Formation of magnetic biskyrmions mediated by an intrinsic emergent monopole-antimonopole pair

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Various properties and potential technological applications of magnetic skyrmions have stimulated a flourishing interest in topological spin textures. Among them, biskyrmions with a rare topological charge of two are observed but their existence is still under debate. In this work, we present the formation of biskyrmion bubbles mediated by emergent monopoles via micromagnetic simulations. We find that biskyrmion bubbles and trivial bubbles share a unified three-dimensional structure, in which the relative position of an intrinsic emergent monopole-antimonopole pair dominates the two-dimensional topological property at the middle plane of magnetic uniaxial films. Biskyrmion bubbles can be transformed from trivial bubbles by the motion of emergent monopoles in confined geometry, paving the way for developing devices. These results highlight the three-dimensional aspect of skyrmion-related nanostructures and the versatile roles of emergent monopoles in topological spin textures.

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INTRODUCTION

Topological phases have attracted widespread interest in condensed matter physics and topological defects play a key role in phase transitions. In spin ordered systems, magnetic skyrmions, particle-like spin textures of nontrivial topology1, have received much attention due to their fundamental research interests and potential applications in spintronics2–5. While intensive efforts have been made to improve the applicability of conventional skyrmions, alternative topological objects have been studied recently6–9. These spin textures can exhibit different properties and bring possibilities for innovative device applications10–13. Among them, biskyrmions observed in centrosymmetric magnets14,15 attract lots of attention due to their rare topological charge of two even in the absence of the Dzyaloshinskii-Moriya interaction. However, it is later proposed that biskyrmions could be misleading images of topologically trivial bubbles observed by Lorentz transmission electron microscopy (TEM)16. Nonetheless, magnetic bubbles cannot explain the observation of a large topological Hall effect (THE)15, although the THE is not sufficient evidence for the existence of skyrmions. In addition, the stability of biskyrmions in films of finite thickness has been confirmed theoretically19. Therefore, the existence of biskyrmions still remains an open problem to be solved. Alternative techniques have to be utilized to provide more experimental evidence. On the other hand, it is imperative to understand the formation mechanism of biskyrmions theoretically, which can serve as guidelines of realizing and utilizing biskyrmions in experiments. In the studies on the formation of skyrmions, emergent monopoles (MPs) or antimonopoles (AMPs) can be driven through the middle plane, inducing the formation of biskyrmion bubbles at the middle plane. This mechanism can be utilized to generate biskyrmion bubbles in a confined-geometry magnetic film.

RESULTS

Emergent monopole-antimonopole pair

Although magnetic skyrmions are usually assumed to be homogeneous through the film thickness, the surface magnetization of a Bloch-type skyrmion can form Néel-caps24–26. Such configurations also exist in the 3D structure of topologically trivial bubbles16. Therefore, we revisit the 3D structure of a type-II bubble as shown in Fig. 1a. As can be seen, the magnetization forms a type-II bubble at the middle plane while it points out (in) radially at the top (bottom). The magnetization indicated by the green (red) arrows is opposite between the middle and the top (bottom) plane. An intrinsic 3D Bloch point (BP) resides between them, considering the magnetization is also opposite inside and outside the domain wall. The contour surface with \( m_z = 0 \) of the 3D structure of a simulated type-II bubble, i.e., the two-dimensional (2D) domain wall, is depicted in Fig. 1b. Two BPs exist on this surface, as indicated by the circles in Fig. 1b.

A BP can be characterized by a 3D charge27:

\[
q = 1/8\pi \int dA \epsilon_{ijk} \mathbf{m} \cdot \partial_i \mathbf{m} \times \partial_j \mathbf{m}
\]

\( \text{(1)} \)

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where \( \mathbf{m} \) is the unit vector of magnetization and the integration is taken over a closed surface \( A \) surrounding the BP. Because the two BPs in Fig. 1b have opposite charges, they can be described as an emergent MP-AMP pair\(^{28}\). Thus, type-II bubbles can be exploited as a platform to study the physics of emergent monopoles.

