



HAL
open science

Electric and antiferromagnetic chiral textures at multiferroic domain walls

J.-Y. Chauleau, T. Chirac, S. Fusil, Vincent Garcia, Mohammad Waseem Akhtar, J. Tranchida, P. Thibaudeau, I. Gross, C. Blouzon, A. Finco, et al.

► **To cite this version:**

J.-Y. Chauleau, T. Chirac, S. Fusil, Vincent Garcia, Mohammad Waseem Akhtar, et al.. Electric and antiferromagnetic chiral textures at multiferroic domain walls. *Nature Materials*, 2020, 19, pp.386-390. 10.1038/s41563-019-0516-z . hal-02909553

HAL Id: hal-02909553

<https://hal.science/hal-02909553v1>

Submitted on 30 Jul 2020

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 and antiferromagnetic chiral textures at multiferroic domain walls

2
3 J.-Y. Chauleau^{1,2}, T. Chirac¹, S. Fusil^{3,8}, V. Garcia³, W. Akhtar⁴, J. Tranchida^{5§}, P. Thibaudeau⁵, I. Gross⁴,
4 C. Blouzon¹, A. Finco⁴, M. Bibes³, B. Dkhil⁶, D.D. Khalyavin⁷, P. Manuel⁷, V. Jacques⁴, N. Jaouen², M.
5 Viret^{1*}

6
7 ¹*SPEC, CEA, CNRS, Université Paris-Saclay, 91191 Gif-sur-Yvette, France.*

8 ²*Synchrotron SOLEIL, 91192 Gif-sur-Yvette, France*

9 ³*Unité Mixte de Physique, CNRS, Thales, Univ. Paris-Sud, Université Paris-Saclay, 91767 Palaiseau,*
10 *France*

11 ⁴*Laboratoire Charles Coulomb, Université de Montpellier and CNRS, 34095 Montpellier, France*

12 ⁵*CEA – DAM le Ripault, BP 16, 37260 Monts, France*

13 ⁶*Laboratoire Structures, Propriétés et Modélisation des Solides, CentraleSupélec, Université Paris*
14 *Saclay, CNRS UMR8580, F-91190 Gif-Sur-Yvette, France*

15 ⁷*ISIS Facility, STFC, Rutherford Appleton Laboratory, Didcot OX11 0QX, UK*

16 ⁸*Université d'Evry, Université Paris-Saclay, Evry, France*

17
18 § : now at 'Multiscale Science Department, Sandia National Laboratories, P.O. Box 5800, MS1322, 87185
19 Albuquerque, NM, United States'

20
21 * e-mail: michel.viret@cea.fr

22 **Chirality, a foundational concept throughout science, may arise at ferromagnetic domain walls¹**
23 **and in related objects such as skyrmions². However, chiral textures should also exist in other types**
24 **of ferroics such as antiferromagnets for which theory predicts that they should move faster for**
25 **lower power³, and ferroelectrics where they should be extremely small and possess unusual**
26 **topologies^{4,5}. Here we report the concomitant observation of antiferromagnetic and electric chiral**
27 **textures at domain walls in the room-temperature ferroelectric antiferromagnet BiFeO₃.**
28 **Combining reciprocal and real-space characterization techniques, we reveal the presence of**
29 **periodic chiral antiferromagnetic objects along the domain walls as well as a priori energetically**
30 **unfavorable chiral ferroelectric domain walls. We discuss the mechanisms underlying their**
31 **formation and their relevance for electrically controlled topological oxide electronics and**
32 **spintronics.**

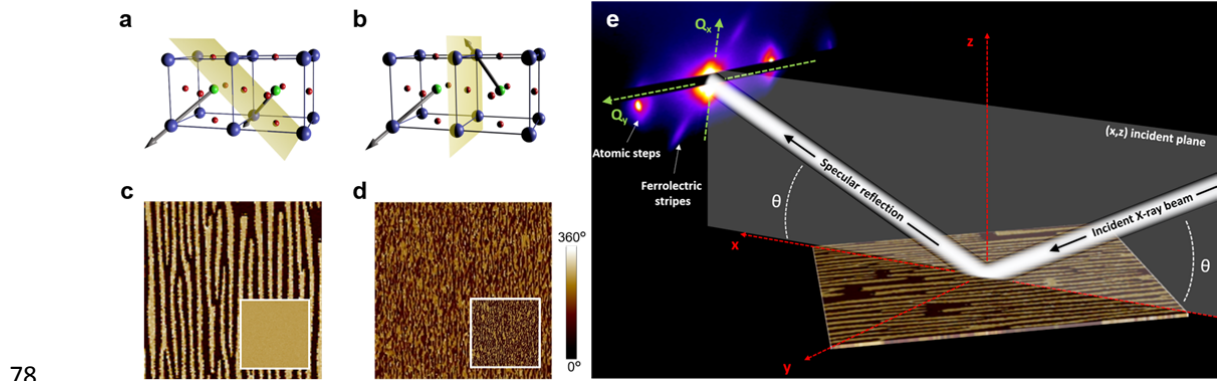
33
34 Metallic ferromagnets have been the elemental bricks of spintronics for the last three decades and
35 continue to hold promises on the basis of non-collinear chiral spin textures such as skyrmions. These
36 topologically protected objects are envisioned to be the future of magnetic data storage thanks to
37 their specific stability, dynamics, and scalability². In parallel, antiferromagnets (AFs) are emerging as a
38 new paradigm for spintronics⁶. They are intrinsically stable (being insensitive to spurious magnetic
39 fields), scalable (no cross talk between neighbouring memory cells), and fast (switching frequencies
40 in the THz regime). The opportunity of gathering the best of these two worlds and realize
41 "antiferromagnetic skyrmions" is then tremendously appealing but faces at least two major
42 challenges. The first one is to achieve antiferromagnetic chirality and the second one is to identify
43 appropriate control stimuli to create, annihilate and move these chiral objects.

44 On one hand, chirality may naturally emerge at domain walls. The antiferromagnetic domain wall
45 structure is a virtually uncharted territory but this is where translational symmetry is broken and spin
46 rotation favoured. On the other hand, AF manipulation is hampered by the intrinsic lack of net
47 magnetization, which prevents a straightforward magnetic actuation. This fundamental issue may be

48 addressed by different strategies. One of them relies on the generation of spin-transfer torque,
 49 mediated by spin currents, to act on the antiferromagnetic order. Antiferromagnetic switching was
 50 indeed recently demonstrated in specific non-centrosymmetric and conducting AFs such as CuMnAs⁷.
 51 However, the vast majority of AFs are insulating, which allows for low operation power and long spin
 52 wave propagation length, although spin-transfer torque remains to be explored in these compounds.

