Quartic asymmetric exchange for monolayer Fe₃GeTe₂ and other two-dimensional ferromagnets with trigonal prismatic symmetry

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We suggest a possible origin of noncollinear magnetic textures in ferromagnets (FMs) with the $D_{3h}$ point group symmetry. Suggested mechanism is different from the Dzyaloshinskii-Moriya interaction (DMI) and its straightforward generalizations. Considered symmetry class is important because a large fraction of all single-layer intrinsic FMs should belong to it. In particular, so does a monolayer Fe₃GeTe₂. At the same time, DMI vanishes identically in materials described by this point group, in the continuous limit. We use symmetry analysis to identify the only possible contribution to the free energy density in two dimensions that is of the fourth order with respect to the local magnetization direction and linear with respect to its spatial derivatives. This contribution predicts long-range conical magnetic spirals with the average magnetization that depends on the spiral propagation direction. We relate the predicted spirals to a recent experiment on Fe₃GeTe₂. Finally, we demonstrate that for easy plane materials the same mechanism may stabilize bimerons.

Introduction. Isolation of graphene in 2004 [1, 2] attracted a remarkable interest to purely two-dimensional (2D) materials. This field has been growing ever since and a large variety of other stable atomically thin crystals has been discovered. It is not only the fundamental interest [3–6] that drives the research on 2D materials, but the potential applications as well. Of a particular importance for the latter is the fact that low-dimensional systems can be tuned in a much more effective way than their bulk counterparts [7, 8]. It is also important that one can combine properties of different 2D materials by stacking them in heterostructures [9, 11].

Potential applications of heterostructures and monolayers include, among others, spin-based computer logic and new ways to store information [12, 13]. In particular, it is assumed that noncollinear magnetic textures, like skyrmions and miniature domain walls, might become the basis for future memory devices [14–16]. However, for more than a decade no atomically thin intrinsic magnets have been realized in experiments. This happened only in 2017 when magnetic order was reported in two-dimensional van der Waals materials Cr₂Ge₂Te₆ [17] and CrL [18]. Soon they were accompanied by a metallic itinerant ferromagnet Fe₃GeTe₂ [19, 20] (FGT).

Recently, spin spirals were reported in thin multilayers of FGT [21] and Néel-type skyrmions were observed in two different heterostructures based on this material [22, 23]. It is however interesting that noncollinear magnetic order in pure FGT cannot be explained by the Dzyaloshinskii-Moriya interaction [24, 25] (DMI). The reason for this is the following. Bulk FGT has an inversion symmetry center, and thus smooth textures cannot originate in associated with DMI contributions to the free energy. Monolayer FGT, on the other hand, does lack the inversion symmetry. But its point group $D_{3h}$ is still so symmetric that any contribution to the free energy density of the form $n_i \nabla_j n_k$ can affect magnetic order only at the sample boundaries. This fact was coined in Ref. [20] and repeated in a recent paper [27] with an illustrative title “Elusive Dzyaloshinskii-Moriya interaction in monolayer Fe₃GeTe₂.” Some of us also mentioned this in Ref. [28]. Here by $n$ we denote the unit vector of the local magnetization direction.

In addition to monolayer FGT, the group $D_{3h}$ describes many other 2D ferromagnets (FMs). For example, some transition metal dichalcogenides (TMDs), when thinned down to a single layer, are predicted to be intrinsically magnetic [29]. Typically, 2D TMDs are formed in either 1T or 2H phases [29, 30], and the latter phase is characterized by $D_{3h}$. Another large group of magnetic monolayers, for which the 2H phase (and the $D_{3h}$ symmetry) is often favourable, are transition metal dichalides [31]. Recently predicted 2D chromium pnictides [32] that are half-metallic ferromagnets with very high Curie temperatures are described by the point group $D_{3h}$ as well. Overall, $D_{3h}$, which is the group of symmetries of a triangular prism, is an important group in the field of intrinsic 2D magnetism. In this Letter, we introduce a novel possible origin of smooth noncollinear magnetic textures in materials described by this point group.

