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

2

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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 BiFeO<sub>3</sub> 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

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37 Topological polar objects are now emerging<sup>1</sup> with experimental demonstrations of polar  
38 vortices, skyrmions and center states in ferroelectric-based superlattices<sup>2,3</sup>, thin films<sup>4,5</sup> and  
39 nanostructures<sup>6-8</sup>. 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<sup>9,7,6,10,8,11</sup>.

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

45 order parameter ceases to vary continuously<sup>12</sup>. 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<sup>6,12</sup>, 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  $\pm 1$ , 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<sup>7,10</sup>, or in etched  
52 islands in the top-down approach<sup>8</sup> in BiFeO<sub>3</sub> thin films. They were also shown to display  
53 different electrical states, with resistances contrasting by several orders of magnitude<sup>7,8,11</sup>,  
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 coupling<sup>13</sup>, in the room-temperature multiferroic BiFeO<sub>3</sub>, harboring  
59 ferroelectricity and a G-type antiferromagnetic order superimposed with a spin cycloid<sup>14,15</sup>. A  
60 comprehensive description of the BiFeO<sub>3</sub> magnetic ordering can be found in the review by  
61 Burns et al.<sup>16</sup> and references therein. Indeed, ferroelectric textures in BiFeO<sub>3</sub> thin films have  
62 been shown to imprint their antiferromagnetic order<sup>13,17,18</sup>. Depending on epitaxial strain,  
63 electric fields were shown to either provide a deterministic control of the spin cycloid  
64 propagation<sup>13</sup> or to change the antiferromagnetic order from canted collinear G-type textures  
65 to spin cycloids<sup>17</sup>.

66 Here we report on the coupled polar and antiferromagnetic topological nanoscale textures  
67 induced by electric-field pulses in submicron devices based on BiFeO<sub>3</sub> 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 BiFeO<sub>3</sub>, allowing deterministic electrical  
77 switching of antiferromagnetic ordering.

78 As previously reported<sup>11</sup>, 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 BiFeO<sub>3</sub> 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 polarization<sup>19</sup>, this degeneracy is lifted  
85 by epitaxial strain in such thin films and the cycloid propagating purely in the film plane is  
86 selected<sup>13</sup>. 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 considered<sup>20</sup>. Indeed, a convenient way to describe the spin  
90 cycloid in BiFeO<sub>3</sub> is to refer to its magnetic polarity<sup>20</sup>: 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 crystals<sup>21</sup> and thin films<sup>20,22</sup>. 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)<sup>23</sup>. 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  $\text{BiFeO}_3$ .

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<sup>6,8,24</sup> and coined as “vector PFM”.

120 In this typical example, a ferroelectric center convergent domain state was stabilized in a 800-  
121 nm-wide BiFeO<sub>3</sub> 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 BiFeO<sub>3</sub> 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 DyScO<sub>3</sub>  
143 substrates, BiFeO<sub>3</sub> has been shown to retain its bulk-like antiferromagnetic spin cycloid<sup>13,17,18</sup>.

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 BiFeO<sub>3</sub> (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<sup>13,17,18</sup>. The aforementioned magnetic polarity  $\lambda$  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 polarization<sup>20-22</sup>. 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  $90^\circ$  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 SmScO<sub>3</sub> substrates,  
186 BiFeO<sub>3</sub> has been shown to destabilize its bulk-like spin cycloid, yielding a pseudo-collinear G-  
187 type antiferromagnetic ordering<sup>17,26,27</sup>. In BiFeO<sub>3</sub>, 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 polarization<sup>28</sup>.  
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)<sup>28</sup>. In turn, when the cycloid is destroyed, the second  
195 Dzyaloshinskii-Moriya interaction, related to the oxygen octahedra tilts along the polarization  
196 direction<sup>29</sup>, 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)<sup>26,28</sup>. 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 SmScO<sub>3</sub> 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 BiFeO<sub>3</sub> 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 spins<sup>26</sup>.

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 walls<sup>30</sup>. 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 compounds<sup>30-32</sup>.  
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 BiFeO<sub>3</sub> 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

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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  $\mu\text{m}$  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  $\theta$ , 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 BiFeO<sub>3</sub> 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  $\lambda$ . **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 BiFeO<sub>3</sub> 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

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359

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424  
425

426 **Methods**

427

428 **Sample fabrication**

429 BiFeO<sub>3</sub> thin films were grown by pulsed laser deposition on DyScO<sub>3</sub>(110) and SmScO<sub>3</sub>(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 BiFeO<sub>3</sub> 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 BiFeO<sub>3</sub> 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 BiFeO<sub>3</sub> 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 ( $40 \text{ N m}^{-1}$ ) for  
456 accurate vertical detection and softer ones ( $7 \text{ N m}^{-1}$ ) for the lateral detection. Regarding the  
457 as-grown state,  $\text{BiFeO}_3$  on  $\text{SmScO}_3$  exhibits a homogeneous vertical signal, corresponding to a  
458 polarization pointing upwards, i.e. away from the substrate, while  $\text{BiFeO}_3$  on  $\text{DyScO}_3$  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 ( $\nu$ ) 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- $\theta$ .

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 BiFeO<sub>3</sub> 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/BiFeO<sub>3</sub>/DyScO<sub>3</sub> 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/BiFeO<sub>3</sub>/SmScO<sub>3</sub> 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**

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