**Magnetic biskyrmion**

Next, we study the magnetization structure in a film with tilted uniaxial anisotropy. Micromagnetic simulations are performed using Mumax3\(^{329}\), considering the exchange, dipolar, uniaxial anisotropy and Zeeman interactions. The material parameters of the biskyrmion host MnNiGa alloy are as follows\(^{16}\): exchange stiffness \( A = 2 \times 10^{-17} \text{J} \cdot \text{m}^{-1} \), saturation magnetization \( M_s = 5.16 \times 10^5 \text{A} \cdot \text{m}^{-1} \), and uniaxial anisotropy \( K_u = 8.7 \times 10^5 \text{J} \cdot \text{m}^{-1} \). Previous studies report that the crystal orientation, thus the magnetic easy axis direction, is a key ingredient to form biskyrmions\(^{39}\). Hence, we perform simulations with the easy axis at a tilted angle of 31° as in the (12\( \bar{2} \)) crystal plane\(^{30}\). The thickness of the film in simulations is 160 nm, corresponding to the typical film thickness in experiments. The values of geometry, cell size, and external field can be varied and are always specified in figure captions or the main text. The simulation is initialized by relaxing a state with random magnetization orientations at an external magnetic field normal to the sample. We start with a sample of 1.2 \( \mu \text{m} \times 1.2 \mu \text{m} \times 160 \text{nm} \) size with cells of 2 nm \( \times \) 2 nm \( \times \) 4 nm. The magnetic easy axis lies in the xz-plane. The simulation is performed with magnetization averaged through the thickness at a defocus of 1.6 mm\(^{36}\).

**Fig. 1** Emergent monopole-antimonopole pair in the 3D structure of type-II bubbles. a A sketch of the 3D structure of a type-II bubble. b The contour surface with \( m_z = 0 \) of a simulated bubble and diagrams of the MP-AMP pair on it. The sample is defined as 2 \( \mu \text{m} \times 2 \mu \text{m} \times 200 \text{nm} \) size with cells of 4 nm \( \times \) 4 nm \( \times \) 4 nm. The simulation is performed with a perpendicular easy axis at a perpendicular external field of 0.06 T. Other parameters are the same as in the main text. The color represents the direction of magnetizations as a conventional color wheel at the right bottom corner. The contour surface is drawn with the software Spirit\(^{35}\).

**Fig. 2** Spin textures of biskyrmions and bubbles and corresponding TEM images. a, b The spin texture of a biskyrmion slightly below the middle plane of the film and a bubble at the middle plane of the film, respectively. The arrow represents the in-plane magnetization \( m_{xy} \). c, d The distribution of topological charge density per unit cell, i.e., the integrand in Eq. (2), of the spin textures in a and b, respectively. g The bubble transformed from the biskyrmion in a at 0.2 T. The external field is increased with steps of 0.02 T, and the state is relaxed in every step. The magnetic easy axis lies in the xz-plane. e, f, h The corresponding Lorentz TEM images of a, b, g, respectively. Calculation is performed with magnetization averaged through the thickness at a defocus of 1.6 mm\(^{36}\).
these spin textures can be calculated as:
\[
Q = \frac{1}{4\pi} \int dx dy \, \mathbf{m} \cdot \nabla_x \mathbf{m} \times \nabla_y \mathbf{m}
\] (2)

The topological charge \(Q\) of the spin texture is 2 in Fig. 2a against 0 in Fig. 2b via numerical calculation. Nevertheless, the spin texture in Fig. 2a is not the very model assumed with Lorentz TEM images, which is composed of two Bloch-type skyrmions with opposite helicities. By contrast, it is a thin-wall biskyrmion bubble, whereas it would still be called a biskyrmion to be distinguished from topologically trivial bubbles in the following. On the other hand, the in-plane magnetization of both the biskyrmion and the bubble is a bound state of two distorted vortices with opposite helicities as illustrated by white arrows in Fig. 2a, b.

We note that the structures in Fig. 2a, b are quite similar. The main difference is the direction of the Bloch lines in domain walls indicated by the dashed circles, which turns out to be the key factor for the topological charge as indicated in Fig. 2c, d. This difference can be better understood by the winding number associated with domain walls, which describes a \(2\pi\) twist along a domain wall\(^{11,12}\). Although this interpretation is equivalent to the topological charge in Eq. (2), it emphasizes the structure of domain walls. In detail, the Bloch lines reverse the magnetization twice and thus make a twist of \(4\pi\) along the domain wall in Fig. 2a, whereas the opposite Bloch lines cancel the twist along the domain wall in Fig. 2b.

When the external field is up to 0.2 T, the biskyrmion in Fig. 2a becomes a standard type-II bubble without in-plane magnetization inside the domain wall, as shown in Fig. 2g. It should be noted that the bubble in Fig. 2b would be similar to the bubble in Fig. 2g under a high field.

We calculate the Lorentz TEM images of these spin textures as displayed in Fig. 2 and compare with previous experimental results. The images in Fig. 2e, f are similar and consistent with previous experimental results at low external field after field cooling\(^{33}\). Moreover, the image shown in Fig. 2h consisting of black and white semicircles is in good agreement with the experimental results observed at high field\(^{14,15}\). These results confirm that tilted bubbles can produce the Lorentz TEM images giving rise to biskyrmion-like configurations, because the TEM is sensitive to the in-plane component of the magnetic flux density averaged through the sample thickness\(^{14}\). Nevertheless, the spin texture corresponding to such an image could still be a biskyrmion.

