

Non-Volatile Electric-Field Toggling Between Antiferromagnetic States

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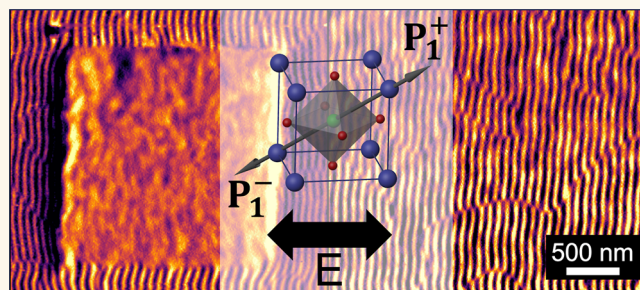
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ABSTRACT: Magnetoelectric multiferroics are foreseen as paramagnetic materials to control magnetism with an electric field, targeting energy-efficient spintronics. The archetypal room-temperature antiferromagnetic ferroelectric, BiFeO₃, harbors an incommensurate antiferromagnetic cycloid whose propagation direction is locked to ferroelectric domains. Epitaxial strain was shown to affect this antiferromagnetic ordering, stabilizing different cycloidal propagation directions, or a collinear antiferromagnetic state. Here we demonstrate the reversible, nonvolatile, electric-field triggered magnetic phase transition between two distinct antiferromagnetic states at room temperature. Using SrTiO₃ vicinal substrates, we stabilize a single ferroelectric domain associated with a single antiferromagnetic cycloidal state in BiFeO₃ epitaxial thin films. Electrically reversing the ferroelectric polarization deterministically within the same ferroelastic domain induces a reversible transition from a cycloidal to a collinear antiferromagnetic state, as directly visualized by scanning nitrogen vacancy (NV) magnetometry. These results bring insights into magnetoelectric devices for ultrafast and low-power spintronics.

KEYWORDS: bismuth ferrite, electric-field control, magnetoelectric, magnetic phase transition, antiferromagnetic, deterministic



TEXT. Future developments of logic circuits will require beyond CMOS alternatives based on new physical mechanisms.¹ Among various emerging device technologies, spintronics appears as a promising field of research with continuous progress made over the past few years, in terms of endurance, writing speed, and CMOS back-end of line integration.² Nevertheless, writing magnetic information using spin-transfer torque or spin-orbit torque still requires large amounts of currents. An alternative consists in using magnetoelectric multiferroics to write magnetic information using electric fields, as recently proposed by INTEL in their magnetoelectric spin-orbit (MESO) device.³ In such a device, the magnetoelectric multiferroic is adjacent to a ferromagnetic memory element. Writing is achieved by a voltage pulse across the multiferroic, exchange coupled to the ferromagnet, with writing energies expected to reach the aJ/bit, i.e., orders of magnitude lower than any existing technology.⁴ Reading is achieved by spin-to-charge conversion in a metal with strong spin-orbit coupling on the other side of the ferromagnet. The experimental implementations of this MESO concept have

been achieved so far on heterostructures derived from BiFeO₃ (refs 5 and 6).

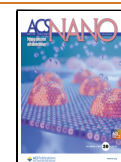
BiFeO₃ is the archetypal multiferroic combining ferroelectricity with robust polarization ($P_r \sim 100 \mu\text{C}/\text{cm}^2$, $T_c \sim 1100 \text{ K}$)^{7,8} and G-type antiferromagnetism forming a long-range spin cycloid (periodicity $\sim 62 \text{ nm}$, $T_N \sim 640 \text{ K}$).^{9,10} Furthermore, antiferrodistortive oxygen octahedral tilts induce an additional space varying canted moment in the form of a spin-density wave locked to the spin cycloid period,¹¹ enabling real-space imaging of the periodic magnetic stray field from the spin cycloid using scanning NV magnetometry.^{12,13} Due to the crystal symmetry of BiFeO₃, eight ferroelectric polarizations along $\langle 111 \rangle_{\text{pc}}$ and three cycloidal propagation vectors along $\langle 1\bar{1}0 \rangle_{\text{pc}}$ are energetically degenerate in bulk (pc refers to the

