

Stabilization and Observation of Large-area Ferromagnetic Bimeron Lattice

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1 **Abstract**

2 Symmetry engineering is an effective approach for generating emergent phases and
3 quantum phenomena. In magnetic systems, the Dzyaloshinskii-Moriya (DM)
4 interaction is essential for stabilizing chiral spin textures. The symmetry manipulation
5 of DM vectors, described in three dimensions, could provide a strategy towards creating
6 abundant topologically magnetic phases. Here, we have achieved to break the rotational
7 and mirror symmetries of the three-dimensional DM vectors in a strongly correlated
8 ferromagnet, which were directly measured through the non-reciprocal spin-wave
9 propagations in both in-plane and out-of-plane magnetic field geometries. Combining
10 cryogenic magnetic force microscopy and micromagnetic simulations, we discover a
11 bimeron phase that emerges between spin spiral and skyrmion phases under an applied
12 magnetic field. Such an artificially manipulated DM interaction is shown to play a
13 critical role in the formation and evolution of the large-area bimeron lattice, a
14 phenomenon that could be realized across a broad range of materials. Our findings
15 demonstrate that symmetry engineering of the DM vectors can be practically achieved
16 through epitaxial strain, paving the way for the creation of diverse spin topologies and
17 the exploration of their emergent functionalities.

18

1 Exploration of topological spin textures in real space not only provides a platform
2 to study the fundamental structures and emergent phenomena of solitons in physics [1-
3 3], but also offers potential applications as ideal information units for beyond-CMOS
4 logic and storage devices [4-6]. This has drawn significant attention from researchers
5 across physics, materials science, and information technology [7-9]. Among these spin
6 textures, magnetic skyrmions are particularly notable [10,11]. Their stabilization is
7 largely governed by the Dzyaloshinskii-Moriya interaction (DMI) alongside the
8 interplay of magnetocrystalline anisotropy, Zeeman energy, and magnetostatic energy.
9 Since the discovery of DMI in ferromagnetic materials, systems that majorly exhibit
10 interfacial-like [12,13] and bulk-like DMI [13-15] have been observed, which results
11 in the formation of Néel-type and Bloch-type skyrmions as shown in Fig. 1(a) and 1(b),
12 as well as merons [16-24], hopfions [25-27], chiral bobbars [28,29] and skyrmion
13 strings[30,31], to name a few.

14
15 In physics, emergent phases or phenomena often arise when symmetry is reduced
16 or broken [32]. In ferromagnets with DMI, the DM vectors can be described in the x -,
17 y -, and z -directions of a three-dimensional Cartesian coordinate system [33], so that the
18 elements and operations of crystal symmetry can therefore be used to describe the
19 symmetry of the DMI [34,35]. In the majority of cases, the DM vectors that generate
20 the topological spin textures [9,10,14-29] adhere to a relatively high degree of
21 symmetry. A natural question arises: can reducing or breaking the symmetry of the DM
22 vectors create opportunities for novel topological variants in magnets? In 2006,

1 Bogdanov *et al.* predicted that lower symmetry of DM vectors may bring about more
2 non-trivial spin configurations [34]. Recent experiments have further suggested that
3 reducing symmetry of DM vectors (i.e., anisotropic DMI) may stabilize exotic spin
4 textures such as antiskyrmions [36-38]. Theoretical studies reveal that DM vectors
5 without rotational and mirror symmetries could also give rise to the emergent
6 topological magnetic textures such as bimeron as shown in Fig. 1(c), which may possess
7 extraordinary dynamic properties under excitation compared to classical skyrmions
8 [33,39]. Therefore, manipulating the symmetry of DM vectors may thus open the door
9 to a much richer variety of spin topologies, as summarized in Fig. S1 and Table SI (for
10 detail see Supplementary Information Section S2 [40]).

