Asymmetric isolated skyrmions in polar magnets with easy-plane anisotropy

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(Dated: April 4, 2017)

We introduce a new class of isolated magnetic skyrmions emerging within tilted ferromagnetic phases of polar magnets with easy-plane anisotropy. The asymmetric magnetic structure of these skyrmions is associated with an intricate pattern of the energy density, which exhibits positive and negative asymptotics with respect to the surrounding state with a ferromagnetic moment tilted away from the polar axis. Correspondingly, the skyrmion-skyrmion interaction has an anisotropic character and can be either attractive or repulsive depending on the relative orientation of the skyrmion pair. We investigate the stability of these novel asymmetric skyrmions against the elliptical cone state and follow their transformation into axisymmetric skyrmions, when the tilted ferromagnetic moment of the host phase is reduced. Our theory gives clear directions for experimental studies of isolated asymmetric skyrmions and their clusters embedded in tilted ferromagnetic phases.

PACS numbers: 75.30.Kz, 12.39.Dc, 75.70.-i.

Magnetic chiral skyrmions are particle-like topological solitons with complex spin structure [1,2] which are the solutions of the field equations of the Dzyaloshinskii’s theory [3]. Recently, skyrmion lattice states and isolated skyrmions were discovered in bulk crystals of chiral magnets near the magnetic ordering temperature [4,5] and in nanostructures with confined geometries over larger temperature regions [6,12]. The small size, topological protection and easy manipulation of skyrmions by electric fields and currents [13,15] generated enormous interest in their applications in information storage and processing [16,17]. Depending on the crystal symmetry of the host materials, distinct classes of skyrmions, such as Bloch and Néel skyrmions, or anti-skyrmions [15] can be realized. In particular, Néel skyrmions were recently found in GaV$_3$S$_8$ and GaV$_4$S$_8$, which are magnetic semiconductors with non-chiral but polar crystal structure [18]. Néel skyrmions emerging in such multiferroic hosts are associated with an electric polarization pattern, which can be exploited for their electric field control [19].

The current interest of skyrmionics is focused on isolated axisymmetric skyrmions within the polarized ferromagnetic (PFM) state of non-centrosymmetric magnets. All the spins around such skyrmions are parallel to the applied magnetic field and point opposite to the spin in the center of the skyrmion, as visualized in Figs. 1(a) & (c). The internal structure of such axisymmetric skyrmions, generally characterized by repulsive skyrmion-skyrmion interaction, has been thoroughly investigated theoretically [21,22] and experimentally by spin-polarized scanning tunneling microscopy in PdFe bilayers with surface induced Dzyaloshinskii-Moriya interactions (DMI) and strong easy-axis anisotropy [23,24]. The existence region of axisymmetric skyrmions was found to be restricted by strip-out instabilities at low fields and a col-

![Fig. 1](color online) Cross-sections of the internal magnetic structures of skyrmions obtained in model (a) with DMI (2) and no axial anisotropy, $k_u = 0$. The magnetic field $\mathbf{h}$ is directed along the $z$ axis. (a) & (c) Color plots of $m_z$ for an axisymmetric skyrmion embedded within the PFM state ($h = 0.55$) in the $xy$ and $xz$ planes, respectively. (b) & (d) the same for a non-axisymmetric skyrmion in the conical phase ($h = 0.3$), with an additional screw-like rotation alongside with the conical phase (white arrow shows the modulation vector $\mathbf{q}$ of the conical phase). The color bar in panel (a) is common for each panel. The in-plane component of the magnetization is represented by black arrows.
FIG. 2. (color online) Numerical solutions for skyrmions using model (1) with DMI (3). (a) - (c) Asymmetric skyrmions within the TFM state for \( h = 0.5 \) and \( k_u = -1.1 \). (d) - (f) Axisymmetric skyrmions within the PFM state for \( h = 1.6 \) and \( k_u = -0.7 \). The top panels are color plots of \( m_z \) in the \( xy \) plane with black arrows representing the in-plane magnetization components. The top insets show the magnetization components in the plane \( xz \) across the skyrmion centers, which are additionally shown as curves in the middle panels. Note that the \( m_x \) and \( m_z \) components seemingly exchange roles in panels (b) & (e), which is due to the different embedding phase, the TFM state with nearly in-plane magnetization and the PFM state with fully out-of-plane magnetization. The bottom panels are color plots of \( P_z \) with white arrows representing the in-plane components of the electric polarization, calculated according to Eq. (4).