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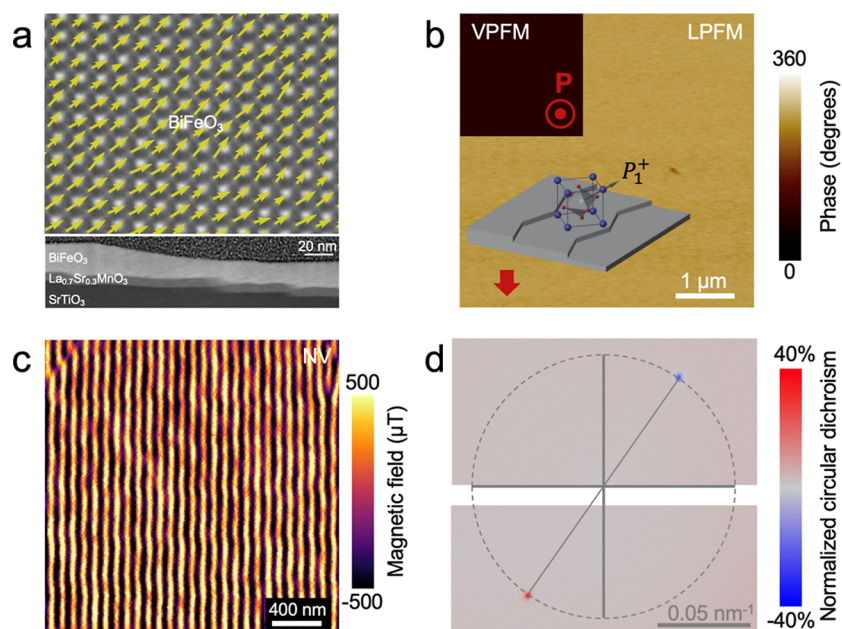


Figure 1. Stabilization of a single-domain multiferroic state. (a) Cross section STEM image of BiFeO₃ grown on a SrTiO₃(001) substrate with a 4° miscut along the [110] direction (zone axis along [110]). These mechanical boundary conditions favor a single ferroelastic domain. The buffer oxide electrode of La_{0.7}Sr_{0.3}MnO₃ gives rise to a single polarization state in BiFeO₃ pointing upward, as visualized by cation displacement maps (zone axis along [100]). (b) PFM images confirm the single ferroelectric domain nature of the BiFeO₃ film with a uniform signal in both lateral (LPFM) and vertical (VPFM, inset) components. The sketch in inset illustrates the single domain nature of the BiFeO₃ film. (c) The corresponding magnetic stray field image performed by scanning NV magnetometry shows a single antiferromagnetic cycloid with a propagation vector parallel to the step edges of the SrTiO₃ substrate. (d) Resonant elastic X-ray scattering circular dichroism at the Fe L edge shows a pair of narrow peaks around the specular reflection with a strong dichroism. This attests for the presence of a single spin cycloid.

pseudocubic notation). This yields a complex antiferromagnetic landscape exhibiting three propagation vectors (k_1 , k_2 , k_3) in bulk single crystals¹⁴ or in thin films^{13,15} for each ferroelectric variant. Thus, stabilizing single-domain BiFeO₃ thin films is essential to eliminate the impact of domain walls, including their associated conductivity¹⁶ and crystal symmetry complexities^{17,18} leading to multiple switching paths. Removing potential conduction paths at ferroelectric domain walls also facilitates low-voltage switching, while the absence of the multiple ferroelectric and antiferromagnetic variants may enable deterministic switching behavior. Anisotropic strain is a powerful knob to manipulate and simplify the multiferroic landscape in BiFeO₃ thin films,^{19,20} limiting this plethora of polar and magnetic states, and eventually promoting binary switching in which two polar states are coupled to two distinct magnetic states.