11

12 Despite the theoretical interests in stabilizing topological variants such as bimeron
13 lattices through controllable symmetry of DM vectors in three dimensions,
14 experimental realization still remains a challenge. Recently, Zhang *et al.* demonstrated
15 that graded strain can effectively break the spatial inversion symmetry of crystals [41],
16 which brings about a hybrid DMI in three dimensions and offers the possibility of
17 engineering the desired symmetry of DM vectors. In this study, we show that DM
18 vectors without rotational (C_n) and mirror (σ) symmetries can be artificially designed
19 in a ferromagnetic oxide thin film, (La,Sr)MnO₃ (LSMO). Such a DMI are formed by
20 the flexoresponse of the magnetic order with the strain gradient [42]. We directly
21 observe the formation and evolution of a large-area bimeron lattice under varying
22 external magnetic field, tracking its emergence from a spin spiral and its subsequent

1 transformation into a skyrmion lattice.

2

3 In general, the DM vectors of magnetic materials can be described by the
4 following tensor (1) [41]

$$5 \quad \mathbf{D} = \begin{bmatrix} \mathbf{D}_x \\ \mathbf{D}_y \\ \mathbf{D}_z \end{bmatrix} = \begin{bmatrix} D_{xx} & D_{xy} & D_{xz} \\ D_{yx} & D_{yy} & D_{yz} \\ D_{zx} & D_{zy} & D_{zz} \end{bmatrix} \quad (1)$$

6 where D_{ij} with $i = x, y, z$ represents the j th component of the DM vector for
7 the spin pairs along [100], [010], and [001] axes, respectively. In the case of non-
8 uniform strain, $D_{ij} = v_{ijmlk} E_{ml,k}$ where v_{ijmlk} are the corresponding flexo-responsive
9 coefficients of the strain-gradient components $E_{ml,k}$. Apart from the conventional DM
10 components in the interfacial-like and bulk-like DMI (D_{xx} , D_{yy} , D_{zz} , D_{xy} , D_{yx}), the
11 existence of z -components (D_{xz} , D_{yz} , D_{zx} , D_{zy}) is essential for achieving complete
12 control over the symmetry of DM vectors in three dimensions. This provides a strong
13 motivation to experimentally explore the configuration of DM components within the
14 LSMO films and further deterministically control the symmetry of DM vectors by
15 modulating epitaxial strain. For this purpose, a high-quality LSMO film with a strain
16 relaxation and thickness of ~ 150 nm was epitaxially grown on NdGaO₃ as shown in
17 Figs. S2 and S3 (for structural and magnetic characterization details see Supplementary
18 Information Section S3 [40]).

19

20 The nonreciprocity of spin-wave propagation is usually used as an identification
21 of the existence and amplitude of DMI [43-47]. In order to evaluate in-plane and out-
22 of-plane DM components in LSMO film with a graded strain, spin-wave propagation

1 in a fully in-plane and out-of-plane magnetized LSMO film was characterized using a
 2 vector network analyzer in combination with a microwave probe (Supplementary
 3 Information Section S1 and S4 [40]). The detailed geometries for measuring the in-
 4 plane (D_{yx}) and out-of-plane (D_{xz}) components are illustrated in Figs. 2 and S4 [40].
 5 For the former, the propagating spin wave along the y -axis was measured under
 6 magnetic saturation along the x -axis (also called Damon-Eshbach, DE mode). With an
 7 applied in-plane field of $\mu_0 H_{\text{ext}} = 140$ mT [Fig. 2(a)], frequency nonreciprocity of
 8 $\delta f_{yx}(k) = f_{\text{DE}}(+k) - f_{\text{DE}}(-k) \approx -0.06$ GHz ($\mathbf{H} \parallel x$) is observed [Fig. 2(c)], which
 9 is proportional to D_{yx} according to the equation (2)

$$10 \quad f_{\text{DE}}(\pm k, H_{\text{ext}}) = \frac{\gamma \mu_0}{2\pi} \sqrt{(Jk^2 - \xi + H_{\text{ext}} + M_s)(Jk^2 + \xi + H_{\text{ext}})} \mp \frac{\gamma k}{\pi M_s} D_{yx} \quad (2)$$

11 where $J = 2A/\mu_0 M_s$ and $\xi = M_s(1 - e^{-2|k|d})/4$ with A being the exchange
 12 constant, M_s the saturation magnetization, μ_0 the vacuum permeability, γ the
 13 gyromagnetic ratio, d the thickness, k the wave vector, and H_{ext} the external field.
 14 The value of the in-plane DM component $D_{yx} = 1.41 \times 10^{-4}$ J m⁻² is estimated by
 15 using the material parameters and $k = 2.07$ rad μm^{-1} .