The internal spin pattern of isolated skyrmions can break the rotational symmetry once placed into the conical phase of bulk helimagnets, such as the cubic B20 compounds. These skyrmions are not uniform along their axes. Though their central core region nearly preserves the axial symmetry, the domain-wall region, which connects the core with the embedding conical state, is asymmetric. This asymmetric profile of the cross-section forms a screw-like modulation along the skyrmion core, as depicted in Figs. (b) & (d). These asymmetric isolated skyrmions, which can exhibit an attractive skyrmion-skyrmion interaction, were proposed to underlie the precursor phenomena near the ordering temperatures in chiral B20 magnets (MnSi, FeGe) and have prospects in spintronics as an alternative to the common axisymmetric skyrmions.

In this Letter we introduce a new type of isolated skyrmions within the tilted ferromagnetic (TFM) state of magnets with polar crystal structure and easy-plane anisotropy. Such skyrmions are forced to develop an asymmetric shape in order to match their spin pattern with that of the TFM state, meanwhile preserving their topological charge \( q = 1 \). We find that—unlike the repulsive axisymmetric skyrmions and the attractive asymmetric skyrmions respectively embedded in the PFM state and the conical phase of chiral magnets—the asymmetric skyrmions emerging in the TFM state of polar magnets exhibit anisotropic inter-skyrmion potential. Depending on the relative orientation of the two individual skyrmions, this potential can be attractive, leading to the formation of biskyrmion or multiskyrmion states, or repulsive.

Chiral solitons and modulated phases can be derived by minimizing the energy functional of a noncentrosymmetric ferromagnet:

\[
w = \sum_{i,j}(\partial_i m_j)^2 - k_u m_z^2 - \mathbf{m} \cdot \mathbf{h} + w_D.
\]

Here, we use reduced values of the spatial variable, \( \mathbf{x} = \mathbf{r}/L_D \) with \( L_D = A/D \) being the periodicity of the modulated states. \( A \) is the exchange stiffness constant. The sign of the Dzyaloshinskii constant \( D \) determines the sense of rotation. \( k_u = K_u A/D^2 < 0 \) is the uniaxial anisotropy of easy-plane type, \( \mathbf{m} = [\sin \theta \cos \psi, \sin \theta \sin \psi, \cos \theta] \) is the unity vector along the magnetization, and \( \mathbf{h} = \mathbf{HA}/D^2 \) is the applied magnetic field in reduced units.

Depending on the crystal symmetry, the DMI energy \( w_D \) includes certain combinations of Lifshitz invariants \( L_i^{(k)} = \partial_i m_j / \partial x_k - m_j \partial_i m_j / \partial x_k \). Particularly, for cubic helimagnets, such as MnSi, FeGe, Cu2OSeO3 and \( \beta \)-type Mn alloys belonging to the chiral 23 (T) and 432 (O) crystallographic classes, the DMI is reduced to the following form

\[
w_D = \mathbf{m} \cdot (\nabla \times \mathbf{m})
\]

and stabilizes Bloch-type modulations. Two types of isolated skyrmions have been found to exist for DMI: axisymmetric skyrmions within the PFM state and asymmetric skyrmions within the conical phase, respectively visualized in Fig. (a) & (b).

