



HAL
open science

Electric-field-induced multiferroic topological solitons

Arthur Chaudron, Zixin Li, Aurore Finco, Pavel Marton, Pauline Dufour,
Amr Abdelsamie, Johanna Fischer, Sophie Collin, Brahim Dkhil, Jirka
Hlinka, et al.

► **To cite this version:**

Arthur Chaudron, Zixin Li, Aurore Finco, Pavel Marton, Pauline Dufour, et al.. Electric-field-induced multiferroic topological solitons. *Nature Materials*, inPress, 10.1038/s41563-024-01890-4 . hal-04604964

HAL Id: hal-04604964

<https://hal.science/hal-04604964>

Submitted on 7 Jun 2024

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

1 **Electric-field induced multiferroic topological solitons**

2

3 Arthur Chaudron¹, Zixin Li², Aurore Finco³, Pavel Marton^{4,5}, Pauline Dufour¹, Amr

4 Abdelsamie^{1,3}, Johanna Fischer¹, Sophie Collin¹, Brahim Dkhil⁶, Jirka Hlinka⁴, Vincent Jacques³,

5 Jean-Yves Chauleau², Michel Viret², Karim Bouzehouane¹, Stéphane Fusil^{1,7*}, Vincent Garcia^{1*}

6

7 *¹Laboratoire Albert Fert, CNRS, Thales, Université Paris-Saclay, 91767 Palaiseau, France*

8 *²SPEC, CEA, CNRS, Université Paris-Saclay, 91191 Gif-sur-Yvette, France*

9 *³Laboratoire Charles Coulomb, Université de Montpellier and CNRS, 34095 Montpellier, France*

10 *⁴Institute of Physics, Academy of Sciences of the Czech Republic, Na Slovance 2, 18221 Praha*

11 *8, Czech Republic*

12 *⁵Institute of Mechatronics and Computer Engineering, Technical University of Liberec,*

13 *Studentská 2, 461 17 Liberec, Czech Republic*

14 *⁶Université Paris-Saclay, CentraleSupélec, CNRS, Laboratoire SPMS, 91190, Gif-sur-Yvette,*

15 *France*

16 *⁷Université d'Evry, Université Paris-Saclay, 91000 Evry, France*

17

18 *email: stephane.fusil@cnsr-thales.fr, vincent.garcia@cnsr-thales.fr

19

20

21 **Topologically-protected spin whirls in ferromagnets are foreseen as the carthorse of**
22 **solitonic information technologies. Nevertheless, the future of skyrmionics may rely on**
23 **antiferromagnets due to their immunity to dipolar fields, straight motion along the driving**
24 **force, and ultrafast dynamics. While complex topological objects were recently discovered**
25 **in intrinsic antiferromagnets, mastering their nucleation, stabilization, and manipulation**
26 **with energy-efficient means remains an outstanding challenge. Designing topological polar**
27 **states in magnetoelectric antiferromagnetic multiferroics would allow one to electrically**
28 **write, detect and erase topological antiferromagnetic entities. Here we stabilize**
29 **ferroelectric center states using a radial electric field in multiferroic BiFeO₃ thin films. We**
30 **show that such polar textures contain flux closures of antiferromagnetic spin cycloids, with**
31 **distinct antiferromagnetic entities at their cores depending on the electric-field polarity. By**
32 **tuning the epitaxial strain, quadrants of canted antiferromagnetic domains can also be**
33 **electrically designed. These results open the path to reconfigurable topological states in**
34 **multiferroic antiferromagnets.**

35

36

37 Topological polar objects are now emerging¹ with experimental demonstrations of polar
38 vortices, skyrmions and center states in ferroelectric-based superlattices^{2,3}, thin films^{4,5} and
39 nanostructures⁶⁻⁸. These entities can be scaled down to nanometer sizes, controlled with
40 energy-efficient electric fields, and detected via non-destructive resistive readout, making
41 them interesting for information technologies and in particular memory applications. Among
42 them, topological center-type domains in ferroelectric thin films are currently garnering
43 attention for their stability, switchability and electrical conduction properties^{9,7,6,10,8,11}.

44 Topological defects here refer to local singularity regions in an ordered medium wherein the

45 order parameter ceases to vary continuously¹². Both center-convergent (head-to-head) or
46 center-divergent (tail-to-tail) polar states are characterized by a finite winding number of +1
47 in a two-dimensional projection^{6,12}, which might provide some topological protection.
48 However, the lack of a full rotation of polarization in the third dimension prevents the
49 topological charge to be ± 1 , as is the case for skyrmions in the homotopy group $\pi_2(S^2)$. Such
50 center convergent or divergent polar textures have been reported in spatially confined
51 nanoscale structures such as as-grown pillars in the bottom-up approach^{7,10}, or in etched
52 islands in the top-down approach⁸ in BiFeO₃ thin films. They were also shown to display
53 different electrical states, with resistances contrasting by several orders of magnitude^{7,8,11},
54 paving the way for ferroelectric topological resistive memories. However, the behavior of their
55 coupled antiferromagnetic order remains an uncharted territory.

56 Controlling spin textures in antiferromagnetic materials is intrinsically challenging due to their
57 insensitivity to magnetic fields. This has been reported to be feasible, taking advantage of the
58 magnetoelectric coupling¹³, in the room-temperature multiferroic BiFeO₃, harboring
59 ferroelectricity and a G-type antiferromagnetic order superimposed with a spin cycloid^{14,15}. A
60 comprehensive description of the BiFeO₃ magnetic ordering can be found in the review by
61 Burns et al.¹⁶ and references therein. Indeed, ferroelectric textures in BiFeO₃ thin films have
62 been shown to imprint their antiferromagnetic order^{13,17,18}. Depending on epitaxial strain,
63 electric fields were shown to either provide a deterministic control of the spin cycloid
64 propagation¹³ or to change the antiferromagnetic order from canted collinear G-type textures
65 to spin cycloids¹⁷.