16

17 Furthermore, the out-of-plane DM component D_{xz} can also lead to a frequency
 18 nonreciprocity in the magnetostatic forward volume (FV) spin waves. Therefore, we
 19 extend our non-local measurement to the FV modes. To the best of our knowledge, this
 20 has remained unexplored due to the absence of materials systems that exhibit an out-
 21 of-plane D_{xz} component. The experimental setup has been designed with an
 22 electromagnet that can be rotated axially to provide a magnetic vector field. The

1 propagating spin wave along the x -axis is then measured under magnetic saturation
 2 along the z -axis in an out-of-plane magnetic field, as shown in Fig. 2(b). During the
 3 measurement, the external magnetic field $\mu_0 H_{\text{ext}} = 550$ mT is applied along the z -axis
 4 for magnetic saturation. The magnetic hysteresis loop can be found in Fig. S3. A
 5 frequency nonreciprocity $\delta f_{xz} = f_{\text{FV}}(+k) - f_{\text{FV}}(-k) \approx 0.04$ GHz is observed [Fig.
 6 2(d)]. The D_{xz} component is estimated through equation (3) [48]

$$7 \quad f_{\text{FV}}(\pm k, H_{\text{ext}}) = \frac{\gamma \mu_0}{2\pi} \sqrt{(Jk^2 + \zeta + H_{\text{ext}} + M_{\text{eff}})(Jk^2 + H_{\text{ext}} + M_{\text{eff}})} \mp \frac{\gamma k}{\pi M_s} D_{xz} \quad (3)$$

8 where $\zeta = M_s[1 - (1 - e^{-kd})/kd]$, $M_{\text{eff}} = \mu_0 H_{\text{ani}} - \mu_0 M_s$, and $\mu_0 H_{\text{ani}}$ is the
 9 perpendicular anisotropy field. $D_{xz} = -0.94 \times 10^{-4}$ J m⁻² is estimated for the out-of-
 10 plane DM component. The spin-wave dispersion of FV modes was further simulated,
 11 as shown in Fig. S5, which confirms that this frequency nonreciprocity originates from
 12 the D_{xz} components. More details of the exploration of additional DM components can
 13 be found in Fig. S6 [40]. The comparable values of the in-plane and out-of-plane DM
 14 components clearly reveal a symmetry configuration of DM vectors in the LSMO film,
 15 e.g. a breaking of rotational (C_n) and mirror (σ) symmetries of the DM vectors as
 16 illustrated in Fig. 1(c) [33]. Such a DM configuration therefore provides an ideal
 17 platform for stabilizing more emergent topological magnetic textures.

18

19 To investigate the possible existence of emergent topological magnetic phases and
 20 their evolutions in real space, we employed cryogenic magnetic force microscopy
 21 (MFM) to image the magnetic domains under varying external magnetic fields (for
 22 detail see Supplementary Information Section S1 [40]). At 3.6 K, a spin spiral state,

1 confirmed by sinusoidal profile of the MFM phase (Fig. S8) [49], was observed in the
2 absence of an applied magnetic field, as shown in the top panel of Fig. 3(a). The
3 intensity of the MFM signal along the dashed path is plotted in the middle panel. It
4 shows the same spiral characteristic as observed at room temperature, exhibiting an
5 excellent periodicity over a large area. This periodicity has been characterized by MFM
6 and resonant elastic x-ray scattering (REXS) and is shown in Fig. S7 [40]. When a
7 magnetic field over -0.12 T was applied perpendicular to the film surface, a decrease in
8 the intensity of MFM contrast was observed, suggesting a reduction of the local out-of-
9 plane magnetization and an appearance of an “intermediate” spiral state as displayed in
10 Fig. S9 [40]. When a perpendicular magnetic field of -0.3 T was applied, the
11 “intermediate” spin spiral phase forms completely [Fig. 3(b)]. Both spiral states share
12 a similar periodicity as confirmed by Fast Fourier Transform (FFT) analysis, as shown
13 in the bottom panels of Fig. 3(a) and 3(b).