Symmetry analysis. Elusive nature of DMI in such materials is characterised by vanishing antisymmetric contributions $n_i \nabla_j n_k - n_k \nabla_i n_j$ to the free energy density. Similar symmetric terms are represented by full derivatives $\nabla_j (n_i n_k)$ and are therefore relevant only close to the edges of the sample. Therefore, in large samples, terms that are quadratic with respect to $n$ do not contribute to formation of smooth textures. The latter are characterized by small spatial derivatives of magnetization, and it is thus worthwhile to consider other contributions that are linear with respect to the derivatives of $n$. Namely we would like to study terms of the form $n_i n_j n_k \nabla_l n_p$. We call them the “quartic asymmetric exchange” terms by analogy with DMI. Physically they can correspond,
for example, to interactions between four spins [33, 34].

We use standard symmetry analysis [35, 36] to identify all quartic asymmetric exchange terms allowed in $D_{3h}$. This group contains a 3-fold rotation around the z-axis, a mirror symmetry with respect to the $xy$-plane, and three 2-fold rotations around axes at the angles 0, ±$2\pi/3$ in the $xy$-plane. Quartic contributions

$$\sum_{ijklp} D_{ijklp} \cdot n_i n_j n_k \nabla n_p$$

(1)

to the free energy density should remain invariant under the transformation

$$D_{ij'k'l'p'} = \sum D_{ijklp} \cdot g_{ii'} g_{jj'} g_{kk'} g_{ll'} g_{pp'}$$

(2)

for every group element $g$. By applying all generators of $D_{3h}$ to Eq. (2), we find that there are precisely seven such invariant contributions in this group, and we collect them in Table I. Surprisingly, up to boundary terms, five of them are not independent. In order to prove this, one should take into account the constraint $n^2 = 1$ (see also Table I). We can choose

$$w_\parallel = n_x (n_x^2 - 3n_y^2) (\nabla_x n_x + \nabla_y n_y)$$

(3a)

$$w_\perp = n_x (n_x^2 - 3n_y^2) \nabla_z n_z$$

(3b)

as the only independent invariants. For a 2D system, the second one can be also disregarded. Hence, if the effects of boundaries are negligible, we are left with a single quartic term $w_\parallel$.

It is useful to relate the structure of $w_\parallel$ to the lattice geometry of a typical 2D crystal described by the point group $D_{3h}$. One can notice that

$$w_\parallel \propto (n \cdot \delta_1)(n \cdot \delta_2)(n \cdot \delta_3) (\nabla_x n_x + \nabla_y n_y),$$

(4)

where $\delta_i$ represent the nearest neighbour vectors. These vectors or their opposite make the angles 0, ±$2\pi/3$ with the positive $z$-axis (see the top part of Fig. I). They also correspond to the three armchair directions of a typical hexagonal lattice generated by $D_{3h}$. Using Eq. (1), one can obtain a classical Heisenberg model for $w_\parallel$. For a site with the spin $S_i$, we have

$$w_{\parallel, H} \propto (S \cdot \delta_1)(S \cdot \delta_2)(S \cdot \delta_3) \sum_i (S_i \cdot \delta_i),$$

(5)

where the spins $S_i$ are the nearest neighbours of $S$.

We note that the effective interaction of Eq. (5) is fundamentally different from the recently proposed [37, 38] interactions of the form $(S_i \times S_j)(S_i \cdot S_j)$. In the continuous limit, the latter are represented by higher order terms with respect to the gradients of magnetization direction. Thus, for smooth textures, interaction of Eq. (5) should be the leading one. At the same time, for textures varying on the scale of a lattice spacing, this is not the case.

TABLE I: All fourth order contributions to the free energy density that are linear with respect to spatial derivatives of $n$.

| Contribution | Expression |
|--------------|------------|
| $I_1$ | $n_x (n_x^2 - 3n_y^2) (\nabla_x n_x + \nabla_y n_y) \equiv w_\parallel$ |
| $I_2$ | $n_x (n_x^2 - 3n_y^2) \nabla_z n_z \equiv w_\perp$ |
| $I_3$ | $n_y (n_y^2 - 3n_x^2) (\nabla_x n_y - \nabla_y n_x)$ |
| $I_4$ | $(n_x^2 + n_y^2) (\nabla_x (n_y^2 - n_x^2) + 2 \nabla_y (n_x n_y))$ |
| $I_5$ | $n_x (n_x^2 - n_y^2) \nabla_x n_x + 2n_x n_y \nabla_y n_x$ |
| $I_6$ | $n_x (n_x^2 - n_y^2) \nabla_z n_x + 2n_x n_y \nabla_y n_x$ |

We also note that there exist many other mechanisms that can produce noncollinear magnetic textures. Our mechanism is a natural replacement of DMI for systems where the latter is forbidden.