53 An alternative pathway could be to resort to materials known as multiferroics in which
 54 antiferromagnetism coexists with ferroelectricity. Some multiferroics also exhibit a magnetoelectric
 55 coupling between these orders, allowed in materials of certain symmetry classes introduced more
 56 than half a century ago^{8,9}. In addition, multiferroic domain walls (DWs) may exhibit singular
 57 properties such as enhanced conductivity¹⁰ associated with their ferroelectric (FE) order, in line with
 58 other exotic properties found in related ferroelastic or antiferroelectric materials^{11,12}. The ability to
 59 write, erase and control domain walls in such systems is the cornerstone of “*the material is the*
 60 *machine*” concept¹³. Extending it to spintronics would offer opportunities unachievable with
 61 conventional magnetic materials.

62 In this study, we focus on bismuth ferrite¹⁴ BiFeO₃ (BFO), the most prototypical multiferroic and one
 63 of the very few known materials presenting spin and dipole ordered phases well above room
 64 temperature. In addition to possessing a very large remanent electric polarization (100 μC/cm²), BFO
 65 is a G-type antiferromagnet forced to rotate in a long-range (64 nm in the bulk) chiral cycloid because
 66 of the presence of magnetoelectric coupling between the two orders. The competition between
 67 symmetric and asymmetric exchange energies make the spins rotate in the plane defined by the
 68 electric polarization and the propagation vector, with an additional small periodic ferromagnetic
 69 canting of the moments in the transverse direction, in a spin density wave fashion (i.e. non-chiral).
 70 Interestingly, an AF skyrmion lattice is just the ‘multi-*q*’ version of the simple cycloid of bulk BFO, but
 71 this state is not stabilized in the bulk because no energy term plays a role equivalent to the magnetic
 72 field for ferromagnetic skyrmions. Our strategy here is to force a multi-*q* state using the frustration
 73 induced on the magnetic and ferroelectric orders by a high-density network of ferroelectric stripe
 74 domains. We use soft resonant elastic X-ray scattering (REXS), neutron scattering, piezoresponse
 75 force microscopy (PFM) and scanning nitrogen-vacancy (NV) magnetometry to observe periodic
 76 magnetic chiral objects stabilized at multiferroic domain walls showing ferroelectric chirality. These
 77 observations are also backed by spin dynamic simulations.

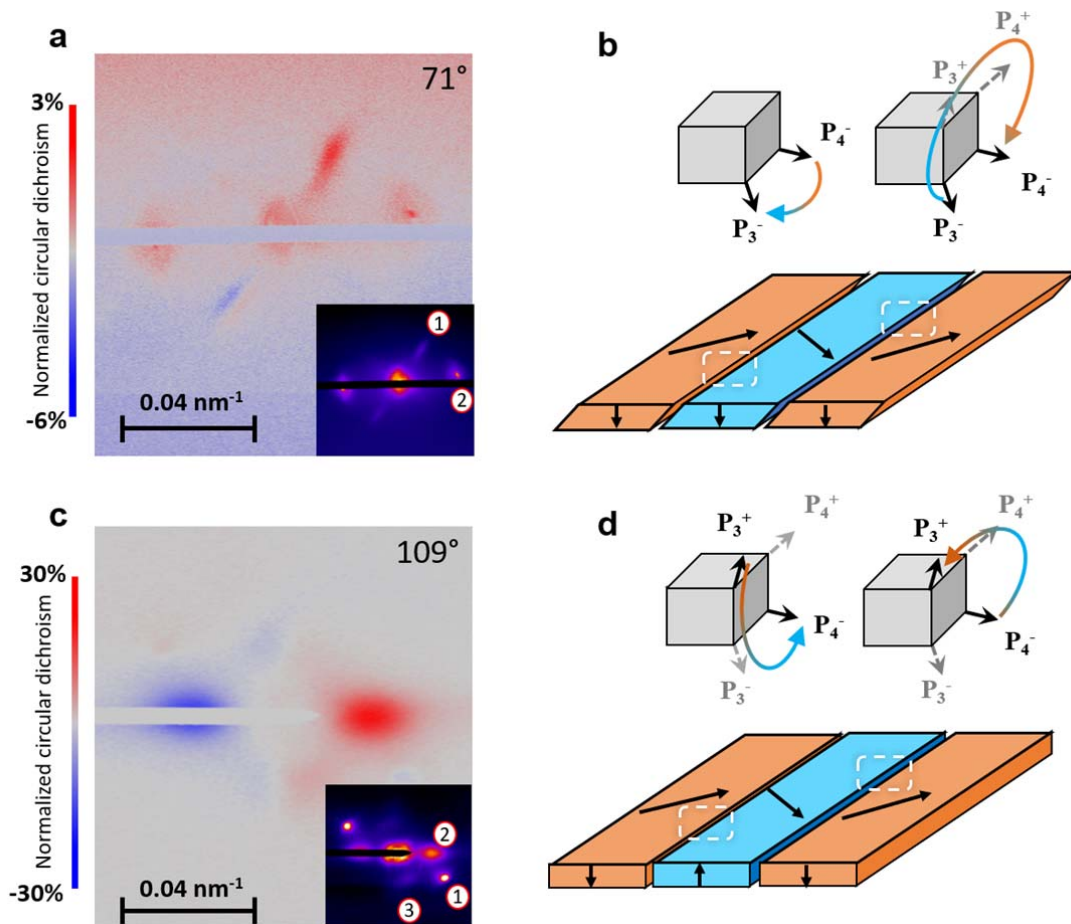


78
 79 **Figure 1: Self-organized ferroelectric patterns and principle of the resonant X-ray diffraction experiments.**
 80 Schematics of **a**, the 71° domain wall in the BiFeO₃ / SrRuO₃ // DyScO₃ epitaxial heterostructure and **b**, the 109°
 81 domain wall in BiFeO₃ / La_{0.7}Sr_{0.3}MnO₃ // DyScO₃ formed by alternating ferroelectric polarization variants. The
 82 dark blue, red, and light green spheres stand for bismuth, oxygen, and iron ions, respectively and the light
 83 yellow planes represent the orientation of ferroelectric domain walls. The grey arrows represent electrical
 84 polarizations. In-plane piezoresponse force microscopy images (4 × 4 μm²) of **c**, the 71° and **d**, the 109°
 85 ferroelectric stripe patterns. Insets: corresponding out-of-plane PFM images. **e**, Sketch of the resonant elastic

86 X-ray scattering experiment in reflectivity geometry with Q_x and Q_y the directions in reciprocal space defined
 87 relative to the incident X-ray beam.