66 Here we report on the coupled polar and antiferromagnetic topological nanoscale textures
67 induced by electric-field pulses in submicron devices based on BiFeO₃ thin films under various
68 epitaxial strains. When subjected to compressive epitaxial strain, we show how ferroelectric

69 center domains yield flux closures of antiferromagnetic spin cycloids, while magnetic
70 simulations give credence to the occurrence of antiferromagnetic singularities at their very
71 center. When subjected to tensile epitaxial strain, we show how those same ferroelectric
72 center domains lead to four-quadrant homogeneous canted antiferromagnetic spin textures.
73 In both cases, we observe a “one-to-one” imprint of the ferroelectric domains into the
74 antiferromagnetic ones, albeit multiple antiferromagnetic states are in principle energetically
75 degenerate for a single ferroelectric domain. This “simple” magnetoelectric coupling holds
76 promise for devices based on topology in multiferroic BiFeO₃, allowing deterministic electrical
77 switching of antiferromagnetic ordering.

78 As previously reported¹¹, applying radial electric fields between a patterned top electrode and
79 a conductive tip (Fig. 1b) has proven to be an effective and reproducible method to induce
80 ferroelectric center states in (001)-oriented BiFeO₃ thin films. Four-quadrants center-
81 convergent (Fig. 1a) and center-divergent (Fig. 1c) polar textures can be written by electric
82 pulses of opposite signs.

83 Conversely to bulk, where cycloids can propagate along three energetically-equivalent
84 crystallographic directions coupled to the ferroelectric polarization¹⁹, this degeneracy is lifted
85 by epitaxial strain in such thin films and the cycloid propagating purely in the film plane is
86 selected¹³. Such a behavior implies that any rotation of the ferroelectric polarization imprints
87 an equivalent rotation of the cycloid propagation direction, while preserving the clockwise
88 (Fig. 1d), or counter-clockwise (Fig. 1g) rotation of the antiferromagnetic spins alongside it, if
89 a single magnetic polarity is considered²⁰. Indeed, a convenient way to describe the spin
90 cycloid in BiFeO₃ is to refer to its magnetic polarity²⁰: this quadratic order parameter is defined
91 as $\lambda = k \times S_i \times S_j$, where S_i and S_j are spins on adjacent i and j sites along the cycloidal
92 propagation direction k . This magnetic polarity unambiguously determines the sense of

93 rotation or handedness of the spin cycloid. It couples to the polarization P through an energy
94 term proportional to $\lambda \cdot P$ in the Landau expansion. Incidentally, a single magnetic polarity
95 (antiparallel to the polarization vector) has been detected within a defined polar domain
96 experimentally, in both single crystals²¹ and thin films^{20,22}. Hence, we will in the following
97 consider the magnetic polarity vector to be antiparallel to the polarization within each
98 ferroelectric variant. Thus, all symmetries pertaining to ferroelectric textures should hold for
99 their antiferromagnetic counterparts, and it becomes possible to rely on the magnetoelectric
100 coupling to induce antiferromagnetic topological states anchored on ferroelectric center
101 domains.

102 Considering these polar topological textures and the magnetoelectric coupling, we ran
103 atomistic simulations of the spin textures for the upwards center-convergent and downwards
104 center-divergent ferroelectric states. In both cases, in addition to the pristine cycloidal
105 rotation of the spins within single domains, another full in-plane rotation is induced at the
106 core of the four-quadrant states, corresponding to the antiferromagnetic equivalents of
107 hedgehog vortices (Fig. 1f) and antivortices (Fig. 1e)²³. These unique spin textures are confined
108 at the junction of several unidirectional k -vectors, which are an essential prerequisite to the
109 emergence of antiferromagnetic skyrmion-like features that have not been reported yet.
110 Controlling these antiferromagnetic singularities with an electric-field is a paramount asset of
111 BiFeO_3 .

112

113 **Results**

114 **Polar mapping and writing**

115 Since the ferroelectric textures are defined as fields of 3D vectors, the need arises to display
116 the dataset in the simplest way to showcase the nature of the electrically-poled polar
117 landscape. In this mindset, Fig. 2 provides insights on the process used to extract a polar map
118 from PFM phase and amplitude measurements. Similar processing and visualization of PFM
119 data has already been used in the literature^{6,8,24} and coined as “vector PFM”.

120 In this typical example, a ferroelectric center convergent domain state was stabilized in a 800-
121 nm-wide BiFeO₃ device confined by a Pt circular electrode (Fig. 2a), when the AFM tip was
122 placed at the center of the device to apply a radial electric field. The vertical piezoresponse
123 force microscopy (VPFM) indicates a uniform signal, corresponding to a polarization pointing
124 upwards (Fig. 2b). The lateral piezoresponse force microscopy (LPFM) responses show a
125 splitting of the bare BiFeO₃ disk into two halves when the cantilever is aligned vertically (Fig.
126 2c), as well as when the cantilever is aligned horizontally (Fig. 2d). The LPFM signals collected
127 at different angles allow the reconstruction of the vector polarization map from the local
128 direction and magnitude of the LPFM responses triggered by electrical excitation in this
129 peculiar design lacking any bottom electrode (a few volts AC bias is applied between the
130 surrounding electrode and the grounded tip to trigger the piezoresponse, see Fig. 1b).