Spin spirals. Now let us find out whether the quartic term $w_\parallel$ can stabilize spin spirals observed in FGT. In order to do this, we consider a conical ansatz

$$(\mathbf{n}(r) = m \cos \alpha + [m_\theta \cos (kr) + m_\phi \sin (kr)] \sin \alpha)$$

(6)

that parameterizes the transition from a collinear state, $\sin \alpha = 0$, to a helix, $\cos \alpha = 0$ (if $k \neq 0$). Here

$$m = (\cos \phi \sin \theta, \sin \phi \sin \theta, \cos \theta)$$

(7a)

$$m_\theta = (\cos \phi \cos \theta, \sin \phi \cos \theta, - \sin \theta)$$

(7b)

$$m_\phi = (- \sin \phi, \cos \phi, 0)$$

(7c)

is a standard basis in spherical coordinates. Vectors $m_\theta$ and $m_\phi$ correspond to oscillations, while $m$ points in the direction of the average magnetization.

We substitute this ansatz into a model that accounts for symmetric exchange ($A$), magnetic anisotropy ($K$), and quartic asymmetric exchange ($D$),

$$f = A \left( (\nabla_x n_x)^2 + (\nabla_y n_y)^2 \right) + K n_z^2 + 2Dw_\parallel,$$

(8)

and average the total free energy $F = \int d^2r f$ over a large volume. Oscillating terms with $kr$ then vanish and the averaged density ($f$) becomes a quadratic function of the wave vector $k$. Therefore, we straightforwardly minimize it with respect to $k$ and find:

$$A \frac{D^2}{2} (f) = - \frac{9}{64} (\sin \alpha + 5 \sin 3\alpha) \sin^4 \theta \cos^2 \theta$$

$$+ AK \frac{2D^2}{2} \sin^2 \theta \sin^2 \alpha + 2 \cos^2 \theta \cos^2 \alpha),$$

(9)

where the combination $AK/D^2$ is dimensionless and further minimization with respect to $\theta$ and $\alpha$ is needed.
The wave vector that corresponds to the minimum is expressed as

$$\begin{pmatrix} k_x \\ k_y \end{pmatrix} = -\frac{3D}{2A} \left(5 \cos^2 \alpha - 1\right) \sin^2 \theta \cos \theta \left(\frac{\sin 2\phi}{\cos 2\phi}\right). \quad (10)$$

Before we proceed with the minimization, it is interesting to note that states described by Eq. (9) are degenerate with respect to $\phi$. In other words, their free energy does not depend on the azimuthal angle of “the average magnetization vector” $\mathbf{m}$. At the same time, for spiral-like textures with a finite wave vector, the angles between $\mathbf{k}$ and $\mathbf{m}$ are different for different values of $\phi$:

$$\mathbf{k} \cdot \mathbf{m} \propto \sin 3\phi, \quad (11)$$

as it follows from Eqs. (7a, 10). Thus, by controlling the direction of the average magnetization (vector $\mathbf{m}$), one should also be able to control the propagation direction of the spiral. Such control can be achieved by an application of a small external magnetic field. The latter couples only to $\mathbf{m}$, therefore it can be used to set the desired value of $\phi$. As we see from Eq. (11), when the in-plane component $m_\parallel$ of the vector $\mathbf{m}$ lies along one of the armchair directions of the lattice ($\phi = 0$, $2\pi/3$, $4\pi/3$), then $\mathbf{k}$ is orthogonal to it. When $\mathbf{m}_\parallel$ is along a zigzag direction ($\phi = \pi/6$, $5\pi/6$, $3\pi/2$), then $\mathbf{k}$ (or $-\mathbf{k}$) points in the same direction (see the bottom part of Fig. 1). We wonder whether such control of the wave vector direction can be realized experimentally.