88

89 Two sets of BFO epitaxial layers have been grown by pulsed laser deposition on DyScO₃ substrates
 90 with buffer layers of either SrRuO₃ or La_{0.7}Sr_{0.3}MnO₃ (Methods). Because BFO has a rhombohedral
 91 symmetry (with a slight monoclinic distortion), 8 ferroelectric variants are possible along the {111}
 92 directions in the pseudo-cubic lattice. Mastering growth modes has allowed us to reduce the number
 93 of variants leading in some cases to highly ordered ferroelectric patterns. Our two representative
 94 samples are composed of ferroelectric stripes as characterized by (vectorial) PFM measurements
 95 (Fig. 1c,d). The first one has strong alternating contrast in the in-plane image and flat out-of-plane
 96 contrast (Fig. 1c) demonstrating that the stripes correspond to 71° FE domains (Fig. 1a) with a
 97 periodicity of about 210 nm. The stripes are less ordered in the second sample with dominant 109°
 98 FE domain walls (Fig. 1b) as shown in the two PFM images (Fig. 1d) with a 200 nm characteristic
 99 length scale.



100

101 **Figure 2: Chiral ferroelectric structures at domain walls.** Dichroic patterns demonstrating that chirality is
 102 associated to electrical polarization in **a**, 71° and **c**, 109° domain walls. Insets: corresponding diffraction
 103 patterns around the O K edge (530 eV). In **a**, two pairs of diffraction spots result from the atomic steps of the
 104 substrate (2) and the periodic 71° FE structures (1). In **c**, three pairs of spots are caused by atomic steps (1) and
 105 109° FE structures (2 & 3). Schematics of polarization (black arrows) leading to chiral arrangements for **b**, 71°
 106 and **d**, 109° domain walls.

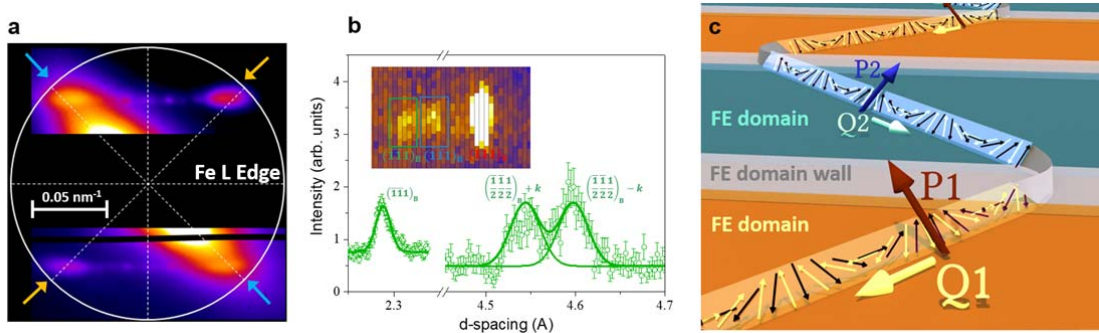
107 REXS measurements were performed at the synchrotron Soleil on the SEXTANTS beamline in
 108 reflectivity geometry (Methods). In the soft energy range, the technique allows accessing complex
 109 charge, magnetic and orbital orders with nanometre spatial resolution. The diffracted intensity of a
 110 spatially modulated order in reciprocal space for a scattering vector Q is given by
 111 $I(Q) \propto |\sum_n f_n \cdot \exp(iQ \cdot r_n)|^2$ where f_n is the scattering amplitude of a single atom at position r_n . This
 112 scattering amplitude is a sum of several contributions, among which some resonant terms are
 113 substantially enhanced when the energy of incoming photons reaches absorption edges. They also
 114 strongly depend on polarization states of incoming and scattered photons. For electron dipole
 115 transitions and in the approximation of a cylindrical symmetry for the considered atoms, the
 116 resonant scattering amplitude f_n^{REXS} can be expanded in a sum of 3 main components¹⁵⁻¹⁷:

$$f_n^{\text{RES}} = f_0(\hat{\epsilon} \cdot \hat{\epsilon}') - i f_1(\hat{\epsilon} \times \hat{\epsilon}') \cdot m_n + f_2(\hat{\epsilon}' \cdot m_n)(\hat{\epsilon} \cdot m_n)$$

117 where $\hat{\epsilon}$ and $\hat{\epsilon}'$ are the polarization states in the Jones formalism of the incident and diffracted
 118 beams, respectively, m_n , the local magnetization unit vector and f_0 , f_1 , f_2 , the photon-energy-
 119 dependent resonance factors for, respectively, the monopole, magnetic dipole and quadrupole parts
 120 of the scattering amplitude. Interestingly, the second term of the right-hand-side, analogous to
 121 circular dichroism in absorption, is sensitive to m whereas the last term, analogous to linear
 122 dichroism in absorption, is sensitive to m^2 and therefore ideal to probe non-collinear
 123 antiferromagnetic orders. In lower symmetry lattices such as in ferroelectrics, parity-odd electric
 124 dipoles do not directly contribute in the electron-dipole approximation. However, the orbital
 125 deformations associated to the electric polarization via spin-orbit coupling can be probed through
 126 the quadrupole part of the scattering amplitude in a more general case¹⁸ than the work of Hannon¹⁵.
 127 The diffraction patterns at the O K edge of Fig. 2a,c indeed provide information about the ordering of
 128 the orbital quadrupole moments reflecting the ferroelectric stripe networks. For the 71° domain
 129 walls, the diffraction peaks ('1' in Fig. 2a) correspond to a ferroelectric stripe pattern with a period of
 130 218 nm \pm 20 nm, as well as more intense 'parasitic' peaks ('2' in Fig. 2a at 49°) generated by the
 131 atomic steps stemming from the miscut of the substrate. This is fully consistent with atomic- and
 132 piezoforce- microscopy measurements (Methods and Supplementary Fig. 1). The case of the 109°
 133 sample is slightly subtler (Fig. 2c) as three populations can be identified. The intense round spots
 134 come from the atomic steps ('1' in Fig. 2c), while the two weaker ones ('2' & '3' in Fig. 2c) come from
 135 the ferroelectric patterns. These correspond to two different period directions: the main (vertical)
 136 stripe structure and some less visible oblique terminations. Interestingly, these diffraction spots
 137 exhibit a clear circular dichroism (Fig. 2a,c), which disappears off-resonance (the full energy
 138 dependence is detailed in Supplementary Fig. 3). This dichroism happens for both systems and
 139 reaches 30 % for the main family of spots of the 109° DWs and 2% for the 71° DWs. The smaller
 140 dichroism of the latter is mainly due to the measuring angle, imposed by the blinding from the
 141 diffracted spots of the regular atomic steps at other angles.