131 The underlying ferroelectric polar texture is approximated by projecting the 3D piezoresponse
132 vectors to the closest crystallographic axes allowed for the dipole moment. The resulting
133 ferroelectric domain mappings are illustrated by the overlaid color scales (Fig. 2e-f). Some
134 examples of full lateral piezoresponse characterizations, obtained before applying the
135 azimuthal angle projection, are provided in Supplementary Fig. 1. Taking advantage of this
136 systematic data processing, all further atomistic magnetic simulations will be based on such

137 polar vector maps, deduced from actual experimentally-designed and measured ferroelectric
138 textures.

139

140

141 **Flux-closure antiferromagnetic spin cycloids**

142 When subjected to -0.35% compressive strain, i.e. thanks to epitaxial growth on DyScO₃
143 substrates, BiFeO₃ has been shown to retain its bulk-like antiferromagnetic spin cycloid^{13,17,18}.

144 In the pristine state, all thin films studied here exhibit a stripy ferroelectric texture consisting
145 of two alternating polarization variants (Fig. 3a) separated by 71° domain walls, and
146 corresponding to the two favored ferroelastic variants. This anisotropy stems from the
147 monoclinic distortion transferred from the (110) scandate substrate to BiFeO₃ (Ref. 25). Each
148 domain is harboring a single cycloid propagating in the film plane and in a direction k_1 (green
149 double arrows in Fig. 3d), and giving rise to the zig-zag magnetic pattern (Fig. 3g), as previously
150 reported^{13,17,18}. The aforementioned magnetic polarity λ is defined within each ferroelectric
151 domain as a green arrow antiparallel to the polarization (Fig. 3d). The two other, non-pre-
152 existing and least stable, ferroelastic variants that are created in center states (Fig. 3b-c) are
153 prone to relax to some extent. However, the four quadrant textures' cores, which contain the
154 topological information, were found to be stable for months. Fig. 3b is a typical example of a
155 divergent center state that underwent minor relaxation, restricted to the edges of the device.
156 The two switchable states are expected to yield opposite rotations of the spin cycloid in all
157 quadrants (Fig. 3e-f), as a single magnetic polarity has been demonstrated for a given
158 polarization²⁰⁻²². This induces two distinct senses of rotation and for the antiferromagnetic
159 spin cycloids around the texture centers.

160 The corresponding magnetic stray field maps, as deduced from scanning nitrogen-vacancy (NV)
161 magnetometry (Methods, Supplementary Fig. 2), are displayed in Fig. 3h-i and nicely
162 corroborate the overall expected antiferromagnetic patterns by displaying four cycloidal
163 directions, rotated one from the other by 90° at each domain wall. These results are
164 highlighting the sharp correlation between ferroelectric and antiferromagnetic domains,
165 whose respective walls overlap perfectly within the resolution limits of these near-field
166 experiments.

167 Supplementary Fig. 3 presents similar magnetometry measurements performed on center-
168 divergent ferroelectric states that were induced on identical disk patterns through the same
169 writing process, attesting for such textures' reproducibility and stability. Although the
170 stitching of antiferromagnetic spin cycloids at domain walls appears to vary from one device
171 to another, the propagation direction of those cycloids in each quadrant is fully deterministic
172 and was found to remain stable, at least for weeks.

173 Moreover, Fig. 3j-k show simulated results for the antiferromagnetic spin textures coupled to
174 the actual center polar states. As previously mentioned, the underlying ferroelectric states
175 were initialized based on the experimental polar domain maps provided by PFM
176 measurements (as in Fig. 2). Scanning NV magnetometry images are reproduced quite reliably,
177 at least regarding their overall topology, since the stitching at domain walls was found to
178 depend on the initial antiferromagnetic state used in the simulations. The insets provide a
179 zoom-in on individual spins around the cores of the two center polar states and give credence
180 to the local stabilization of two distinct antiferromagnetic singularities, although their
181 existence cannot be corroborated experimentally due to a lack of resolution.

182

183

184 **Pseudo-collinear G-type antiferromagnetic quadrants**

185 When subjected to +0.50% tensile strain, i.e. thanks to epitaxial growth on SmScO₃ substrates,
186 BiFeO₃ has been shown to destabilize its bulk-like spin cycloid, yielding a pseudo-collinear G-
187 type antiferromagnetic ordering^{17,26,27}. In BiFeO₃, the investigation on the local coupling
188 between ferroelectric polarization and pseudo-collinear G-type antiferromagnetic domains
189 has been very shallow, with no clear experimental report to the best of our knowledge. It was
190 still reported that the antiferromagnetic vector L_i in such domains could only lie along three
191 energetically-equivalent crystallographic directions coupled to the ferroelectric polarization²⁸.
192 Those three directions happen to be the same as the aforementioned bulk-like spin cycloid
193 propagation directions that belong to the pseudo-cubic (111) plane perpendicular to the
194 polarization (Supplementary Fig. 4a)²⁸. In turn, when the cycloid is destroyed, the second
195 Dzyaloshinskii-Moriya interaction, related to the oxygen octahedra tilts along the polarization
196 direction²⁹, gives rise to a net canted magnetization $\pm M_i$ along the direction perpendicular to
197 both the antiferromagnetic Néel vector L_i and the ferroelectric polarization P
198 (Supplementary Fig. 4b-c)^{26,28}. Taking its sign into account, this weak moment can then lie
199 along six possible orientations within a given ferroelectric domain.

