + \alpha) + J_{l} c \theta_{s,l+1} + c \theta_{s,l-1} \\
&+ H_{ex}^z \cos \varphi_{s,l} s \theta_{s,l} + H_{ox}^z c \theta_{s,l} - K c^2 \theta_{s,l} + 2 \sum_{\mu=x,y} J_{\mu} (1 - \cos k_{\mu} a)
\end{align*}
\]

(S18)

(S19)

(S20)

(S21)

(S22)

(S23)

(S24)

(S25)

Note the relation \((K_{l,m}^\mu(k_{\perp}))^* = K_{l,m}^\mu(k_{\perp} = K_{l,m}^\mu(-k_{\perp})\), and the other components are zero. Our equation of motion is given by \(\frac{d\tilde{M}_{l,m}^{k_{\perp}}}{dt} = -\tilde{M}_l \times \left(-\frac{\partial H}{\partial \tilde{M}_l}\right)\), which now reads

\[
-\omega \left( \tilde{M}_{k_{\perp},l}^{x,m} \right) = \left( \frac{-\partial H}{\partial \tilde{M}_{k_{\perp},l}^{y,m}} \right) \left( \frac{-\partial H}{\partial \tilde{M}_{k_{\perp},l}^{x,m}} \right) \sum_m \left( -K_{l,m}^{yx}(k_{\perp}) K_{l,m}^{zx}(k_{\perp}) - K_{l,m}^{zy}(k_{\perp}) K_{l,m}^{xz}(k_{\perp}) \right) \left( \tilde{M}_{k_{\perp},l}^{x,m} \right).
\]

(S26)

Numerical scheme

We numerically diagonalize Eq. (S23) to obtain the excitation spectra and eigenvectors using a software of CPPla-pack. We consider the sufficiently large finite-size lattice chain, the number of the site in the direction, \(N_z\), is set to 2000 \((l = 0, \cdots, N_z - 1)\). The free boundary condition is given by \(\tilde{M}_{l=1} = \tilde{M}_{l=N_z} = 0\). First we solve the mean field equation to obtain the static profile \(\tilde{M}_{l,t}\) and then investigate the excitation modes on \(\tilde{M}_{l,t}\). In order to exclude the surface twist structure at around \(l = N_z - 1\), we use sites \(l = 0, \cdots, N_z/2 - 1\) for calculation of excitation spectra. In this case, we can approximately deal with a semi-infinite system with boundary at \(l = 0\). The boundary condition for the diagonalization is correspondingly given by \(\tilde{M}_{l=1,N_z/2} = 0\). Note that the condition at \(N_z/2\) gives finite size effects, but the effects on the localized mode are negligible and those on the extended mode are not very important in the following.

Instability modes

We consider the same case of \(D_1 = 0\) as in the main text. In this case, instabilities are caused by a mode uniform in the plane perpendicular to the helical axis (\(z\)-axis), and we set \(k_{\perp} = 0\). We remark that the non-reciprocity appears only when \(D_1 \neq 0\). Spin profiles of excited modes are shown for the basis \(\tilde{e}^\mu_l(\mu = x, y, z)\) in the following.

First we show the spin profile of an excitation mode leading to the surface instability for \(H_{ex}^z/H_d = 5.0\), discussed in the main text. We set \(H_{ex}^z/H_d = 0.1474264\). The spin modulation is localized around the surface, which leads to the penetration of a soliton. Then we see the excitation spectrum when we enter the PI region without crossing the barrier field \(H_b\) in Fig. S3. We set \(H_{ex}^z/H_d = 5.2\). There is also a low energy state separated from the continuum spectra, but the weight of its wave function is away from the surface with distance about the size of the surface twist structure. The static configuration at \(H_{ex}^z/H_d = 0.1370791\) is shown in Fig. S4(a), and the wave function of the lowest excited state is shown in Fig. S4(b). This excited state is an instability mode leading to a distorted conical order. Because there is one low energy branch of the surface instability, we can expect the crossover behavior of its wave function weight between two instabilities in the vicinity of the field \(H_{ex}^z\). The instability is a penetration of a soliton.
Figure S2. (a) Static configurations to calculate the excitation spectra for \((H_{ex}^{x}, H_{ex}^{z})/H_d = (0.1474264, 5.0)\). The spin profile is uniform far from the surface and a surface twist structure appears around the surface. (b) The lowest energy excitation bound to the surface. This mode causes the penetration of a soliton at the surface.

Figure S3. (a) Excitation spectra for \(H_{ex}^{z}/H_d = 5.2\). (b) is a magnified image of (a) near the instability region. Red circles are the spectra for the static configuration given by Fig. S4 (a). There is a low energy mode apart from the continuum spectra as well as for \(H_{ex}^{z}/H_d = 5.0\).

for \(H_{ex}^{z} < H_{ex}^{z^*}\) and a development of a distorted conical order for \(H_{ex}^{z} > H_{ex}^{z^*}\). However it is difficult to access this region because of enormous numerical costs. The instability to a distorted conical order occurs at higher field than the LA line obtained by the linear analysis. In the linear analysis, we assume the uniform state as a static configuration.

In the present case, we consider the surface twist structure, which breaks the translational symmetry, and the conical order nucleates there. This is not unique to the surface structure; If there is a remnant soliton in the bulk, it becomes a nucleation point. Whether the nucleation process of the conical order occurs at the surface or an isolated soliton depends on \(H_{ex}^{z}\) (when we change \(H_{ex}^{x}\) to cause an instability).

Finally we remark that there is another instability at higher field side when there is an isolated soliton. An isolated soliton destabilizes at some field value, and it is called the \(H_0\) line introduced in the skyrmion system at finite temperature[S3]. We identify this instability as the Landau instability by studying the chiral sine-Gordon model. The details about instabilities associated with an isolated soliton are given in Ref. [S4].

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Figure S4. (a) Static configurations to calculate the excitation spectra for \((H_{ex}^x, H_{ex}^z)/H_0 = (0.1370791, 5.2)\), similar to Fig. S2(a). (b) The lowest energy excitation. This mode leads to a distorted conical order. The weight is localized around \(l \sim 50\), which is about the size of the surface structure. This mode stands for the development of the oscillation in the tail of the surface structure.

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