Despite two decades of intensive research on magneto-electric coupling in BiFeO₃, direct and reversible electric-field modification of the magnetic ground state remains an outstanding challenge. To date, electric field has been shown primarily to redistribute antiferromagnetic domain populations²¹ or manipulate magnetic polarity in strain-engineered heterostructures.²² In one instance, electric-field application enabled the emergence of different spin configurations in (111)-oriented, remarkably thick single-domain BiFeO₃ films, although internal fields within the film may also contribute.²³ However, the (111) crystallographic orientation is known for rapid polarization fatigue,²⁴ making it unsuitable for memory devices. In a separate case, electric field modulated the magnetic state under large capacitors.²⁵ These effects rely on constrained ferroelastic switching conditions (71° switching)

and are similarly demonstrated in relatively thick films, limiting their translation to dense and energy-efficient memory architectures. At the MESO device level, transduction or readout of the magnetic state typically necessitates an additional ferromagnetic layer, complicating device architecture and integration.^{6,26} Indeed, the voltage output tunability is limited owing to the spin polarization of the ferromagnet and the exchange coupling between the ferromagnet and BiFeO₃. Therefore, using magnon transport within BiFeO₃ itself has opened promising pathways recently.^{27–30} Consequently, a strong intrinsic voltage-driven magnetic phase transition in single-domain thin BiFeO₃ layer, amenable to direct electrical readout, has yet to be realized, calling for an innovative engineering approach.

We therefore turned to the BiFeO₃/La_{0.7}Sr_{0.3}MnO₃ bilayers, a system which is known by the emergence of strongly asymmetric interfacial physical properties governed by the ferroelectric polarization direction in BiFeO₃. Beyond charge-driven conductivity and magnetic phase transitions in La_{0.7}Sr_{0.3}MnO₃ induced by adjacent ferroelectric layers,³¹ coupling with BiFeO₃ has also been shown to produce exchange bias effects, possibly influenced by the cycloidal spin structure.^{32,33} These effects were only shown at temperatures well below practical device operation. While the precise origin of this phenomenon remains unclear, the experimental evidence suggests that reversible structural and electronic reconstructions in BiFeO₃ play a significant role.^{34–36} These findings point to the potential for electric-field control of the antiferromagnetic state in BiFeO₃, mediated by changes at the BiFeO₃/La_{0.7}Sr_{0.3}MnO₃ interface.

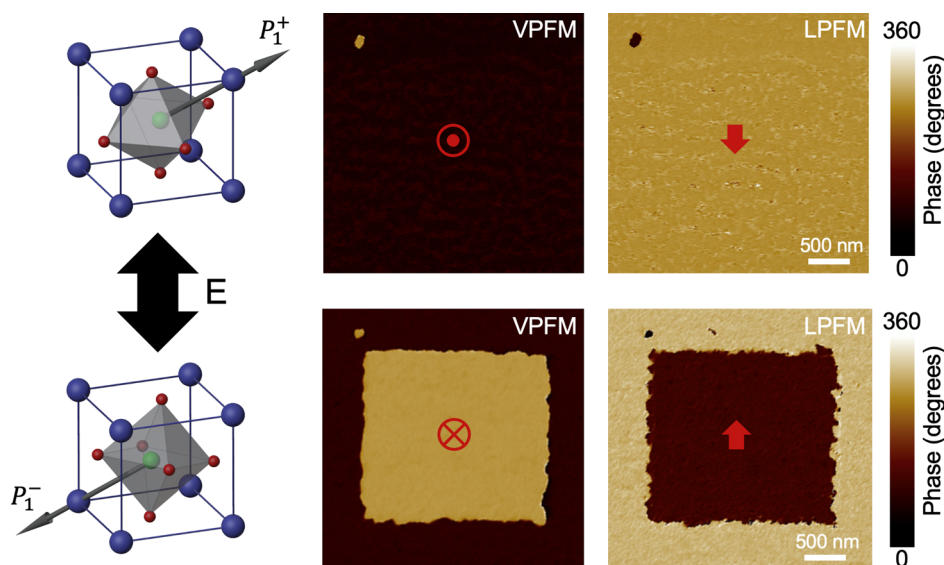


Figure 2. Deterministic control of the ferroelectric polarization state. Using an electric field, the polarization of the single-domain as-grown ferroelectric state (top row) is switched by 180° to another single domain ferroelectric state (bottom row). The left column shows the sketch of the two ferroelectric states, P_1^+ and P_1^- . The middle column shows the vertical PFM (VPFM) and the right column the lateral PFM (LPFM). The ferroelectric state has been switched (bottom row) by scanning the PFM tip with a negative voltage applied to the bottom $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ electrode in a $2 \times 2 \mu\text{m}^2$ square.