200 The pristine polar landscape of the films grown on SmScO₃ is similar to the previously
201 described compressive strain case (Fig. 4a). Fig. 4d shows how the pseudo-collinear G-type
202 antiferromagnetic domains overlap their ferroelectric counterparts in as-grown BiFeO₃ under
203 tensile strain. It seems that the net magnetization stays uniform within each ferroelectric
204 monodomain, and exhibits two distinct alternating configurations coupled to the ferroelectric
205 stripes. Micromagnetic simulations were run considering a uniform net magnetization
206 pointing in any of the 6 permitted crystallographic directions within each experimentally-
207 measured ferroelectric domain (Supplementary Note 1). This allows us to compute the

208 magnetic stray field landscape radiated at the NV center height for all possible canted moment
209 configurations, and to identify the ones experimentally observed. The results are depicted in
210 the schematics of Fig. 4g, indicating that both ferroelectric variants contain the same magnetic
211 variant M_3 (more information in Supplementary Fig. 5). However, this local one-to-one
212 correspondence behavior does not always yield at larger scale, where patches of different
213 alternating configurations may be observed (Supplementary Fig. 6). Comparing these large-
214 scale NV images with simulations (Supplementary Fig. 7), we identify up to two magnetic
215 variants M_3 and M_2 being harbored in each ferroelectric domain.

216 Nonetheless, the local “one-to-one” magnetoelectric coupling was found to be retained
217 through electrical writing of center domains, as exhibited by Fig. 4e-f, where scanning NV
218 magnetometry images show either the highest field or highest field gradient at the
219 approximate location of the ferroelectric domain walls (more information in Supplementary
220 Fig. 8 and Supplementary Fig. 9). Both center-convergent (Fig. 4b) and center-divergent (Fig.
221 4c) polar textures thus imprinted a single antiferromagnetic domain over each of their
222 quadrants. In order to identify the actual canted moment configurations, magnetic-field
223 simulations were again performed from all 6^4 configurations a priori permitted by the center
224 domains. Out of all those simulated configurations, a single one qualitatively matched
225 scanning NV magnetometry measurements for the downwards center-divergent ferroelectric
226 texture, and is illustrated in Fig. 4h. Supplementary Fig. 8 presents the comparison of the
227 measured and simulated magnetic field along the three cartesian axes for this specific net
228 magnetization configuration.

229 This matching process was found to be more cumbersome for the downwards center-
230 convergent ferroelectric texture. Supplementary Fig. 9 displays the comparison between the
231 measured and simulated magnetic field projected for the most compatible magnetization

232 configuration displayed in Fig. 4i. Other simulated magnetic configurations were found to
233 radiate a magnetic field in qualitative agreement with the measured one (Supplementary Fig.
234 10 and Supplementary Fig. 11). Fig. 4g-i sum up the multiferroic topological states deduced
235 from simulations for both the as-grown ferroelectric stripes as well as the written divergent
236 and convergent center states.

237

238 Repeating the writing process identically on multiple disk-shaped devices did not necessarily
239 yield identical net magnetization configurations. The magnetoelectric coupling then appears
240 less deterministic for the G-type antiferromagnetic ordering than for the bulk-like cycloidal
241 one. However, evidence suggests that the net magnetization direction lies along either $\pm[-211]$
242 or $\pm[1-21]$ (for a ferroelectric polarization oriented along $[111]$), and actually never occurs
243 along the $\pm[11-2]$ variant. In terms of their respective associated antiferromagnetic vectors, it
244 means that only the out-of-plane $[0-11]$ or $[10-1]$ are favored, and the $[-110]$ one lying strictly
245 in the thin film plane is discarded (Supplementary Fig. 4). This is in line with Landau-Ginzburg
246 theory predicting that compressive epitaxial strain should favor spins in the (001) film plane
247 while tensile strain leads to out-of-plane spins²⁶.

248 We now focus on the nature of the antiferromagnetic domain boundaries embedded in these
249 different polar textures. The identification of the M domains in each polar configuration allows
250 us to determine the direction of the Néel vectors. As we cannot distinguish antiphase
251 boundaries, we define L_i as green double arrows in Fig. 4g-i. In the as-grown striped domain
252 structure, the ferroelectric domain walls always coincide with antiferromagnetic domain walls,
253 defining them as multiferroic domain walls³⁰. In addition, pure antiferromagnetic domain
254 walls can be identified within the ferroelectric stripes (Supplementary Fig. 12a). In the electric-
255 field induced center polar states, these multiferroic domain walls are accompanied by pure

256 ferroelectric domain walls through which the antiferromagnetic Néel vector is preserved
257 (Supplementary Fig. 12b-c). These observations of purely ferroelectric, purely
258 antiferromagnetic, or multiferroic domain walls are reminiscent of the seminal papers from
259 M. Fiebig et al. on other multiferroic compounds³⁰⁻³².
260 Thus, although tensile strain promoting pseudo collinear antiferromagnetism prevents a fully
261 deterministic control of the canted antiferromagnetic state via the magnetoelectric coupling,
262 it still allows for the discrimination of several net magnetization configurations, with a rich
263 zoology of embedded antiferromagnetic domains and domain walls, paving the path for
264 further study of this magnetoelectric coupling in such exotic and largely unexplored G-type
265 antiferromagnetic thin films with different crystalline orientations.

266

267 **Discussion**

268 In this work, we reported the successful design and writing of two different families of
269 multiferroic topological entities. The first set was obtained on BiFeO₃ epitaxial thin films
270 subjected to compressive strain, and consist in antiferromagnetic vortices induced by spin
271 cycloids coupled to ferroelectric center domains. The second was achieved on similar films
272 subjected to tensile strain, and amount to four-quadrant canted G-type antiferromagnetic
273 domains imprinted from ferroelectric center domains.

274 In the first case, epitaxial strain discriminates the spin cycloids propagation directions
275 permitted by magnetoelectric coupling, and yields only the direction lying strictly in the
276 sample plane. In the second case, it also discriminates the allowed antiferromagnetic vectors,
277 resulting in the prohibition of the one lying strictly in the sample plane.