14
15 When the applied field strength exceeds -0.4 T, an unexpected magnetic phase
16 emerges, characterized by pairs of semicircles with reversed out-of-plane
17 magnetization cores, completely forming as a triangular arrangement [Fig. 3(c)].
18 Further increasing the external magnetic field strength to -0.465 T results in a transition
19 to the skyrmion phase [Fig. 3(d)]. Notably, the FFT pattern of this emergent magnetic
20 phase displays a distorted hexagonal periodicity, corresponding the arrangement in real
21 space, as shown in dashed circles in the top panel of Fig. 3(c). It is distinct from the
22 symmetric hexagonal periodicity characteristic of the skyrmion phase, as shown in the

1 bottom panels of Fig. 3(c) and 3(d). This lattice of paired semicircles, exhibiting a
2 periodic arrangement, is an emergent phase rarely observed in magnets with traditional
3 DMI [11,50]. The detailed evolution, including an analysis of the corresponding
4 magnetic structures and periodicities as a function of magnetic field, is presented in Fig.
5 S10 [40].

6

7 To better understand the nature and origin of the emergent magnetic phase with
8 reversed cores and associated chirality, micromagnetic simulations were conducted. For
9 the simulations, the DM vectors with both out-of-plane and in-plane components like
10 Fig. 1(c) were assumed [33]. The magnetic textures were simulated using
11 MUMAX3[51] with the out-of-plane DM component taken into account [33]. The
12 value of the parameters adopted in the simulation can be found in the Supplementary
13 Information Section S1 [40]. As plotted in Fig. 4, all the experimentally observed
14 phases are reproduced. The comparison between simulated results in Fig. 4(a) and 4(b)
15 suggests that the transition of the both spiral states as observed in Fig. 3(a) and 3(b),
16 corresponding to the relative shrinking of two up/down magnetized domains under the
17 perpendicular magnetic field. For the emergent phase with paired semicircles, the
18 magnetization distribution of each bi-core unit exhibits a spin texture consistent to that
19 of the predicted ferromagnetic bimeron [33]. The ferromagnetic bimeron lattice will
20 further evolve into the magnetic skyrmion phase at increased magnetic fields. As shown
21 in Fig. 4(c) and 4(d), the number of bimerons is equal to that of skyrmions, indicating
22 the one-by-one transformation between these two topological spin textures. The spin

1 configuration at the top layer is shown in Fig. 4(a)-(d) bottom panel. Those at the middle
2 and bottom layers are shown in Fig. S11. Thus, the detailed magnetization distribution
3 of the observed bimeron lattice is further analyzed. Figure 4(f) reveals the combination
4 of two specific chirality along the x - and y -axes in a bimeron as illustrated in Fig. 4(e),
5 induced by the in-plane and out-of-plane DM components, i.e., D_{yx} and D_{xz} ,
6 respectively.

7
8 It is also noteworthy that, simulations with only interfacial DMI or without any DMI
9 cannot be the case in the present observation, as shown in Fig. S12. With this, we
10 conclude that the bimeron phase are driven by the three-dimensional DM vectors. This
11 symmetry engineering of DM vectors facilitates the large-area stabilization of the
12 bimeron lattice observed experimentally as shown in Fig. S13 [40]. The temperature-
13 magnetic-field phase diagram [Fig. S14], which captures intermediate states, provides
14 critical insights into understanding this transition and filling a previously unexplored
15 gap in the exploration of the bimeron phase that occurs between spiral and skyrmion
16 states. Our findings propose a practical strategy for tailoring magnetic textures by
17 designing and controlling symmetry of DM vectors. Graded strain offers a precise
18 method for engineering them via the interplay of crystal structure (e.g., rhombohedral,
19 tetragonal, orthorhombic, monoclinic) and the strain configuration (e.g., tensile,
20 compressive, and shear strain, and their relaxations)[41,42]. A deterministic control of
21 the lattice, such as the relaxation of shear strain or anisotropic biaxial strain, may
22 independently break the rotational or mirror symmetry of the DM vectors and generate

1 further variants of magnetic textures. This approach may extend beyond bimerons and
2 skyrmions to a broader spectrum of magnetic phases, enabling the on-demand design
3 of topological magnetic textures.

4

5

6 **Conclusions**

7 In summary, a three-dimensional configuration of DM vectors, breaking rotational
8 and mirror symmetries, was artificially designed in a strain-graded LSMO thin film.