142 Although X-rays cannot directly access electrical polarization in the electron-dipole approximation,
 143 they can however indirectly be sensitive to chiral polar arrangements^{19,18}. The dichroism at the O K
 144 edge therefore indicates the presence of chirality in the ferroelectric order. This is only possible if the
 145 polarization does not vanish at the domain walls, which are non-Ising like, in agreement with recent
 146 reports²⁰. This also imposes a homochiral FE winding (Fig. 2b,d), inconsistent with a zigzag winding of
 147 DWs linking in the simplest way, the polarizations on each side. It is easy to imagine how this is
 148 possible for 109° DWs as going from say the P_3^+ polarization direction of the first domain to the P_4^- of
 149 the second rotating via P_3^- or via P_4^+ are energetically degenerate (Fig. 2d). Thus, although we do not
 150 know what forces such a state, our measurements indicate that the two windings alternate: via P_3^- in
 151 one set of DWs and via P_4^+ for the neighbours, thus imposing a net chirality. Note that a slight
 152 dichroism is also visible at the Fe L edge (not shown), which finds a similar explanation as recently
 153 developed by Lovesey and van der Laan¹⁸ and therefore corroborate the polar chirality in our
 154 systems. The case of the 71° DWs is more puzzling as a homochiral state imposes every second wall
 155 to wind in a 'long angle'. For instance, when P directly rotates from P_4^- to P_3^- in one DW, it then goes

156 from P_3^- to P_3^+ then P_4^+ to reach P_4^- in the neighbouring one (Fig. 2b). This is obviously higher in
 157 energy than the direct P_3^- to P_4^- route and singles out the nature of this set of walls. Note here that
 158 P_3^+ and P_4^+ regions are not visible in the PFM images implying that they are narrow, but their
 159 presence makes the whole FE structure chiral.

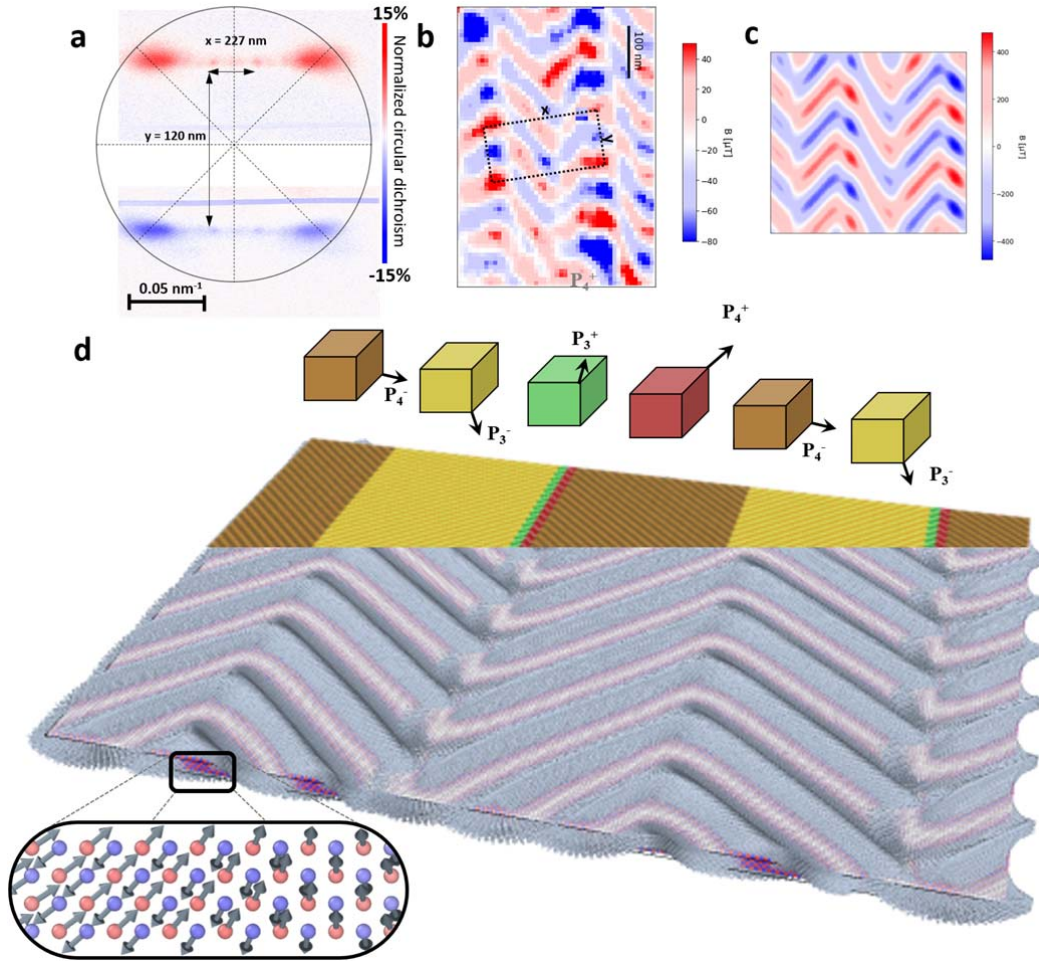


160

161 **Figure 3: Non-collinear magnetic structure in ferroelectric domains by neutron and resonant X-ray scattering.**
 162 **a**, X-ray diffraction at the Fe L edge (707.5 eV) for the 71° periodic ferroelectric patterns. The four main spots
 163 (pointed by blue and yellow arrows) visible in **a** stem from the two families of cycloids sketched in **c** and
 164 correspond to a cycloidal period of about 80 nm. **b**, Neutron scattering measurements (integrated intensity and
 165 2D in inset) show the splitting of the Bragg peaks confirming the cycloidal order. Open circles are experimental
 166 data and solid lines are fits assuming a bulk-like spin cycloid with a period of 80 nm. **c**, Sketch of the two
 167 cycloids (one in each polarization domain) propagating in BiFeO₃. P_i and Q_i stand for the polarization vector of
 168 the domains and the wavevector of the cycloids, respectively.

169

170 Turning now to the magnetic order, the (circularly polarized) X-ray wavelength is tuned to the Fe L
 171 edge (Methods and Supplementary Fig. 2). The measured REXS diffraction patterns are now
 172 completely different, as four peaks appear in a square pattern as shown in Fig. 3a for the 71° FE
 173 stripe sample. These should be viewed as paired symmetrically with respect to the specular beam
 174 (thus in diagonal). They stem from two cycloidal families, one in each ferroelectric domain, with their
 175 propagation vector in the film plane and perpendicular to the local polarizations. This is also
 176 confirmed by time of flight neutron diffraction measurements (Fig. 3b, see Methods and
 177 Supplementary Fig. 5). Thus, only one out of the three symmetry allowed cycloidal families is present
 178 in each polarization domain (Fig. 3c), as the monoclinic distortion lowers further the initial 3-fold
 179 symmetry. The period is found to be 80 ± 8 nm, a little greater than the bulk period (64 nm). A closer
 180 examination of the elongated shape of the diffraction peaks indicates that they are actually
 181 composed of two spots, which stem from a splitting of the cycloidal AF peaks due to the modulation
 182 caused by the FE stripe pattern. Polarized X-rays and neutrons have the further capability to locate
 183 the spins direction, as previously demonstrated in BFO^{21,22,23}. To our knowledge, the extra ability of
 184 both techniques to study the chiral nature of the AF arrangements has not been applied to BFO thin
 185 films. With their ultra-high intensity, synchrotron X-rays are the ideal probe of chirality in thin films.
 186 This was previously demonstrated in ferromagnets by analysing circular dichroism, i.e. the intensity
 187 difference between right and left polarized X-rays²⁴. The magnetic scattering term used to measure
 188 ferromagnetic chirality is $-i f_1(\hat{\epsilon} \times \hat{\epsilon}') \cdot m_n$. Here, we extend this type of analysis to antiferromagnetic
 189 chiralities through the $f_2(\hat{\epsilon}' \cdot m_n)(\hat{\epsilon} \cdot m_n)$ term, giving rise to diffraction peaks located at $2q_{\text{cycloid}}$
 190 in reciprocal space. Similarly, the dichroic plot of Fig. 4a for the 71° sample shows that +q and -q spots
 191 have opposite dichroism, indicating that the antiferromagnetic cycloids are chiral. This change of sign
 192 also reverses after an azimuthal sample rotation of 180° (Methods and Supplementary Fig. 4), which
 193 is consistent with a winding uniquely linked to the direction of P in each ferroelectric domain as
 194 expected theoretically²⁵ and measured in the bulk²³. We point out here that this feature cannot be
 195 due to the (non-chiral) periodic magnetic uncompensation, which would appear at q_{cycloid}
 196 in reciprocal space.