278 All multiferroic textures discussed in this work were written by simple electric pulses from
279 their as-grown state, and can be switched from the center-convergent to the center-divergent

280 ferroelectric states by reversing the voltage sign. Doing so reverses the spin cycloid vortex
281 rotation in the case of compressive strain, or changes the net four-quadrant magnetization
282 configuration in the case of tensile strain. Such a switchable behavior may lay the foundations
283 for topological multiferroic memories.

284

285 **Acknowledgements**

286 We are grateful to Nicolas Reyren for fruitful discussions. We are thankful for support from
287 the French Agence Nationale de la Recherche (ANR) through the project TATOO (ANR-21-
288 CE09-0033) and ESR/EquipEx+ program e-DIAMANT (Grant No. ANR-21-ESRE-0031), the
289 European Union's Horizon 2020 research and innovation programme under the Grant
290 Agreements No. 964931 (TSAR) and No. 866267 (EXAFONIS). This work is supported by a
291 public grant overseen by the ANR as part of the "Investissements d'Avenir" program (Labex
292 NanoSaclay, reference: ANR-10-LABX-0035). The Sesame Ile de France IMAGE SPIN project (No.
293 EX039175) is also acknowledged.

294

295 **Authors contributions**

296 V.G. and S.F. designed the experiments. V.G. coordinated the work. P.D., A.A. and J.F.
297 synthesized the thin films by pulsed laser deposition. S.F. and S.C. fabricated the devices using
298 e-beam lithography and sputtering deposition. A.C. switched and examined the polar textures
299 using piezoresponse force microscopy. A.C. investigated the coupled spin textures using
300 scanning NV magnetometry with the help of K.B. Z.L., J.-Y.C., and M.V. performed the atomic
301 spin simulations. A.F. and V.J. performed the micromagnetic simulations. A.C., S.F. and V.G.
302 wrote the core of the manuscript. All the authors discussed the results and contributed to the
303 manuscript preparation.

304 **Competing interests**

305 The authors declare no competing interests

306

307 **Figure captions**

308 **Fig. 1. Concept of multiferroic topological states.** **a** and **c**, Sketches illustrating the convergent (**a**) and
309 divergent (**c**) center polar (white arrows) states. **b**, Atomic force microscopy topography of the device
310 geometry with lateral PFM phase signal overlaid. The bright (dark) color corresponds polarization
311 pointing to frontside (back side) as sketched in the inset. Two holes (1 μm diameter in a 30 nm thick
312 Pt line) on the left (right) of the AFM tip were poled with negative (positive) bias, the other holes
313 correspond to pristine domains. **d** and **g**, Sketches illustrating a four-quadrant center polar texture
314 with associated antiferromagnetic clockwise (**d**) and counter-clockwise (**g**) spin rotations (blue and red
315 arrows stand for the antiferromagnetic spins), given the single magnetic polarity of the spin cycloid. **e**
316 and **f**, Zoomed-in computed spin textures at the core of the 'geometrically perfect' center domains
317 showing antiferromagnetic hedgehog antivortices (**e**) and vortices (**f**) for the convergent and divergent
318 cases, respectively. The color scale displays the z-component of the Néel vectors.

319

320 **Fig. 2. Polar maps extracted from piezoresponse force microscopy.** PFM characterization of an
321 upwards center-convergent ferroelectric texture written by a +60 V pulse. **a**, Topography (color bar in
322 nm). **b**, Vertical piezoresponse extracted from the cantilever deflection. **c-d**, Lateral piezoresponse
323 extracted from the cantilever torsion, for different angles of the sample with respect to the cantilever
324 direction. Color bars in b-d in arbitrary units. The insets on the bottom left of a-c show the cantilever
325 orientation during measurement. **e**, 3D sketch of the resulting polar configuration, considering a single
326 polarization in each quadrant. **f**, 2D in-plane piezoresponse vector map of the area indicated by a circle,
327 extracted pixelwise by trigonometric regression on all lateral piezoresponse images measured for
328 different cantilever orientations. Vector lengths and directions show the local magnitude and direction
329 of the surface in-plane deformation. The overlapped color map provides the projection of this direction

330 on the closest crystallographic axes permitted for the ferroelectric polarization. The color bar displays
331 the corresponding angle θ , as defined from the sketch. Scale bars are 200 nm.

332

333 **Fig. 3. Magnetoelectric coupling in ferroelectric center states written on a (001)-oriented 33-nm-**
334 **thick BiFeO₃ thin film under -0.35% compressive strain.** Scale bars are 100 nm. **a-c**, Piezoresponse
335 vector maps and underlying ferroelectric textures for the as-grown (a), downwards center-divergent
336 (b) and upwards center-convergent (c) states. The insets display a 3D sketch of the polar configuration.
337 **d-f**, Schematics of the associated multiferroic topological states. The cycloid propagation vectors are
338 depicted as green double arrows. Considering a single magnetic polarity in each domain results in
339 opposite spin cycloid vortex helicities around the core of center-divergent and center-convergent polar
340 states, as depicted by the center-convergent and center-divergent magnetic polarities λ . **g-i**, Out-of-
341 plane magnetic field radiated by the antiferromagnetic textures from NV magnetometry (dark lines
342 highlight the ferroelectric domain walls). **j-k**, Spin-level simulations of the antiferromagnetic textures,
343 initialized with the ferroelectric domains extracted from experimental measurements. The computed
344 out-of-plane magnetic field is displayed, as well as a zoom in the core configuration of the
345 antiferromagnetic spins.