On the other hand, the 3D structures of the biskyrmion in Fig. 2a and the bubble in Fig. 2b are quite similar as shown in Fig. 3a and b. The magnetization still points radially on the surfaces as in Fig. 1, but the cores are eccentric due to the tilted easy axis. The tilt of the easy axis also aligns Bloch lines roughly along its in-plane projection direction (x-axis). Thus, the easy axis with the surface normal defines a symmetry plane throughout the sample thickness and the Bloch lines roughly lie in this plane. As discussed earlier, the Bloch lines in the biskyrmion are opposite to the bubble. Therefore, the Bloch lines reverse at different points between the biskyrmion and the bubble (see sketches of the 3D structures in Supplementary Fig. 2), which can be illustrated by the distribution with \(m_z = -1\) (cones in Fig. 3a, b). The peaks of the contour cones represent the reverse points of the Bloch lines and thus the locations of the BPs. The Bloch lines in the biskyrmion are inside the contour cones and terminate at the peaks, while the Bloch lines in the bubble are deeper than the peaks. The height of the contour cones can be defined as the depth of the corresponding BPs. In Fig. 3, the depths of the MP and the AMP are from the top and bottom surfaces, respectively.

Comparing Fig. 3a with Fig. 3b shows that the difference of the 3D structures between the biskyrmion and the bubble is mainly due to the relative position of the MP-AMP pair. In other words, a biskyrmion forms in the overlapping depth of the MP and the AMP, while a bubble exists in the gap between the MP and the AMP. These findings can also be confirmed by the dependence of the topological charge \(Q\) of 2D spin textures in the xy-plane on the perpendicular position \(z\), as shown in Fig. 3c, d. The surface of the 3D structures of both the bubble and the biskyrmion have a topological charge of 1, because of Néel-caps. Such structures through the thickness thus can dominate the measurement of the THE. As a result, even topologically trivial bubbles can exhibit the THE. Meanwhile, the topological charge is different near the middle plane. Therefore, the topological charge changes in two transition positions through the thickness. A change of the topological charge through the thickness has to be attributed to a BP. The transition positions of topological charges agree with the locations of BPs depicted by the blue shades, which provides compelling evidence for the role of the MP-AMP pair in the formation of biskyrmions.

It should be noted that biskyrmions have two polarized cores as shown in Fig. 3e. As a result, the polarized core of the 3D structure of the biskyrmion is a torus with two holes rather than a tube, as demonstrated in Fig. 3f.

In addition, it can be inferred that the motion of the MP-AMP pair accounts for the transition from biskyrmions to bubbles derived from the increase of the external field. Increasing the field lifts BPs towards the surfaces and expands the zero-Q thickness. In summary, biskyrmions and bubbles are topologically different as 2D structures, while their 3D structures are topologically equivalent. In such a 3D model, biskyrmions can be transformed into bubbles smoothly via the motion of BPs along the thickness direction. Also, this 3D structure is topologically nontrivial because of the existence of the BPs. This result emphasizes the significance of the 3D aspect of topological objects in magnetism.

**Formation of biskyrmions under zero field**

We have demonstrated that the relative position of the MP-AMP pair is the key factor to form biskyrmions. The remaining problem is the origin of different locations of the MP-AMP pair in the 3D structures between biskyrmions and bubbles. The location of the biskyrmion in Fig. 2a in the film (Supplementary Fig. 1) inspires us to investigate the effect of film edges. After confirming the edge effect in another simulation (Supplementary Fig. 3), we create biskyrmions in geometry-confined films, which is also the common case in application. As a result, two biskyrmions are generated in a rectangle film under zero field with the easy axis tilted at an angle of 15°, displayed in Fig. 4a.

It turns out that type-I bubbles are favorable in confined geometry. Therefore, a \(y\)-component field is introduced to produce type-II bubbles at first. These bubbles are created by relaxing the randomly magnetized state at an external magnetic field of \((0, 0.05, 0.14)\) T. Then the \(y\)-component field \(B_y\) is removed with steps of 0.01 T, and the state is relaxed in every step. After this, the \(z\)-component field \(B_z\) is removed likewise. The elimination of \(B_y\) leads to enlargement of bubbles and transformation into biskyrmions. Figure 4b shows the variation of \(Q\) versus \(z\) during removing \(B_y\), demonstrating the process of biskyrmion transition. Also, relaxing the bubble state under zero field can directly obtain quite similar structures (Supplementary Fig. 4), indicating that the history of field decreasing has little influence on the formation of biskyrmions. In contrast, eliminating the external field below a certain threshold value makes bubbles and biskyrmions turn into stripe domains if the tilted angle is large as in Fig. 2.