197

198 **Figure 4: Chiral magnetic textures at ferroelectric domain walls seen in reciprocal and real spaces.** **a**, Intensity
 199 difference between left and right X-ray polarizations for the two-cycloid system showing opposite dichroism for
 200 each set of diffraction spots taken as a signature of chirality. **b**, Scanning NV magnetometry image of the stray field 60 nm above the sample confirming in real space the two cycloidal populations and the rectangular array of bubbles at ferroelectric domain walls. **c-d**, Magnetic simulations using an atomistic code and showing the cycloidal arrangements in (a four-atom-thick slab of) the FE stripe domains, as well as their stitching at the walls (**d**). Every second wall, the polarization (black arrows) rotates along a long winding consistent with the chirality measured for the FE order. The stray field extracted from the simulations (**c**) shows excellent agreement with NV magnetometry in **b**.

207 In addition, the diffraction pattern of Fig. 4a comprises some extra features in the form of spots
 208 spreading along horizontal lines. These are representative of periodic structures generated by the FE
 209 stripes. Indeed, the two pairs of bright spots have a horizontal separation corresponding to the FE
 210 stripes' period of about 220 nm and a vertical separation giving 120 nm in real-space, close to the
 211 length of the cycloids projected onto the DWs ($\sqrt{2} \times 80$ nm). Therefore, this pattern results from the
 212 merging of the cycloids at the periodic domain walls. Importantly, its dichroic character indicates that
 213 the structures are chiral. Moreover, the weaker middle spots have an intensity inconsistent with the
 214 individual diffraction of two periodically striped cycloidal families. We thus attribute their presence
 215 to the diffraction of a chiral rectangular pattern stemming from the stitching of the cycloids.

216 To corroborate the presence of these chiral objects, we have combined REXS with real-space imaging
 217 by means of scanning NV magnetometry (Methods)^{26,27}. Fig. 4b shows a magnetic image in a sample
 218 with 71° DWs grown under the exact same conditions. In addition to a vertical periodicity
 219 corresponding to the cycloids in different domains, regular bright spots are visible along one domain

220 wall out of two. They are therefore related to the different cycloidal windings. It is worth noting that
221 in the simplest case of the short angle rotation of the ferroelectric polarization, the AF cycloid
222 stitching can be straightforwardly almost continuous. In that case, the discontinuity in magnetization
223 is minimum and the stray field should be close to that in the domain. On the other hand, for a wall
224 with the FE polarization rotating along the long angle (as sketched in Fig. 2b), a frustration appears
225 and the stitching is more complex. Interestingly, the measured hot spots on the scanning NV
226 magnetometry image can be considered as the embryos of multi-q topological objects, especially as
227 the dichroic diffraction patterns evidence their chirality. In order to better illustrate this DW effect,
228 we have carried out numerical simulations using a dynamical atomistic magnetic simulations code²⁸.
229 We minimized the convergence time by dividing all distances by 10 and correspondingly multiplying
230 by 10 the magnetoelectric interactions. This scaling does not affect the qualitative topography of the
231 calculated magnetic configuration (Methods and Supplementary Fig. 6). The underlying striped FE
232 state is taken with a 1-nm slab of a reversed polarization inserted every second wall to account for
233 the ‘reverse winding’. Fig. 4d shows the obtained spin arrangement where AF cycloids are clearly
234 visible in the stripes and a peculiar stitching is observed on the walls where the FE polarization
235 rotates along the wide angle. The stray field generated by the uncompensated moments shows
236 excellent agreement with the recorded scanning NV magnetometry image (Fig. 4c) and notably the
237 hot spots measured every second wall. This provides further evidence supporting our conclusions on
238 both electric and magnetic chiralities. Because of the magnetoelectric interaction, these are
239 entangled at domain walls to create very special entities. Our results call for a proper hybrid
240 modelling of these peculiar objects and of their interplay with charge currents, spin currents and
241 light, in analogy with the topological Hall effect²⁹ and related emergent electromagnetic
242 phenomena³⁰ predicted for skyrmion systems.

243

244 **Acknowledgments**

245 We thank Horia Popescu for his assistance during the synchrotron runs, Antoine Barbier for
246 discussions regarding diffraction as well as Yves Joly and Gerrit van der Laan for fruitful discussions
247 about theoretical aspects of resonant x-ray scattering. We also acknowledge the company QNAMI for
248 providing all-diamond scanning tips containing single NV defects. V.J. acknowledges financial support
249 by the European Research Council (ERC-StG-2014, Imagine). The authors also acknowledge support
250 from the French Agence Nationale de la Recherche (ANR) through projects Multidolls, PIAF and
251 SANTA as well as the ‘programme transversal CEA ACOSPIN and ELSA’. This work was also supported
252 by a public grant overseen by the ANR as part of the ‘Investissement d’Avenir’ programme (LABEX
253 NanoSaclay, ref. ANR-10-LABX-0035).

254

255

256 **Authors contributions:**

257 JYC, MV and NJ planned the REXS experiment and carried it out with CB. VG and SF prepared the
258 samples and carried out the PFM measurements. BD, DK and PM carried out the neutron
259 measurements while WA, IG, IF and VJ carried out the NV magnetometry. TC, JT and PT wrote,
260 optimized and ran the simulation code. All authors participated in scientific discussions.

261

262 **Data Availability:** All relevant data are available from the authors, and/or are included with the
263 manuscript.