346

347 **Fig. 4. Magnetoelectric coupling in ferroelectric center states written on a (001)-oriented 32-nm-**
348 **thick BiFeO₃ thin film grown under +0.50% tensile strain.** Scale bars are 100 nm. **a-c**, Piezoresponse
349 vector maps and underlying ferroelectric textures for the as-grown (a), downwards center-divergent
350 (b) and center-convergent (c) states. The insets display a 3D sketch of the polar configuration. **d-f**,
351 Associated magnetic field radiated by the antiferromagnetic textures from NV magnetometry (dark
352 lines highlight the ferroelectric domain walls). **g-i**, Schematics of the resulting multiferroic states: red
353 and purple vectors show the in-plane projections of polarization and the weak moment stemming from
354 the canting of the G-type antiferromagnetic spins. The green double arrows indicate the corresponding

355 Néel vectors. All magnetization configurations allowed by magnetoelectric coupling were simulated
356 and compared, with the ones selected here yielding the best fit to experimental data.

357

358 References

359

- 360 1. Junquera, J. *et al.* Topological phases in polar oxide nanostructures. *Rev Mod Phys* **95**,
361 025001 (2023).
- 362 2. Yadav, A. K. *et al.* Observation of polar vortices in oxide superlattices. *Nature* **530**, 198–201
363 (2016).
- 364 3. Das, S. *et al.* Observation of room-temperature polar skyrmions. *Nature* **568**, 368–372 (2019).
- 365 4. Zhang, Q. *et al.* Nanoscale Bubble Domains and Topological Transitions in Ultrathin
366 Ferroelectric Films. *Adv. Mater.* **29**, 1702375 (2017).
- 367 5. Han, L. High-density switchable skyrmion-like polar nanodomains integrated on silicon. **603**,
368 63–67 (2022).
- 369 6. Kim, K.-E. *et al.* Configurable topological textures in strain graded ferroelectric nanoplates.
370 *Nat. Commun.* **9**, 403 (2018).
- 371 7. Ma, J. *et al.* Controllable conductive readout in self-assembled, topologically confined
372 ferroelectric domain walls. *Nat. Nanotechnol.* **13**, 947–952 (2018).
- 373 8. Yang, W. *et al.* Quasi-one-dimensional metallic conduction channels in exotic ferroelectric
374 topological defects. *Nat. Commun.* **12**, 1306 (2021).
- 375 9. Li, Z. *et al.* High-density array of ferroelectric nanodots with robust and reversibly switchable
376 topological domain states. *Sci. Adv.* **3**, e1700919 (2017).
- 377 10. Han, M.-J. *et al.* Shape and Surface Charge Modulation of Topological Domains in Oxide
378 Multiferroics. *J. Phys. Chem. C* **123**, 2557–2564 (2019).
- 379 11. Yang, W. *et al.* Nonvolatile Ferroelectric-Domain-Wall Memory Embedded in a Complex
380 Topological Domain Structure. *Adv. Mater.* **34**, 2107711 (2022).
- 381 12. Mermin, N. D. The topological theory of defects in ordered media. *Rev. Mod. Phys.* **51**, 591–
382 648 (1979).
- 383 13. Gross, I. *et al.* Real-space imaging of non-collinear antiferromagnetic order with a single-spin
384 magnetometer. *Nature* **549**, 252–256 (2017).
- 385 14. Sosnowska, I., Neumaier, T. P. & Steichele, E. Spiral magnetic ordering in bismuth ferrite. *J.*
386 *Phys. C Solid State Phys.* **15**, 4835–4846 (1982).
- 387 15. Lebeugle, D. *et al.* Electric-Field-Induced Spin Flop in BiFeO₃ Single Crystals at Room
388 Temperature. *Phys. Rev. Lett.* **100**, 227602 (2008).
- 389 16. Burns, S. R., Paull, O., Juraszek, J., Nagarajan, V. & Sando, D. The Experimentalist’s Guide to
390 the Cycloid, or Noncollinear Antiferromagnetism in Epitaxial BiFeO₃. *Adv. Mater.* **32**, 2003711 (2020).
- 391 17. Haykal, A. *et al.* Antiferromagnetic textures in BiFeO₃ controlled by strain and electric field.
392 *Nat. Commun.* **11**, 1704 (2020).
- 393 18. Zhong, H. *et al.* Quantitative Imaging of Exotic Antiferromagnetic Spin Cycloids in BiFeO₃ Thin
394 Films. *Phys. Rev. Appl.* **17**, 044051 (2022).
- 395 19. Finco, A. *et al.* Imaging Topological Defects in a Noncollinear Antiferromagnet. *Phys. Rev.*
396 *Lett.* **128**, 187201 (2022).
- 397 20. Waterfield Price, N. *et al.* Electrical Switching of Magnetic Polarity in a Multiferroic BiFeO₃
398 Device at Room Temperature. *Phys. Rev. Appl.* **8**, 014033 (2017).
- 399 21. Johnson, R. D. *et al.* X-Ray Imaging and Multiferroic Coupling of Cycloidal Magnetic Domains
400 in Ferroelectric Monodomain BiFeO₃. *Phys. Rev. Lett.* **110**, 217206 (2013).
- 401 22. Waterfield Price, N. *et al.* Strain Engineering a Multiferroic Monodomain in Thin-Film BiFeO₃.
402 *Phys. Rev. Appl.* **11**, 024035 (2019).
- 403 23. Thiaville, A., Miltat, J. & Rohart, S. 1 - Magnetism and topology. in *Magnetic Skyrmions and*