Let us proceed with consideration of the functional defined by Eq. (9). Perturbative analysis with respect to small $AK/D^2$ provides almost a perfect fit for its minimum in the entire range of parameters. States with $k \neq 0$ can exist when $-0.98 \lesssim AK/D^2 \lesssim 2.18$, and for such states we find

$$\begin{align*}
\sin^2 \theta &= \frac{2}{3} + \frac{9}{128} \frac{AK}{D^2} + \ldots, \\
\sin^2 \alpha &= \frac{4}{15} - \left(\frac{9}{32\sqrt{10}} \frac{AK}{D^2}\right)^2 + \ldots,
\end{align*} \quad (12a, 12b)$$

where only the leading and the subleading order terms are shown. For other values of the parameter $AK/D^2$, the state is collinear: out-of-plane for $K < 0$ and in-plane for $K > 0$ (see Fig. 2). This resembles a typical situation with magnetic textures determined by DMI: when the absolute value of the DMI strength $D$ exceeds some critical value $D_c \propto \sqrt{|K|}$, the system is found in a helical ground state, while for $|D| < D_c$ the uniform magnetization is favoured. Helical states described by Eqs. (12) are in a reasonable agreement with spirals found in FGT. For $\phi = 0$ and large enough $|D|$, we have a helical texture with finite $k$ pointing along the $y$-direction. The components $n_y$ and $n_z$ of this texture oscillate with a phase difference of $\pi/2$ (see Eqs. (6), (7)). This is precisely what has been reported in Ref. [21]. In addition, however, we have an oscillating $n_x$ component, with a slightly smaller amplitude that is, basically, equal to $\cos \theta$. It is not clear whether this is a crucial disagreement with the experiment or something that was not seen in it. We should also note that $x$ and $y$ axes in Ref. [21] describe the coordinates of detectors, not the crystal axes. Hence the $x$ component of magnetization in that paper can indeed correspond to our $n_y$.

**Skyrmions and bimerons.** It turns out that circular skyrmions cannot be stabilized by $w_\parallel$. For a standard ansatz $n(r) = (\cos \Phi \sin \Theta, \sin \Phi \sin \Theta, \cos \Theta)$, with
Φ = QΦ + δ and Θ = Θ(r), integration over φ nullifies w|| when Q is an integer. At the same time, there exist predictions of skyrmions with more complex axial symmetry [12], including trigonal [13-14]. Such symmetry is natural for D3h and we have checked that indeed, for “trigonal skyrmions”, w|| is generally finite after the angle integration. We do not think that this can explain skyrmions observed in the experiments of Refs. 22, 23, but the spatial symmetries of heterostructures studied in these works are different from D3h anyway.

FGT is an easy axis magnet [3]: K < 0. For materials with an easy plane anisotropy, K > 0, our model can stabilize bimerons – the in-plane skyrmions [45-47]. As we have mentioned, a collinear state is preferred over spin spirals when D2 ≲ AK/2.18 ≈ 0.46AK. In this case, a bimeron can exist as a metastable transition from one in-plane state to another. In order to demonstrate this, let us consider a parameterization

\[ n(r) = R_z[φ_0] \begin{pmatrix} \cos Θ, \cos φ \sin Θ, \sin φ \sin Θ \end{pmatrix}, \] (13a)

\[ Φ = Q(φ + φ_0) + δ, \quad Θ = Θ(r), \] (13b)

with the boundary conditions Θ(0) = π, Θ(∞) = 0. Here Q is the bimeron’s topological charge, and R_z[φ_0] is a matrix of rotation by an arbitrary angle φ_0 with respect to z. Eqs. (13) describe a bimeron magnetized at \( r = ∞ \) in the direction set by the polar angle φ_0. The inset in the bottom panel of Fig. 3 provides an illustration of a bimeron that corresponds to \( δ = π/2, φ_0 = 0, Q = 1 \).

We substitute Eqs. (13) with Q = 1 into Eq. (8), integrate over φ, and compute a functional derivative of the result. This brings us to the Euler-Lagrange equation

\[ \Theta''(r) + \frac{\Theta'(r)}{r} - \frac{\sin 2Θ(r)}{2r^2} - \frac{K \sin 2Θ(r)}{4A} - \frac{3D \sin (3φ_0 + δ)}{2Ar} \sin^2 Θ(r) \left[ 5 \cos^2 Θ(r) - 1 \right] = 0. \] (14)

It is very similar to the equation that describes skyrmions in the presence of the DMI term \( n_z(∇ \cdot n) - (n \cdot ∇) n_z \). From Refs. [14-18] we know that the radial profile of such skyrmions can be very well approximated by a domain wall with two parameters. We can use the approach of these papers to analyze solutions of Eq. (14) as well.