9 This was quantitatively characterized by the frequency non-reciprocity of the spin-wave
10 propagation. Such a DM configuration facilitated the stabilization of a large-area

11 ferromagnetic bimeron lattice, which was directly observed during the phase evolution
12 from a spin spiral to skyrmion state. Micromagnetic simulations highlighted the crucial

13 role of the DM configuration with both out-of-plane and in-plane DM components in
14 shaping this emergent topological spin texture. Our results demonstrate that artificially

15 designed symmetry of DM vectors and its associated topological variants can be
16 extended to a wide range of materials. The large-area bimeron phase observed in this

17 strongly correlated oxide provides a novel platform for exploring emergent
18 functionalities and quantum phenomena in both ground and excited states.

19

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15

16 **Author contributions**

17 M.C., Y.Z. and J.Z. conceived and designed the experiments. D.S. and J.R. were
18 responsible for the thin film fabrication. M.C., Z.L., performed structural and magnetic
19 characterization. M.C. and X.B. performed magnetic force microscopy. M.C., D.S., Y.Z.
20 and H.Y. designed and fabricated the CPWs and performed the spin-wave propagation
21 measurements. S.W., L.Q. and K.S. performed the micromagnetic simulation. S.Z.,
22 M.K. and N.J. were responsible for the designs or measurements of resonant elastic x-
23 ray scattering. J.Z. supervised the experimental study, and H.Y., M.K., N.J., G.L. and
24 T.H. provided related discussion. K.S. supervised the theoretical and simulated study.
25 M.C., S.W., Y.Z., K.S. and J.Z. wrote the paper. M.K., S.Z., N.J., G.L. and T.H.
26 contributed to the revision.