In the phase diagram of noncentrosymmetric ferromagnets with DMI and easy-plane uniaxial anisotropy, the conical phase is the only stable modulated state. Other states including skyrmion lattices and spirals are only metastable solutions of the model, though can be stabilized in real materials by several means, such as thermal fluctuations and confined geometry. Axisymmetric isolated skyrmions exist as metastable excitations of the
On the other hand, the isolated Néel skyrmions embedded in the TFM phase are confined by the following in-plane boundary conditions:

$$\theta(0) = \pi, \theta(\infty) = \theta_{TFM} = \arccos(h/2k_u).$$  \hspace{1cm} (5)

These boundary conditions violate the rotational symmetry, forcing the skyrmions to develop an asymmetric shape.

The results of the numerical minimization of the energy functional [1] with boundary conditions [5] are shown in Figs. 2 (a) & (b). When the canted moment of the TFM state is along the y axis, the asymmetry is clearly reflected in both \(m_x\) and \(m_z\). As implied by \(m_z\), such skyrmions consist of a strongly localized nearly axisymmetric core and an asymmetric transitional region toward TFM state. From the left side of the depicted skyrmion, the magnetization rotates directly from \(\theta_{TFM}\) to \(\theta = \pi\) in the skyrmion center. In contrast, at the right side, the magnetization first passes through \(\theta = 0\) \((m_z = 1)\) and then converges back to \(\theta_{TFM}\). This is the reason of the crescent-shaped anti-skyrmion-like region with a positive energy density over the TFM state, shown in Figs. 3 (b) & (c), which is necessary to maintain the topological charge \(q = 1\) of such a highly distorted asymmetric skyrmion.

As already mentioned, Néel skyrmions in insulating hosts can have a polar dressing in addition to their magnetic pattern. The asymmetry of Néel skyrmions embedded in the TFM phase is also reflected in the spatial pattern of their electric polarization, as visualized in Fig. 2 (c) in comparison with the polar pattern of an axisymmetric Néel skyrmion in Fig. 2 (f). The magnetically induced polarization was calculated using the lowest-order magnetoelectric terms allowed in materials with \(C_{4v}\) or \(C_{6v}\) symmetry:

$$P = \left[\alpha m_x m_z, \alpha m_y m_z, \beta m_z^2 + \gamma(m_x^2 + m_y^2)\right].$$  \hspace{1cm} (6)

For the polarization patterns shown in Figs. 2 (c) and (f), we set \(\alpha=\beta=\gamma\) and used the normalization condition, \(|\mathbf{m}|=1\). While the feasibility of electric-field-driven switching has been demonstrated already for Néel skyrmions [30], the asymmetry of the polarization pattern, characteristic to the isolated skyrmions studied here, can be exploited to control their orientation by in-plane electric fields.

Asymmetric skyrmions within the TFM phase can be considered as \(xy\) cross-sections of asymmetric skyrmions in the conical phases, shown in Figs. 1 (b) & (d) [23,29]. However, the DMI term [3] stabilizing Néel skyrmions does not support any modulation along the z axis. Thus, the asymmetry of Néel skyrmions created within the TFM state is uniform along the z axis, which results in a non-trivial character of inter-skyrmion potential, displayed in Fig. 3 (a). Figs. 3 (b) & (c) present energy density distributions in skyrmion pairs for two mutual orientations, head-to-head and side-by-side. In the
head-to-head configuration, skyrmions form pairs with a fixed inter-skyrmion distance, implying the attractive nature of their interaction, as clear from the green curve in Fig. 3 (a). Therefore, these skyrmions are expected to form 1D chains running along the canted magnetization component of the TFM phase. The calculated inter-skyrmion potential for the side-by-side configuration, the blue curve in Fig. 3 (a), reveals the repulsive character of skyrmion-skyrmion interaction at large distances with a local minimum (or saddle point) at smaller distances. Such a behavior of the inter-skyrmion potential is related to the positive and negative asymptotics of the energy density toward the TFM state. In general, we argue that the inter-skyrmion potential inherently contains a number of minima separated by saddle points (see also Supplement for additional details).