404 *Their Applications* (eds. Finocchio, G. & Panagopoulos, C.) 1–30 (Woodhead Publishing, 2021).
405 doi:10.1016/B978-0-12-820815-1.00012-2.
406 24. Park, M. *et al.* Three-dimensional ferroelectric domain imaging of epitaxial BiFeO₃ thin films
407 using angle-resolved piezoresponse force microscopy. *Appl. Phys. Lett.* **97**, 112907 (2010).
408 25. Chen, Z. H., Damodaran, A. R., Xu, R., Lee, S. & Martin, L. W. Effect of “symmetry mismatch”
409 on the domain structure of rhombohedral BiFeO₃ thin films. *Appl. Phys. Lett.* **104**, 182908 (2014).
410 26. Sando, D. *et al.* Crafting the magnonic and spintronic response of BiFeO₃ films by epitaxial
411 strain. *Nat. Mater.* **12**, 641–646 (2013).
412 27. Agbelele, A. *et al.* Strain and Magnetic Field Induced Spin-Structure Transitions in Multiferroic
413 BiFeO₃. *Adv. Mater.* **29**, 1602327 (2017).
414 28. Chauleau, J.-Y., Haltz, E., Carrétéro, C., Fusil, S. & Viret, M. Multi-stimuli manipulation of
415 antiferromagnetic domains assessed by second-harmonic imaging. *Nat. Mater.* **16**, 803–807 (2017).
416 29. Ederer, C. & Spaldin, N. A. Weak ferromagnetism and magnetoelectric coupling in bismuth
417 ferrite. *Phys. Rev. B* **71**, 060401 (2005).
418 30. Fiebig, M., Lottermoser, Th., Fröhlich, D., Goltsev, A. V. & Pisarev, R. V. Observation of
419 coupled magnetic and electric domains. *Nature* **419**, 818–820 (2002).
420 31. Meier, D. *et al.* Topology and manipulation of multiferroic hybrid domains in MnWO₄. *Phys.*
421 *Rev. B* **80**, 224420 (2009).
422 32. Fiebig, M., Lottermoser, T., Meier, D. & Trassin, M. The evolution of multiferroics. *Nat. Rev.*
423 *Mater.* **1**, 16046 (2016).
424
425

426 **Methods**

427

428 **Sample fabrication**

429 BiFeO₃ thin films were grown by pulsed laser deposition on DyScO₃(110) and SmScO₃(110)
430 orthorhombic substrates using a KrF excimer laser (248 nm), as detailed in Ref. 17.

431

432 **Top electrode patterning**

433 The 15-nm-thick Pt electrode was deposited by sputtering after performing an electronic
434 beam lithography to induce nanostructures of various geometries and sizes where BiFeO₃ is
435 left bare. Disk shaped devices were found to host the most accurate and reproducible
436 ferroelectric center domains after electric pulse writing, exhibiting clear monodomains in each
437 of the quadrants, as well as low roughness and straight domain walls. Diameters of around
438 800 nm were used to keep the devices as small as possible while allowing the 200-nm-wide
439 NV-center diamond tip to come in contact with the BiFeO₃ surface over an area large enough
440 for proper imaging.

441

442 **Writing of center domains**

443 Ferroelectric center states were induced through electrical interaction between a conductive
444 tip and the patterned top electrode. While the tip was grounded and stationed in direct
445 contact with BiFeO₃ at the center of an 800 nm disk pattern, a DC pulse of around +50 V or -
446 50 V was applied on the surrounding electrode to write a head-to-head or tail-to-tail center
447 domain respectively. Pulse duration was not found to have any significant impact beyond a
448 few milliseconds, and was therefore set around 100 ms.

449

450 **Piezoresponse force microscopy**

451 The experiments were conducted with an atomic force microscope (Nanoscope V multimode,
452 Bruker) and two external lock-in amplifier detectors (SR830, Stanford Research) for the
453 simultaneous acquisition of lateral and vertical piezoresponses. An external ac source (DS360,
454 Stanford Research) was used to excite the Pt patterned top electrode at a frequency of 35 kHz
455 while the conducting Pt-coated tip was grounded. We used stiff cantilevers (40 N m^{-1}) for
456 accurate vertical detection and softer ones (7 N m^{-1}) for the lateral detection. Regarding the
457 as-grown state, BiFeO_3 on SmScO_3 exhibits a homogeneous vertical signal, corresponding to a
458 polarization pointing upwards, i.e. away from the substrate, while BiFeO_3 on DyScO_3 exhibits
459 a quasi-homogeneous vertical signal, corresponding to a polarization pointing downwards, i.e.
460 towards the substrate. All images were obtained with phase shifts close to 180 degrees
461 between in-plane domains, as well as between out-of-plane domains.

462

463 **Scanning NV magnetometry**

464 Experiments were performed using a commercial scanning N-V magnetometer (Qnami,
465 ProteusQ) operated under ambient conditions, with commercial all-diamond scanning-probe
466 tips containing single NV defects (Qnami, Quantilever MX). The tip was integrated into a
467 tuning-fork based atomic force microscope combined with a confocal microscope optimized
468 for single NV defect spectroscopy. Magnetic fields radiated by the sample are detected by
469 recording the Zeeman shift of the NV defect's electronic spin sublevels through optical
470 detection of the electron spin resonance.

471 The scanning NV magnetometer was operated in the full-B imaging mode by continuously
472 monitoring the photoluminescence (PL) intensity and sweeping the applied microwave
473 frequency (ν) at each point of the scan. A Lorentzian fit on $\text{PL}(\nu)$ then provides the

474 aforementioned local resonance frequency. Experiments were performed with a NV-to-
475 sample distance of approximately 50 nm and a bias magnetic field of 1 mT applied along the
476 NV quantization axis. Thermal drift was a significant source of error in our slow full-B
477 quantitative measurements, therefore all textures were also imaged in the faster dual-iso-B
478 mode to confirm the overall patterns with limited drift.