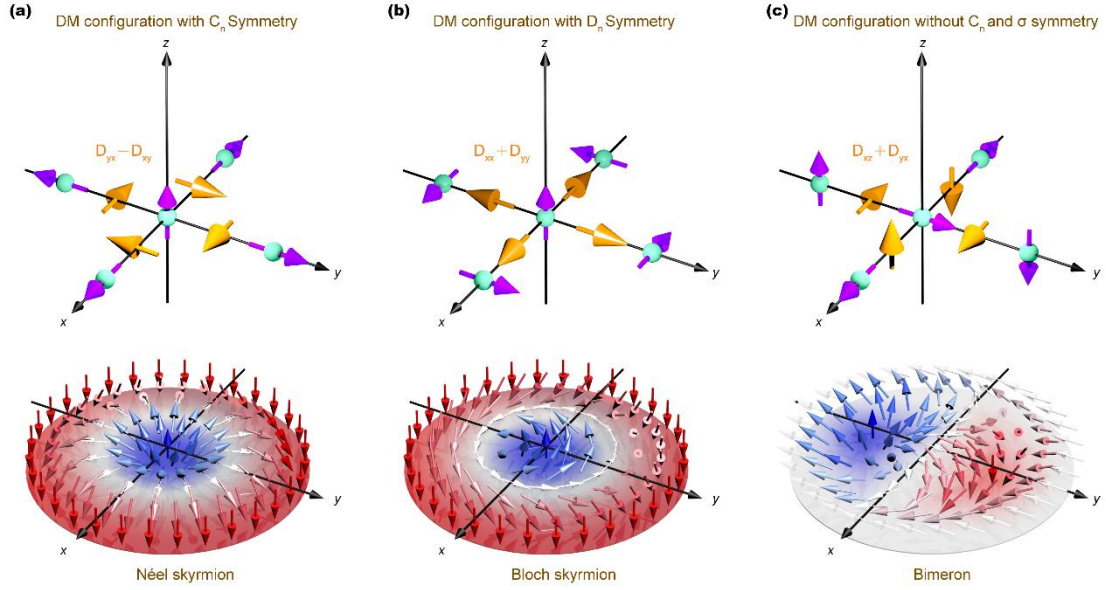
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8 of DM configurations and related magnetic textures; Section S3: structural and magnetic
9 characterizations; Section S4: nonreciprocal spin-wave spectra; Section S5: MFM image and REXS
10 characterization of the ground state magnetic order; Section S6: MFM analysis of the evolution of
11 the magnetic textures; Section S7: micromagnetic simulations of magnetic structures; Section S8:
12 large-area stabilization of bimeron lattice and the detailed phase diagram.
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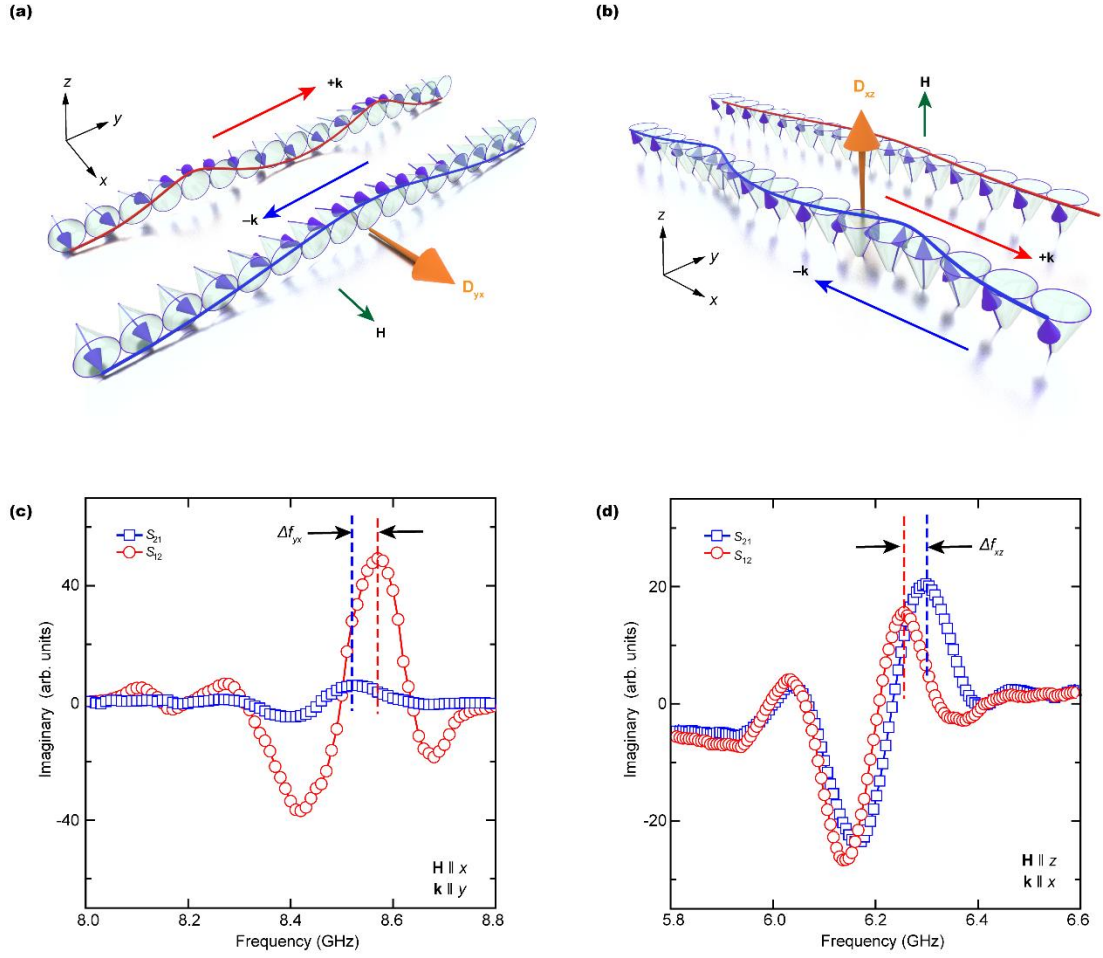
1 Figures



2

3 **FIG. 1. DM configurations with different symmetries and corresponding energy-**
 4 **preferred topological spin textures. (a), (b)** Classical DM configurations with
 5 rotational (C_n) or mirror (σ) symmetries and corresponding energetically stabilized Néel
 6 and Bloch-type skyrmions, respectively. (c) Three-dimensional DM configuration
 7 without C_n and σ symmetries and its associated bimeron state. Schematics of DM vector
 8 distributions are shown in the top panels and the detailed magnetic textures are depicted
 9 in the bottom panels.