In contrast, the asymmetric pattern of Bloch skyrmions is tightly linked to the conical modulation of the host phase and rotates around the z axis in the same way for each individual skyrmions. This synchronized screw-like rotation of the asymmetry for a pair of such skyrmions leads to an overall attractive potential, by averaging over head-to-head, side-by-side and intermediate configurations alternating along the z axis.

The asymmetric skyrmions within the TFM state can exist only for \( k_u < -1 \), i.e. require a relatively strong easy-plane anisotropy. For \( k_u > -1 \) isolated skyrmions undergo an instability towards the elliptical cone state, as demonstrated in Fig. 3 (a) (see Ref. 22 and Supplement for a detailed information on the structure of the elliptical cone and its lability region). This instability resembles the elliptical instability of skyrmions into spirals considered in Ref. 22 and allows to generalize the considered phenomenon: isolated skyrmions tend to elongate into one-dimensional states (elliptical cones or spirals) which have smaller energy for given control parameters.

The TFM state turns into the PFM state with \( \theta_{PFM} = 0 \) at the line \( h = 2k_u \). In the PFM state the rotational symmetry is recovered and isolated skyrmions become axisymmetric with \( \theta = \theta(\rho) \) and \( \psi = \phi \), as shown in Figs. 2 (d) & (e). The core region and the surrounding ring have positive and negative energy densities, respectively, implying a repulsive skyrmion-skyrmion interaction.

Néel skyrmions can also form the thermodynamically stable skyrmion lattices. Skyrmions within unit cells of such lattices have perfectly hexagonal shape, as seen in Fig. 3 (b), and do not bear any hint on the asymmetric skyrmion structure or skyrmion instability into the elliptical cone.

Results obtained within the model (1) with DMI are valid for bulk polar magnets with axial symmetry as well as for thin films with interface induced DMI. In particular, bulk polar magnetic semiconductors \( \text{GaV}_4\text{S}_8 \) and \( \text{GaV}_4\text{Se}_8 \) with the \( C_{3v} \) symmetry possess uniaxial anisotropy of easy-axis and easy-plane type, respectively. Since the magnitude of the effective anisotropy in these lacunar spinels strongly varies with temperature, these material family provides an ideal arena for the comprehensive study of anisotropic effects on modulated magnetic states. Skyrmions were also studied experimentally in various systems with interface induced DMI. In these thin film structures, the rotational symmetry can also be broken by different anisotropic environments due to lattice strains or reconstructions in the magnetic surface layers, as has been discussed recently for the double atomic layers of Fe on Ir(111). These structural anisotropies also promote the formation of asymmetric skyrmions.

In conclusion, we found a new type of isolated skyrmions emerging in tilted FM states of polar magnets with easy-plane anisotropy. These novel solitonic states are characterized by an asymmetric shape and an anisotropic inter-skyrmion potential. Our results are of particular interest for 2D materials like thin films, surfaces, interfaces, where easy-plane anisotropy can coexist with Rashba-type spin-orbit coupling, activated by the broken surface-inversion symmetry. In order to fully explore their characteristics and functionalities, the internal structure of these asymmetric skyrmions should be studied experimentally, as was done for the axisymmetric individual skyrmions within polarized FM states.

The authors are grateful to K. Inoue, A. Bogdanov, and Y. Togawa for useful discussions. This work was funded by JSPS Core-to-Core Program, Advanced Research Networks (Japan). This work was supported by the Hungarian Research Fund OTKA K 108918.