479

480 **Extraction of 3D magnetic field**

481 Full-B NV-center magnetometry provides a quantitative measure of the magnetic field
482 radiated at the defect's height and along the defect's symmetry axis. Making use of Maxwell's
483 equations in air in the absence of currents and time-dependent electric fields, $\nabla \cdot \mathbf{B} = 0$ and
484 $\nabla \times \mathbf{B} = \mathbf{0}$, one can compute the two remaining components of B perpendicular to the
485 symmetry axis. This computation was simplified by assuming a uniform absolute height of the
486 NV-center over the whole measured area. The resulting 3D magnetic field was then projected
487 along all three sample axes, and either all projections or the most relevant one are displayed
488 here, although it should be noted that a single projection already holds the full information.

489

490 **Data noise filtering**

491 Most piezoresponse and magnetometry images were filtered by Gaussian kernels whose
492 standard deviations were kept lower than the about 10 nm lateral resolution of the scanning
493 probe microscopes. Such filters were applied on the X-Y coordinates of the piezoresponse
494 before converting them into R- θ .

495

496 **Four-quadrant spin cycloid simulations**

497 Simulating antiferromagnetic textures on a large scale remains a challenging task because of
498 the large number of involved magnetic moments. Indeed, they have to be performed on the
499 atomic level in order to grasp a realistic picture of their magnetic properties. For this work,
500 atomic spin simulations have been performed using a GPU based home-made code. This code
501 is based on solving the Landau-Lifschitz-Gilbert equation for every atomic magnetic moment.
502 In order to reproduce the BiFeO₃ spin textures, the effective Hamiltonian includes all relevant
503 energy terms as described in Ref. 33. The main ones are: the direct antiferromagnetic
504 exchange which favors the antiparallel alignment of spins, the magneto-electric coupling
505 which promotes the stabilization of the antiferromagnetic cycloid, the “alternating” second
506 DMI term which is responsible for the emergence of the spin density wave. Besides, two
507 anisotropy terms are also needed namely a uniaxial easy-axis anisotropy long direction of the
508 electric polarization, i.e. one of the [111] directions, and a weaker 6-fold anisotropy in the
509 perpendicular plane. All parameters have been taken from Ref. 34. However, for the sake of
510 computing time, the magnetoelectric amplitude has been increased by a factor 10 which leads
511 to a shortening of the period of antiferromagnetic cycloid. It therefore allowed to simulate
512 smaller volumes, yet keeping a qualitative agreement.

513 In the present work, the simulations have been performed for a 120 nm × 120 nm square (in
514 the [100] and [010] directions respectively) and for a thickness of 0.78 nm (in the [001]
515 direction) which corresponds to 303 × 303 × 2 atomic magnetic moments. Noteworthy, while
516 boundary conditions are periodic in the [001] direction, they are let free in the in-plane
517 directions. Indeed, in the presence of the spin cycloid, periodic boundary conditions in these
518 directions could be more detrimental than free boundary condition and non-negligibly affect
519 the cycloid properties. In our case, the borders are far enough so that the region of interest is
520 unaffected. The 4-quadrant electrical landscape is reproduced by defining four regions with

521 corresponding orientations of the magneto-electric term and the DMI term. The initialization
522 conditions are the expected antiferromagnetic cycloid properties far from the ferroelectric
523 domain walls which are subsequently relaxed to obtained

524 In order to compare the atomic simulations to the scanning NV magnetometry images, the
525 magnetic fringing field has been computed in a standard way. However, the period of the
526 antiferromagnetic spin cycloid being 10 times shorter, the magnetic field has been mapped at
527 a 5 nm height from the sample surface, i.e. typically 10 times closer than in the NV-center
528 measurements.

529

530 **Four-quadrant net magnetization simulations**

531 The simulations of the magnetic stray field resulting from the G-type antiferromagnetic
532 quadrants were done assuming that only the canted moments are contributing (more
533 information in Supplementary Note 1). These moments can have 3 possible directions with
534 respect to P, as shown in Supplementary Fig. 4, and considering that their sign is unknown, we
535 end up to 6 possible directions of M in each quadrant. Finally, the whole number of possible
536 configurations is thus 6^4 .

537 The magnetic state used for each simulation corresponds to 4 domains with boundaries
538 extracted from the PFM data, and in each domain one of the 6 possible orientations of M for
539 the associated direction of P.

540 The stray field maps are then obtained by separating the magnetic sample in 5 nm x 5 nm cells
541 and summing up the contribution of each cells at every position of the map, using the method
542 described in Ref. 35.

543

544 **Sample and device statistics**

545 Piezoresponse force microscopy data displayed in Fig. 3a-c for the as-grown, center-divergent,
546 center-convergent polar states, respectively, were collected on three different devices of the
547 same $5 \times 5 \text{ mm}^2$ Pt/BiFeO₃/DyScO₃ sample. The very same devices were imaged by scanning
548 NV magnetometry in Fig. 3g-i. Piezoresponse force microscopy data displayed in Fig. 4a-c for
549 the as-grown, center-divergent, center-convergent polar states, respectively, were collected
550 on three different devices of the same $5 \times 5 \text{ mm}^2$ Pt/BiFeO₃/SmScO₃ sample. The very same
551 devices were imaged by scanning NV magnetometry in Fig. 4d-f. The polar configurations
552 displayed in Fig. 1, 2, 3, 4 were reproduced on at least 20 devices for the two different samples.
553
554

555 **Data availability**

556 The data that support this work are available in <https://zenodo.org/records/10875992>.

557

558 **Methods-only references**

- 559 33. Li, Z. *et al.* Multiferroic skyrmions in BiFeO₃. *Phys. Rev. Res.* **5**, 043109 (2023).
560 34. Fishman, R. S. The microscopic model of BiFeO₃. *Phys. B* **536**, 115–117 (2018).
561 35. Abert, C., Selke, G., Kruger, B., & Drews, A. A Fast Finite-Difference Method for
562 Micromagnetics Using the Magnetic Scalar Potential. *IEEE Trans. Magn.* **48**, 1105–1109 (2012).
563