The only difference between the Euler-Lagrange equation for skyrmions and Eq. (14) is the presence of the term \( 5 \cos^2 Θ(r) - 1 \) in the latter. At small values of r, its effect on the solutions is minimal. But for larger r, when \( \cos Θ(r) ≈ ±1/√5 \), this term becomes important. Therefore, we can expect that the bimeron profile Θ(r) is a superposition of a domain wall and some additional structure that is relevant at large r. Being optimistic, one can hope that at least some properties of Θ(r) can be captured from the analysis of its domain wall “component” alone. It turns out that this is indeed the case.

We employ the ansatz of Ref. [18]:

\[ Θ_{dw}(r) = 2 \arctan \left[ \frac{\sinh (R/Δ)}{\sinh (r/Δ)} \right]. \] (15)

where Δ is the width of the domain wall, and R is the profile radius: \( Θ_{dw}(R) = π/2 \). Assuming \( R ≫ Δ \) and repeating considerations of Ref. [18], we can estimate the free energy F of this ansatz as

\[ F ≈ 4π \left[ A \left( \frac{R}{Δ} + \frac{Δ}{R} \right) + \frac{KRΔ}{2} + \frac{3πDR}{16} \sin (3φ_0 + δ) \right]. \]

Based on this result, we can argue that the minimal energy corresponds to \( \sin (3φ_0 + δ) = -\text{sign} D \). Alternatively, one can see this from the direct minimization of the above expression (using the fact the R should be positive). Either way, we minimize our expression for F with respect to both R and Δ to obtain

\[ Δ = \frac{3π|D|}{16K}, \quad R = \frac{Δ}{\sqrt{1 - KΔ^2/2A}}. \] (16)

For \( D^2/\text{AK} \lesssim 0.46 \), the square root can be safely ignored and we are left with \( R = Δ = 3π|D|/16K \).

This result obviously contradicts the initial assumption \( R ≪ Δ \). Nevertheless, it works astonishingly well when
\[ r \lesssim R \], as can be seen from Fig. 3. There we plot numerical solutions of Eq. (14) that was supplemented with the condition \( \sin (3 \phi_0 + \delta) = - \text{sign} \> D \). For \( \Theta(r) \geq \pi/2 \), the ansatz \( \Theta_{dw}(r) \) correctly reproduces the shape of the bimeron (top panel) and allows us to get a good estimate of its radius (bottom panel). Out of curiosity, we also solved Eq. (14) for \( D^2/\Delta K = 4 \) (inset of the top panel). The result looks like a superposition of two domain walls that match at \( \cos \Theta(r) \approx 1/\sqrt{5} \). This is of course by far the “spiral-region” of our model.

Two more comments should be made. First, the actual relation between the bimeron direction \( \phi_0 \) and its phase \( \delta \) can be more complex than \( \sin (3 \phi_0 + \delta) < 0 \), solutions of Eq. (14) are strong minimums of the free energy [19]. Thus \( w_l \) can indeed stabilize a bimeron (regardless of the assumptions we used to estimate its radius).

**Conclusion.** We used symmetry analysis to obtain all contributions to the free energy density of the form \( n_i n_j n_k \nabla m_p \) that are allowed by the point group \( D_{3h} \). There are exactly seven such contributions. Only two of them can be chosen as independent if boundary terms are ignored, and only one of these two does not vanish in a 2D system. We demonstrated that the quartic term \( w_l = n_x (n_x^2 - 3 n_y^2) (\nabla n_x + \nabla n_y) \) looks compatible with spin spirals observed in a recent experiment on FGT. It does not stabilize circular skyrmions, but for FMs with an easy plane it can stabilize bimerons. We estimated the radius and energy of such bimerons analytically and calculated their profiles numerically by solving the Euler-Lagrange equation. We argue that the quartic asymmetric exchange term \( w_l \) introduced in this Letter should be considered as a potential source of noncollinear magnetic textures in many 2D intrinsic ferromagnets. To investigate the role of this term numerically, one may use the effective Heisenberg model that we derived.