264

265

266 **References**

- 267 1. Emori, S., Bauer, U., Ahn, S.-M., Martinez, E. & Beach, G. S. D. Current-driven
268 dynamics of chiral ferromagnetic domain walls. *Nat. Mater.* **12**, 611–616 (2013).
- 269 2. Fert, A., Reyren, N. & Cros, V. Magnetic skyrmions: advances in physics and potential
270 applications. *Nat. Rev. Mater.* **2**, 17031 (2017).
- 271 3. Barker, J. & Tretiakov, O. A. Static and Dynamical Properties of Antiferromagnetic
272 Skyrmions in the Presence of Applied Current and Temperature. *Phys. Rev. Lett.* **116**,
273 147203 (2016).
- 274 4. Nahas, Y. *et al.* Discovery of stable skyrmionic state in ferroelectric nanocomposites. *Nat.*
275 *Commun.* **6**, 8542 (2015).
- 276 5. Pereira Gonçalves, M. A., Escorihuela-Sayalero, C., Garca-Fernández, P., Junquera, J. &
277 Íñiguez, J. Theoretical guidelines to create and tune electric skyrmion bubbles. *Sci. Adv.*
278 **5**, eaau7023 (2019).
- 279 6. Jungwirth, T., Marti, X., Wadley, P. & Wunderlich, J. Antiferromagnetic spintronics. *Nat.*
280 *Nanotechnol.* **11**, 231–241 (2016).
- 281 7. Wadley, P. *et al.* Electrical switching of an antiferromagnet. *Science* **351**, 587–590
282 (2016).
- 283 8. Smolenskii, G. A. & Chupis, I. E. Ferroelectromagnets. *20* (1982).
- 284 9. Dzyaloshinsky, I. A thermodynamic theory of “weak” ferromagnetism of
285 antiferromagnetics. *J. Phys. Chem. Solids* **4**, 241–255 (1958).
- 286 10. Seidel, J. *et al.* Conduction at domain walls in oxide multiferroics. *Nat. Mater.* **8**, 229–234
287 (2009).
- 288 11. Van Aert, S. *et al.* Direct Observation of Ferrielectricity at Ferroelastic Domain
289 Boundaries in CaTiO₃ by Electron Microscopy. *Adv. Mater.* **24**, 523–527 (2012).
- 290 12. Wei, X.-K. *et al.* Ferroelectric translational antiphase boundaries in nonpolar materials.
291 *Nat. Commun.* **5**, (2014).

- 292 13. Bhattacharya, K. APPLIED PHYSICS: The Material Is the Machine. *Science* **307**, 53–54
293 (2005).
- 294 14. Catalan, G. & Scott, J. F. Physics and Applications of Bismuth Ferrite. *Adv. Mater.* **21**,
295 2463–2485 (2009).
- 296 15. Hannon, J. P., Trammell, G. T., Blume, M. & Gibbs, D. X-Ray Resonance Exchange
297 Scattering. *Phys. Rev. Lett.* **61**, 1245–1248 (1988).
- 298 16. Hill, J. P. & McMorrow, D. F. X-ray Resonant Exchange Scattering: Polarizaion
299 Dependence and Correlation Function. *Acta Crystallogr. A* **52**, 236–244 (1996).
- 300 17. van der Laan, G. Soft X-ray resonant magnetic scattering of magnetic nanostructures.
301 *Comptes Rendus Phys.* **9**, 570–584 (2008).
- 302 18. Lovesey, S. W. & van der Laan, G. Resonant x-ray diffraction from chiral electric-
303 polarization structures. *Phys. Rev. B* **98**, 155410 (2018).
- 304 19. Shafer, P. *et al.* Emergent chirality in the electric polarization texture of titanate
305 superlattices. *Proc. Natl. Acad. Sci.* **115**, 915–920 (2018).
- 306 20. Cherifi-Hertel, S. *et al.* Non-Ising and chiral ferroelectric domain walls revealed by
307 nonlinear optical microscopy. *Nat. Commun.* **8**, 15768 (2017).
- 308 21. Waterfield Price, N. *et al.* Coherent Magnetoelastic Domains in Multiferroic BiFeO₃
309 Films. *Phys. Rev. Lett.* **117**, (2016).
- 310 22. Lebeugle, D. *et al.* *Phys Rev Lett* **100**, 227602 (2008).
- 311 23. Johnson, R. D. *et al.* X-Ray Imaging and Multiferroic Coupling of Cycloidal Magnetic
312 Domains in Ferroelectric Monodomain BiFeO₃. *Phys. Rev. Lett.* **110**, (2013).
- 313 24. Dürr, H. A. *et al.* Chiral magnetic domain structures in ultrathin FePd films. *Science* **284**,
314 2166–2168 (1999).
- 315 25. Mostovoy, M. Ferroelectricity in Spiral Magnets. *Phys. Rev. Lett.* **96**, (2006).

- 316 26. Rondin, L. *et al.* Magnetometry with nitrogen-vacancy defects in diamond. *Rep. Prog.*
317 *Phys.* **77**, 056503 (2014).
- 318 27. Gross, I. *et al.* Real-space imaging of non-collinear antiferromagnetic order with a single-
319 spin magnetometer. *Nature* **549**, 252–256 (2017).
- 320 28. Tranchida, J., Plimpton, S. J., Thibaudeau, P. & Thompson, A. P. Massively parallel
321 symplectic algorithm for coupled magnetic spin dynamics and molecular dynamics. *J.*
322 *Comput. Phys.* **372**, 406–425 (2018).
- 323 29. Bruno, P., Dugaev, V. K. & Taillefer, M. Topological Hall Effect and Berry Phase in
324 Magnetic Nanostructures. *Phys. Rev. Lett.* **93**, (2004).
- 325 30. Nagaosa, N. & Tokura, Y. Emergent electromagnetism in solids. *Phys. Scr.* **T146**, 014020
326 (2012).
- 327
- 328

329 **Methods**

330 **Sample preparation and characterization**

331 The epitaxial thin film heterostructures were grown by pulsed laser deposition using a KrF excimer
332 laser ($\lambda = 248$ nm, 1 J cm^{-2}) on an orthorhombic DyScO_3 (110) single crystal substrate. The SrRuO_3
333 (resp. $\text{La}_{1/3}\text{S}_{2/3}\text{rMnO}_3$) bottom electrode (1.2 nm) was grown with 5 Hz repetition rate at 650 °C under
334 0.2 mbar of oxygen for the sample exhibiting 71° (resp. 109°) stripe domains. The BiFeO_3 films (32
335 nm) were subsequently grown at 650 °C under 0.36 mbar of oxygen at 1 Hz repetition rate. The
336 heterostructures were slowly cooled down under high oxygen pressure. In both cases, the film
337 surface exhibits single-unit-cell atomic steps (with additional ferroelastic deformations related to the
338 ferroelectric variants of the 109° sample).