10



1

2 **FIG. 2. Coexisting spin-wave frequency nonreciprocities induced by in-plane and**

3 **out-of-plane DM components. (a), (b) Schematics of in-plane and out-of-plane DM**

4 **vectors and their corresponding nonreciprocal spin-wave propagation, respectively. The**

5 **DM component D_{yx} and D_{xz} are shown by orange arrows. The corresponding spin-wave**

6 **vector $\pm\mathbf{k}$ (blue and red arrows) aligns along the y -axis (x -axis), and the external**

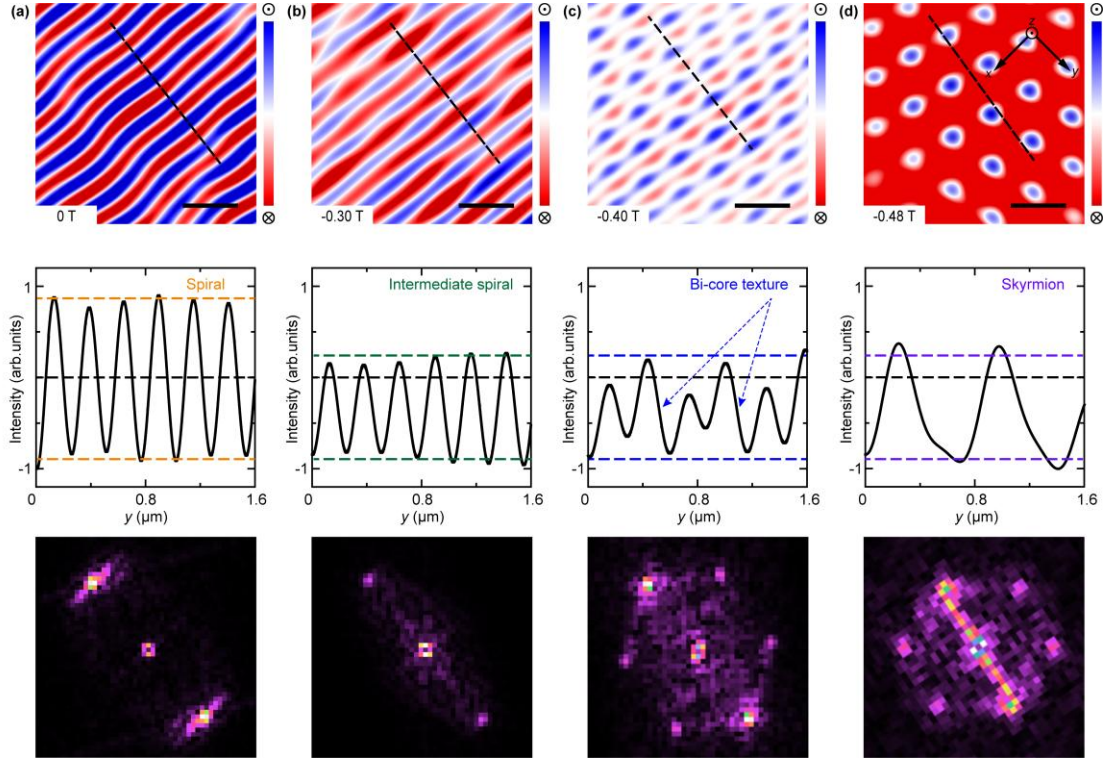
7 **magnetic field \mathbf{H} (green arrows) aligns along the x -axis (z -axis), respectively. (c), (d)**

8 **Experimental transmission spectra of nonreciprocal spin-wave propagation at room**

9 **temperature under $\mathbf{H} \parallel x$ and $\mathbf{H} \parallel z$ configurations, respectively. Spin waves in the**

10 **transmission spectrum S_{21} ($+\mathbf{k}$) propagate from coplanar waveguide 1 (CPW1) to CPW2,**

11 **while the S_{12} ($-\mathbf{k}$) spectrum describes propagation in the reverse direction.**



1

2 **FIG. 3. Field-dependent evolution of spin textures. (a)-(d)** Real-space images of spin

3 textures under varying out-of-plane external fields (z -axis), captured by magnetic force

4 microscopy (MFM) imaging (top panel). The dashed circles highlight the periodicity of

5 the bi-core lattice. All scale bars represent 500 nm. The middle panels show line plots