To clarify the mechanism, we inspect the transition process of the left biskyrmion in Fig. 4a during removing \(B_y\). As can be seen in Fig. 4b, the variation of the dependence of \(Q\) on \(z\) indicates the motion of the MP-AMP pair during this process. To address the origin of the MP-AMP pair’s motion, the variations of energy terms are presented in Fig. 4c. The anisotropy energy \(E_A\) and exchange
energy $E_x$ only increase slightly. The change of $B_z$ makes the Zeeman energy $E_z$ increase, while the magnetostatic energy $E_D$ is the only decreasing term. Apart from $E_z$, the total energy is reduced via $E_D$. Therefore, it can be concluded that the magnetostatic energy dominates the transition process. Furthermore, the distribution of magnetostatic energy density $e_D$ on the domain wall shown in Fig. 4d, e reveals the relation between the MP-AMP pair and $E_D$. Specifically, the energy distribution above the AMP is larger than that below the AMP in Fig. 4d. As $B_z$ decreases, the AMP is driven up to reduce the magnetostatic energy as in Fig. 4e, accompanied by the enlargement of the biskyrmion. This mechanism can also account for the observed edge effect, which can be attributed to the influence of the demagnetizing field. On the other hand, the formation of biskyrmions may be triggered by other effects via influencing BPs, such as magnetic impurities.

**DISCUSSION**

Although this study should be confirmed in experiments eventually, it sheds light on the actual structure of experimentally observed biskyrmions. The simulations are performed with parameters same to experimental materials and reproduce TEM images quite well. Therefore, it is plausible that the 3D model is the actual structure corresponding to the observed biskyrmions. The question whether the 2D structures at the middle plane are biskyrmions is less important since the 3D structures of biskyrmions and bubbles are topologically equivalent.

In terms of experiments, the THE may still be a convenient approach to confirm our findings since reconstructing 3D magnetic structures by imaging techniques is technologically difficult. In detail, the dependence of the THE on the external field can be observed while the number of bubbles remains constant. Quantitative experiments can be performed with geometry-confined films to confirm the existence of biskyrmions. In addition, the presence of MP-AMP pairs introduces an in-plane emergent field, which could affect the measurement of the THE.

In conclusion, we find the intrinsic MP-AMP pair mediates the formation of biskyrmion bubbles by inspecting their 3D structure. Biskyrmion bubbles and type-II bubbles can form in a magnetic film with tilted uniaxial anisotropy and produce similar images under Lorentz TEM. In fact, biskyrmions and bubbles share a unified 3D structure with nontrivial topology, in which the relative position of an intrinsic MP-AMP pair dominates the topological...
charge at the middle plane of the film. It is found that the MP-AMP pair can be driven by the film edge and this effect can be attributed to the demagnetizing field. Therefore, it can be assumed that biskyrmion bubbles can be generated in many materials because it only requires geometry-defined ferromagnetic films with tilted uniaxial anisotropy.

The 3D model in this work may inspire both fundamental research and application development. The capability of encoding data using the binary topological charge at the middle plane of a unified 3D object can lead to architectures of data storage devices and other spintronics applications. On the other hand, this work demonstrates a formation mechanism of 2D topological structures induced by the motion of 3D topological defects in contrast to the creation or annihilation of such defects. These findings about biskyrmion bubbles may enlighten the creation of standard biskyrmions since the transition from standard biskyrmions to biskyrmion bubbles is non-topological and can be induced by modulating experimental parameters. Moreover, the 3D structure of these bubbles is the same class of particle-like excitations as the chiral bobber, composed of a magnetization field and magnetic singularities. These results draw attention to the three-dimensional aspect of magnetic nanostructures and the role of emergent MPs in their formation, thus opening avenues to study topological structures in ferromagnetic systems.

DATA AVAILABILITY
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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FIG. 4 Transition from bubbles into biskyrmions. a Biskyrmions at the middle plane of a rectangle film. The sample is defined as 1 μm × 0.5 μm × 160 nm size with cells of 4 nm × 4 nm × 4 nm. The magnetic easy axis lies in the yz-plane. b The dependence of the topological charge \( Q \) on the perpendicular position of the left biskyrmion in a during the process of removing \( B_z \). c The variations of different energy terms \( \Delta E \) during the process of removing \( B_z \). The initial values at \( B_z = 0.14 \) T are subtracted from the energy terms for better illustration. \( E_Z, E_A, E_X, \) and \( E_D \) denote Zeeman, anisotropy, exchange, and magnetostatic energy, respectively. d, e The distribution of magnetostatic energy density \( \epsilon_D \) on the contour surface with \( m_z = 0 \) at \( B_z = 0.14 \) T and 0 T, respectively. Gray dots depict the location of the AMPs.
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AUTHOR CONTRIBUTIONS
C.-J.W. designed the work and performed the numerical simulations and calculations. C.-J.W., P.W., Y.Z., W.W., and F.S. wrote the manuscript. J.D. supervised the research. All authors contributed to the discussions of the results.

COMPETING INTERESTS
The authors declare no competing interests.

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