339 The structural thin film properties were assessed by X-ray diffraction (XRD, Xpert Panalytical)
340 performing reciprocal space mappings (RSMs) along different directions of the monoclinic DyScO_3
341 substrate. PFM experiments were conducted with an atomic force microscope (Nanoscope V
342 multimode, Bruker) and two external SR830 lock-in detectors (Stanford Research) for simultaneous
343 acquisition of in-plane and out-of-plane responses. A DS360 external source (Stanford Research) was
344 used to apply the AC excitation to the bottom electrodes at a frequency of 35 kHz while the
345 conducting Pt coated tip was grounded. The ferroelectric configurations of the pristine BFO samples
346 were identified by vectorial PFM, i.e. probing the different in-plane variants when rotating the
347 sample crystallographic axes compared to the PFM cantilever long axis. Alternated light/dark stripes
348 are observed in the in-plane PFM phase image acquired with the cantilever aligned along the
349 pseudo-cubic $[100]_c$ direction.

350 The PFM and AFM images show structures that can be compared to the resonant scattering
351 diffraction through FFT transforms. Two different features can be attributed to diffraction by
352 substrate atomic steps and electrical polarization stripes, but peak No3 of the REXS does not appear
353 in the PFM+AFM images. We attribute it to some domains' terminations, which can be guessed from
354 the real-space PFM image but not ordered enough to appear in the Fourier transforms.

355 **Resonant soft x-ray elastic scattering**

356 A. Experimental details and setup configuration

357 The experiment has been performed at SOLEIL synchrotron on the SEXTANTS beamline in the RESOXS
358 [31] diffractometer. The diffractometer is equipped with a CCD camera (2048x2048 pixels of 13.5 μm)
359 that is on the detector arm at 26 cm from the sample. The solid angle probed by the camera alone is
360 6.3° but it can be further moved perpendicularly to the scattering plane by +/- 15mm to increase the
361 accessible reciprocal space. A set of beamstops placed a few mm before the CCD chip can be
362 precisely aligned with few 10nm precision in order to block the specular beam or unwanted strong
363 Bragg reflections.

364 Data were collected at both O K edge and Fe L edges using circularly polarized x-rays delivered from
365 an Apple 2 undulator located at the I14M straight section of SOLEIL storage ring. A typical photon flux
366 of 10^{13} ph/s with 100meV energy resolution was used for the experiment.

367

368 B. Energy dependence

369 Prior to any conclusions relating to circular dichroism and chirality, the energy dependence of circular
370 dichroic diffraction patterns has been measured both around the Fe L_3 edge and the O K edge.
371 Supplementary Fig. 2 presents the normalized circular dichroism for the upper diffraction pattern as

372 function of the incident photon energy around the Fe L₃ edge. The dashed line stands for the
373 normalized specular intensity at an incident x-ray angle of 30°. On the left side of the figure, a
374 selection of normalized circular dichroisms of the cycloidal diffraction patterns are presented. As
375 expected, there is no signal off-resonance and two maxima are observed for the 2p to 3d t_{2g}
376 transition (707.5 eV) and about 1 eV above the 2p to 3d e_g transition (710 eV). However, when
377 reaching the maximum of the main resonance (709 eV), the circular dichroism is either vanishing or
378 even reversing. Yet, at this energy, the intensity is one order of magnitude larger and additional
379 effects might play a non-negligible role. Indeed, dynamical effects and phenomena such as x-ray
380 birefringence [32] or magneto-optical effects [33] are known to occur. One crucial consequence is
381 that the X-ray polarization could be modified during the crossing of the material (and not only by
382 scattering processes). Because it is challenging to account for these processes, we have avoided this
383 energy for the analysis of the magnetic chirality. A similar argument holds for the O K edge, as can be
384 seen in Supplementary Fig. 3. A strong energy dependence is evidenced, yet no inversion of circular
385 dichroism is observed in the peaks under consideration. It is interesting to notice that the maxima in
386 circular dichroism are found in the feet of the main resonances.

387

388 C. Azimuthal dependence

389 Topological structures generate a dichroic signal that is of opposite sign in +q and -q. This depends
390 on the sign of the chirality and it is therefore linked to the antiferromagnetic winding in the sample.
391 This provides us with another means to further confirm that the dichroism stems from the AF
392 cycloids. Indeed, as Supplementary Fig. 4 shows, the dichroism reverses when the sample is rotated
393 by 180° in the azimuthal plane.

394 Neutron scattering

395 We used time of flight neutron diffraction to confirm the presence of the cycloidal ordering in our
396 BFO/DSO thin film. The measurements were carried out in four different scattering geometries
397 schematically shown in Supplementary Fig. 5. For each sample orientation (angle ϕ in Supplementary
398 Fig. 5a), two distinct pseudo-cubic diagonals {111}_D-type, referring to the DSO substrate unit cell,
399 were in the scattering plane, but as only one of them could be optimized with respect to the neutron
400 flux and resolution for the target magnetic reflections, two measurements (angle ω in
401 Supplementary Fig. 5a) were performed. The target reflections were the satellites of the $\{\frac{1}{2}\frac{1}{2}\frac{1}{2}\}\pm k$
402 type expected at a *d*-spacing range around 4.5-4.6 Å. The measurements were performed at high
403 scattering angles (typically above 120°) to achieve a good momentum transfer resolution and the
404 reflecting geometry helped to avoid absorption from the substrate. After 16 hours of exposition time,
405 the (hhh)-type reflections of the BFO film could be clearly observed (Supplementary Fig. 5b,c). In two
406 out of the four scattering geometries, two signals, spatially separated from the substrate on the
407 detectors array, could be resolved (Supplementary Fig. 5b), revealing the presence of structural
408 domains. In the other two measurements, the domains could not be resolved (Supplementary Fig.
409 5c). Data, focused around these (hhh)-type reflections and plotted as a function of *d*-spacing, are
410 presented in Supplementary Fig. 5d-f. The patterns clearly reveal the presence of magnetic
411 reflections in the expected *d*-spacing range. The magnetic satellites scatter very close to the nuclear
412 (hhh)-type reflections and the angular resolution of the data is not sufficient to spatially isolate the
413 nuclear and magnetic reflections on the detector array. The *d*-spacing positions and intensities of the
414 observed magnetic reflections can be quantitatively fitted by the model implying a cycloidal magnetic
415 order, similar to that in bulk BFO, and the presence of two equi-populated ferroelectric domains
416 illustrated in Supplementary Fig. 5g,h. It is remarkable that the data can be satisfactorily modelled
417 only assuming a single magnetic domain state with the propagation vector $k=(-\delta,0,\delta)$, $\delta\approx 0.043(3)$ for
418 both ferroelectric domains. As possible reasons for suppression of the other two magnetic domains

419 with $k_2=(0,-\delta,\delta)$ and $k_3=(-\delta,\delta,0)$ is the small monoclinic distortion of the ferroelectric domains
420 imposed by the misfit with the substrate as discussed below.