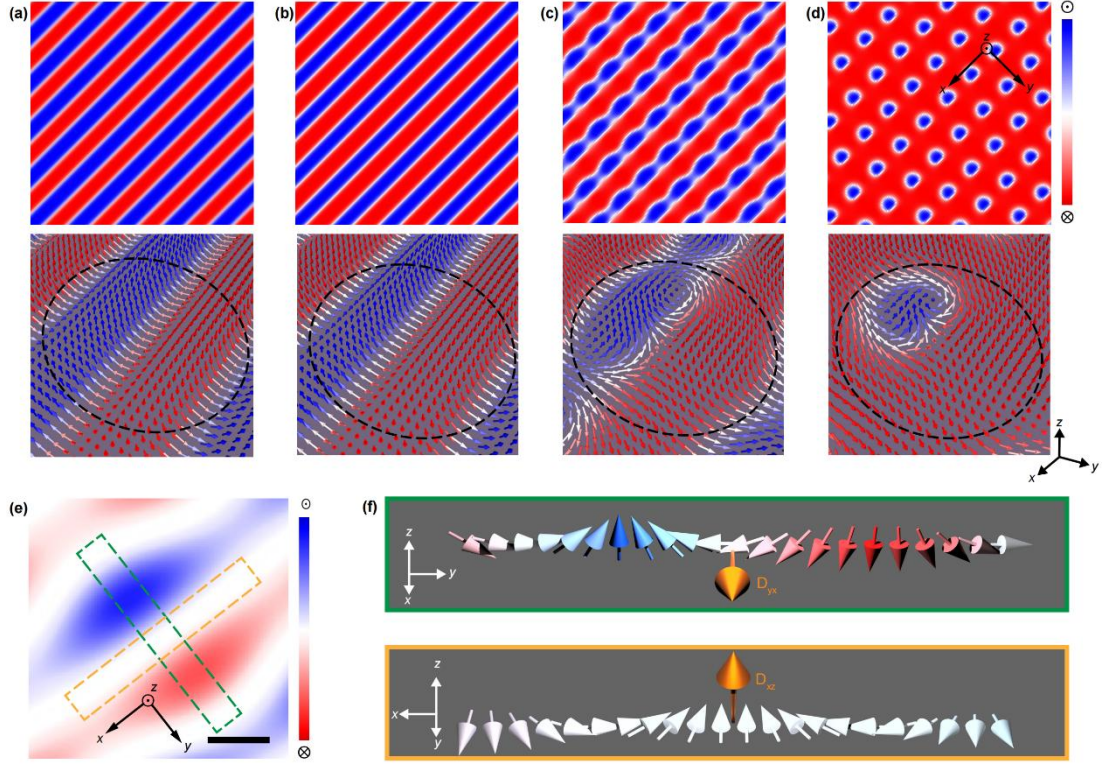
6 along the y -axis, as indicated by the respective dashed lines above. The normalized

7 stray field intensity from the local magnetization highlights the intensity differences

8 between the spiral phase, intermediate spiral, bi-core, and skyrmion phase. The FFT

9 patterns of the different magnetic textures, which illustrate their periodicities, are

10 displayed in the bottom panels.



1

2 **FIG. 4. The appearance of ferromagnetic bimeron lattice driven by in-plane and**

3 **out-of-plane DM vectors. (a)-(d) The simulated lattice states of spiral, intermediate**

4 **spiral, bimeron and skyrmion textures. Their corresponding spin arrangements at the**

5 **top layer are presented in detail by colorful arrows at the bottom and the black circles**

6 **point out the in-situ transformations of the above textures. (e) The unit of bimeron from**

7 **experimental observation. Yellow and green boxes indicate that the chiralities of spin**

8 **arrangements along the x - and y -axes are different. The scale bar is 250 nm. (f) The**

9 **different spin chiralities in ferromagnetic bimeron. The spin arrangement along the y -**

10 **axis (green box in e) is shown at the top and exhibits out-of-plane chirality, which is**

11 **induced by the in-plane DM vector D_{yx} . In contrast, the spin arrangement along the x -**

12 **axis (yellow box in e) is shown at the bottom and exhibits in-plane chirality, which is**

13 **induced by the out-of-plane DM vector D_{xz} .**