I. SUPPLEMENTARY INFORMATION

A. Elliptical cone

We analyze solutions of the model (1) starting from one-dimensional cycloids and elliptical cones (Fig. 3). We direct their \( \mathbf{q} \)-vectors along \( x \), and thus get the polar and azimuthal angles as functions of only one spatial coordinate: \( \theta = \theta(x) \), \( \psi = \psi(x) \). The twisting DMI acquires the following form:

\[
w_D = - \cos(\psi) \partial_x \theta + (1/2) \sin(2\theta) \sin(\psi) \partial_x \psi \tag{7}
\]
For a cycloid, $\psi = 0$ and thus $w_D = \partial_x \theta$ (the notation $\partial_x = \partial/\partial x$ is used). For $h = 0$ the average value of DMI energy equals $1/2$ like in undistorted spirals with uniform rotation (Fig. 7(b)). In the applied magnetic field, the rotational part of the cycloids is squeezed to narrow domain walls between wide domains polarized along the field. Thus, the rotational energy gradually decreases to 0. Usually, such a cycloid is considered to satisfy the boundary conditions: $\theta(0) = 0$, $\theta(p/2) = \pi$ where $p$ is a pitch of a cycloid subject to minimization. Thus, in the case of the easy-plane uniaxial anisotropy as well as in the case of easy-axis anisotropy $^{32}$ the cycloid is believed to infinitely expand its period and transform into the TFM state at the line $a - A - C$ (Fig. 5) see also Fig. 12 in Ref. $^{31}$.

In our simulations, however, we show that the cycloidal spiral rather gives rise to the elliptical cone (Fig. 6) at the line $A - B$ (Fig. 5). An elliptical cone (EC) was introduced in Ref. $^{33}$ Spins in this phase trace out a cone with an elliptical cross-section (Fig. 6(a) - (e)). The angle of this cone is defined by the corresponding angle of the tilted FM state, $\theta = \arccos(h/2k_u)$. Such an EC develops from a cycloidal state by the second-order phase transition and gradually increases its $m_y$-component (Fig. 7(a)). At the line $h = 2k_u$, EC continuously transforms into the polarized FM state (Fig. 5). For $k_u < -1$ the EC transforms into the tilted FM state. We note that the TFM state is isotropic in plane. On the contrary, the EC develops its in-plane component perpendicular to the $\mathbf{q}$-vector. By this, the unidirectional sense of the magnetization rotation in EC is
preserved, and the DMI energy of EC (7) is modified by the additional $\psi(x)$-dependence. At the lines $k_u = -1$ and $A = 2k_u$ the rotational energy (7) falls to 0 (Fig. 7 (b)).

B. The phase diagram of states

The complete phase diagram (Fig. 8) of states of the model (1) has been reproduced from Ref. 33 and includes stability regions of modulated phases and regions of metastable skyrmions. Points A-D indicate parameters for different solutions used in the manuscript: point A – asymmetric skyrmions within TFM phase (Fig. 2 (a) - (c)); point B – axisymmetric skyrmions within PFM phase (Fig. 2 (d) - (f)); point C - hexagonal skyrmion lattice (Fig. 4 (b)), point D - instability of asymmetric skyrmions with respect to the elliptical cone (Fig. 4 (a)).

The phase diagrams also allows to generalize the processes of skyrmion lattice formation. Along the line $a - b$ the skyrmion lattice appears as a result of condensation of isolated skyrmions (building blocks of the hexagonal skyrmion lattice), as found for axisymmetric skyrmions in the easy-axis case. Along the first-order phase transition line $a - d$, however, none of the aforementioned mechanisms is appropriate. Presumably, domains of the skyrmion lattice and the elliptical cone state coexist with non-trivial domain boundary between them.

C. Inter-skyrmion potential

To calculate the skyrmion-skyrmion interaction potential, the following procedure was proved to be appropriate for axisymmetric skyrmions and asymmetric skyrmions within the conical phase. The energy density (1) is minimized with the constraint $m_z = -1$ imposed at the centers of two skyrmions as a function of the distance between skyrmion centers. This procedure, however, faces the following difficulty when applied to asymmetric skyrmions within TFM states: skyrmions will locally deform the surrounding TFM state to achieve the minimum of the system. Thus, only positions of (local and global) minima can be found precisely. To reconstruct the interaction potential with all underlying details, one should impose a control over the in-plane component of the TFM state, i.e. violate its in-plane isotropy.
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