421 The pseudocubic unit cell metric of the orthorhombic DSO substrate (Supplementary Fig. 5i) is
422 characterised by two distances ($a_D=c_D \neq b_D$) and one angle ($\beta_D \neq 90^\circ$). This implies two non-equivalent
423 $\{100\}$ -type BFO films with the b_D -axis either parallel or perpendicular to the surface. In our case, the
424 b_D -axis is parallel to the surface. The unit cell is also characterised by two long $[11-1]_D/[1-1-1]_D$ and
425 two short $[111]_D/[1-11]_D$ diagonals as shown in Supplementary Fig. 5i. Our neutron diffraction data
426 indicate that this type of substrate imposes a selection of two ferroelectric domains in the BFO film.
427 The polar $[111]$ axes of these domains are along the long diagonals $[11-1]_D$ and $[1-1-1]_D$ of the
428 substrate (Supplementary Fig. 5g-i). Moreover, the pseudocubic unit cell metric of the BFO film
429 ($a_f=b_f=c_f$, $\alpha_f=\beta_f=\gamma_f \neq 90^\circ$) does not match the metric of the substrate, resulting in additional structural
430 distortions. These could be responsible for the selection of the single magnetic domain state,
431 through magneto-elastic coupling, within the structural ferroelectric domains. The resolution of our
432 neutron diffraction data, however, is not sufficient to directly measure these distortions.

433 In the scattering geometries optimized to measure magnetic satellites, close to the $(11-1)_D$ and $(1-1-1)_D$
434 reflections, the two domains cannot be separated and a contribution from both domains is
435 present in the focused data (Supplementary Fig. 5c,f). The refined value of the propagation vector
436 $\delta=0.043(3)$ is slightly smaller than that of the bulk BFO indicating a slightly longer period of the
437 cycloidal modulation with respect to that of the bulk. This is in a good agreement with the
438 synchrotron data.

439 **Scanning NV magnetometry**

440 Scanning-NV magnetometry is performed with a commercial all-diamond scanning-probe tip
441 containing a single NV defect (QNAMI, Quantilever MX). This tip is integrated into a tuning-fork-
442 based atomic force microscope (AFM) combined with a confocal microscope optimized for single
443 defect spectroscopy. Details about the experimental setup can be found in [34]. Quantitative
444 magnetic field distributions are obtained by monitoring the Zeeman shift of the NV defect electron
445 spin sublevels through optical detection of the magnetic resonance [35]. Experiments are performed
446 under ambient conditions with a NV-to-sample distance of 60 nm.

447 **Atomistic spin dynamic computation**

448 Unlike micromagnetic simulations applied to ferromagnets, large-scale simulations of complex
449 antiferromagnetic textures remain a challenge. Indeed, the micromagnetic approximation consisting
450 in averaging local magnetic moments into large enough computational cells and supposing that this
451 quantity only changes in direction and not in magnitude, fails for antiferromagnets. In order to
452 provide a simulation support for our experimental findings, atomistic spin dynamics simulations have
453 been performed where each atom carries a spin. The relevant interactions for BFO have been
454 considered, namely exchange, magnetoelectric, anisotropy and asymmetric Dzyaloshinskii-Moriya
455 interactions on a periodically striped ferroelectric pattern. In order to model as closely as possible
456 the antiferromagnetic configurations of our samples, the ferroelectric domain walls have been
457 designed as measured, with a chiral winding. For the 71° pattern, one wall out of two was taken to
458 rotate following the ‘long winding’ schematically represented in Fig. 4b of the main text. The
459 simulations have been carried out using the SPIN package of LAMMPS [36, 37] which assesses spin
460 textures and their dynamics by solving the Landau-Lifshitz-Gilbert (LLG) equation :

$$\frac{ds_i}{dt} = \omega_i \times s_i + \alpha s_i \times \frac{ds_i}{dt}$$

461 with s_i the unit spin vector at the i^{th} position, α the Gilbert damping coefficient and ω_i the effective
462 spin pulsation defined as:

$$\omega_i = \frac{-1}{\hbar} \frac{\partial H}{\partial s_i}$$

463 For the present study, the three main components of the Hamiltonian (H) of N interacting spins are
464 considered, namely the symmetric exchange, the magneto-electric (ME) and the asymmetric
465 Dzyaloshinskii-Moriya interactions (DM) expressed as follows:

$$H = - \sum_{i,j,i \neq j}^N J_{ij} s_i \cdot s_j + (P \times e_{ij}) \cdot (s_i \times s_j) + (p \times e_{ij}) \cdot (s_i \times s_j)$$

466 with J_{ij} the symmetric exchange coefficient, e_{ij} the unit direction vector between the i^{th} and j^{th}
467 spins, P the global ferroelectric polarization and p the local polarization induced by the octahedral
468 tilts and alternating from site to site. Noteworthy, the consequence of this latter is the small cycloidal
469 wriggling leading to an apparent spin density wave.

470 The simulations have been performed for $N=N_x(240)*N_y(172)*N_z(4)$ spins with periodic boundary
471 conditions in x and z . A cubic lattice with a 3.96 \AA constant was considered. The x -direction
472 corresponds to the (100) crystallographic direction. Note that while the antiferromagnetic exchange
473 interaction has been kept to its known value (-4.5 meV for nearest neighbour interaction), ME and
474 DM interactions were multiplied by a factor 10 (1.09 meV and 0.54 meV respectively) to reduce the
475 simulated volume and hence the calculation time. The cycloidal period scales correspondingly.

476 The spins are divided into 60 atom wide stripes with different orientations of P in order to mimic the
477 71° ferroelectric striped pattern. Every second ferroelectric domain wall is defined following the
478 assumptions explained in the article. The result is shown in Supplementary Fig. 6.

479 Subsequently, the magnetic stray field of the converged spin texture is calculated and projected
480 along the NV defect quantization axis [34], leading to the result shown in figure 4 d of the main
481 article.

482

483 References

484

- 485 [31] N. Jaouen, J.-M. Tonnerre, G. Kapoujian, P. Tautier, J.-P. Roux, D. Raoux, and F. Sirotti, J.
486 Synchrotron Radiat. **11**, 353 (2004).
- 487 [32] Y. Joly, S. P. Collins, S. Grenier, H. C. N. Tolentino, and M. De Santis, Phys. Rev. B **86**, 220101
488 (2012).
- 489 [33] M. W. Haverkort, N. Hollmann, I. P. Krug, and A. Tanaka, Phys. Rev. B **82**, 094403 (2010).
- 490 [34] L. Rondin et al., Appl. Phys. Lett. **100**, 153118 (2012).
- 491 [35] L. Rondin, et al., Rep. Prog. Phys. **77**, 056503 (2014).
- 492 [36] S. Plimpton, J. Comput. Phys. **117**, 1 (1995).
- 493 [37] J. Tranchida, S. J. Plimpton, P. Thibaudeau, and A. P. Thompson, J. Comput. Phys. **372**, 406
494 (2018).