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SUPPLEMENTARY MATERIAL
Quartic asymmetric exchange for monolayer Fe$_3$GeTe$_2$ and other two-dimensional ferromagnets with trigonal prismatic symmetry

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In this Supplementary Material we discuss the bimeron stability. We also comment on the relation between the bimeron direction $\phi_0$ and its phase $\delta$.

A. Dimensionless equations and stability

We introduce $d = D \sin (3\phi_0 + \delta)$ and $\rho = r|d|/A$. This allows us to rewrite the Euler-Lagrange Eq. (14) in the dimensionless form

$$\Theta''(\rho) + \frac{\Theta'(\rho)}{\rho} - \frac{\sin 2\Theta(r)}{2\rho^2} - \frac{AK \sin 2\Theta(\rho)}{d^2} + \frac{3\sigma}{2\rho} \sin^2 \Theta(\rho) \left[ 5 \cos^2 \Theta(\rho) - 1 \right] = 0,$$

(s1)

where $\sigma = -\text{sign} \{D \sin (3\phi_0 + \delta)\}$ and it is assumed that $d \neq 0$. Sufficient conditions for an extremum are considered with the help of the Jacobi accessory equation [s2]. It reads

$$h''(\rho) + \frac{h'(\rho)}{\rho} - \left\{ \left( \frac{2}{\rho} + \frac{\rho AK}{d^2} \right) \cos 2\Theta(\rho) + 3\sigma \sin 2\Theta(\rho) - (5/2) \sin 4\Theta(\rho) \right\} \frac{h(\rho)}{2\rho^2} = 0,$$

(s2)

where $\Theta(\rho)$ is a solution of Eq. (s1).

In Eqs. (s1) and (s2), $\sigma = 1$ or $\sigma = -1$. It is not known a priori which one of these two possibilities is realized. In the main text of the manuscript we have minimized the bimeron free energy for the domain wall ansatz. According to this analysis, the following condition should hold:

$$\sin (3\phi_0 + \delta) = -\text{sign} \, D.$$

(s3)

It corresponds to $d = -|D|$ and $\sigma = 1$. We use the latter fact as a motivation to first solve Eqs. (s1) and (s2) for this value of $\sigma$. The solutions are obtained numerically for several values of the parameter $d^2/AK$. The Jacobi accessory equation is solved with the initial conditions $h(0) = 0$, $h'(0) = 1$. It turns out that none of the solutions $h(\rho)$ have zeroes different from the one at the origin. Moreover, the second derivative of the free energy with respect to $\Theta'$ is equal to the combination $4\pi A\rho$ that is nonnegative. Hence, the sufficient conditions for a strong minimum are satisfied [s2]. Therefore, we argue that for $D \sin (3\phi_0 + \delta) < 0$ the ansatz of Eqs. (13) describes a strong minimum of the free energy, i.e. the bimeron is stabilized by the quartic asymmetric exchange term $w_{\parallel}$.

FIG. s1: Solutions $\Theta(\rho)$ of Eq. (s1) (dashed lines) and the corresponding solutions $h(\rho)$ of Eq. (s2) (solid lines), for $\sigma = 1$ and three different values of the parameter $d^2/AK$.

In the opposite case, $\sigma = -1$, we are unable to find any solutions of Eq. (s1), using the shooting method. We anticipate that in this case it does not have solutions at all (for the boundary conditions $\Theta(0) = \pi$, $\Theta(\infty) = 0$).
other words, if $D \sin (3\phi_0 + \delta) > 0$, the bimeron is expected to be unstable. This is in line with the fact that for the domain wall ansatz the minimum of the free energy is reached only when $\sigma = 1$ (see Eq. (s3)).

B. Relation between $\phi_0$ and $\delta$

Considerations of the previous section suggest that the condition $D \sin (3\phi_0 + \delta) < 0$ ensures the bimeron stability. However, the concrete relation between $\phi_0$ and $\delta$ that corresponds to a global minimum should be obtained for all particular values of $AK/D^2$ and sign $D$ by minimizing the bimeron free energy with respect to $\phi_0$ and $\delta$. The result can be more complex than that of Eq. (s3). Nevertheless, it will be also given by a periodic function of $\phi_0$ with a period of $2\pi/3$.

[s1] If $d = 0$, then $\Theta(r)$ in Eq. (14) describes a skyrmion profile in the absence of DMI. Such a skyrmion has a zero radius [s3].
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