RESULTS AND DISCUSSION

Here we show that the multiferroic textures of BiFeO_3 can be drastically simplified by growing the films onto a vicinal substrate. The BiFeO_3 thin films were grown by pulsed laser deposition (Methods). Single crystal $\text{SrTiO}_3(001)$ substrates, with a 4° miscut along the $[110]$ direction, were prepared. A buffer oxide electrode of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ was first grown and reflection high-energy electron diffraction oscillations were used to monitor the growth, yielding an electrode thickness of 5.5 nm (15 unit cells). Subsequently, a BiFeO_3 film with a thickness of 18 nm was deposited. X-ray diffraction reveals that the films are compressively strained by the SrTiO_3 substrate and show a single ferroelastic domain (Figure S1). Furthermore, reciprocal space maps, performed along symmetric $\text{SrTiO}_3(002)$ or asymmetric $\text{SrTiO}_3(113)$ peaks, show a small misalignment of 0.2° of the $\text{BiFeO}_3(001)$ planes with respect to the SrTiO_3 in the direction of the monoclinic distortion while the film is coherently strained perpendicularly to this direction. The resulting compressive epitaxial strain is estimated to about −1.3% (−1.4%) along (perpendicularly to) the monoclinic in-plane direction (Figure S1).

Scanning transmission electron microscopy (STEM) and scanning probe microscopy were performed to get some insights into the polar and spin textures of the films at the nanometer scale (Figure 1, Methods). STEM cross section images show how the large miscut of the SrTiO_3 substrate is accommodated by the $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ and BiFeO_3 layers (Figure 1a, bottom). The large miscut angle of the SrTiO_3 substrate generates strong ferroelastic distortions in BiFeO_3 with local thickness fluctuations between 15 and 25 nm. Piezoresponse force microscopy (PFM) reveals that the as-grown state of the films displays a uniform vertical piezoresponse (inset of Figure 1b), with the dark phase contrast corresponding to an upward polarization state. Furthermore, the lateral PFM signal is also uniform (Figure 1b) and is consistent with a single ferroelectric domain state. The in-plane component of polarization is perpendicular to the

step edges of the miscut substrate, as confirmed by STEM (Figure 1a, top) and illustrated by the sketch in Figure 1b. Using scanning NV magnetometry, we imaged the magnetic stray field corresponding to this single-domain ferroelectric state (Figure 1c). A periodic magnetic stray field is clearly visible, in agreement with the presence of the antiferromagnetic spin cycloid of BiFeO_3 . The high ordering of the magnetic pattern (single period and propagation direction) suggests the presence of a single spin cycloid in the film with a propagation vector lying in the film plane, along the step edges of the miscut substrate. The cycloidal period is estimated to 86 ± 5 nm from several scanning NV magnetometry images acquired at random locations. This increased cycloidal period, with respect to that of bulk BiFeO_3 (~62 nm), is attributed to the influence of compressive epitaxial strain, in line with previous experiments.^{13,37} To further confirm the single spin cycloid nature of the films on a macroscopic scale (several hundreds of microns), we used resonant elastic X-ray scattering (REXS).^{13,17,19} REXS was performed at the Fe L edge (Methods). The dichroic diffraction pattern at the Fe L edge is displayed in Figure 1d. A very clear pair of intense and narrow spots at +q and −q, symmetric with respect to the specular beam, is observed on the REXS pattern, with a very strong magnetic circular dichroism (Figure S2). This REXS pattern attests for the presence of a periodic antiferromagnetic structure with a single propagation direction all over the probed area (typically $50 \times 100 \mu\text{m}^2$). The corresponding period of the spin cycloid is estimated to be 88 ± 2 nm, in excellent agreement with real space measurements performed using NV magnetometry. Thus, the combination of X-ray diffraction, PFM, NV magnetometry and REXS corroborates the presence of a single ferroelectric domain coupled to a single spin cycloid in our $\text{BiFeO}_3(001)$ thin films epitaxially grown on vicinal $\text{SrTiO}_3(001)$ substrates.

We now explore the influence of the electric field on the ferroelectric and magnetic states of these single-domain multiferroic films. Using the scanning PFM tip connected to

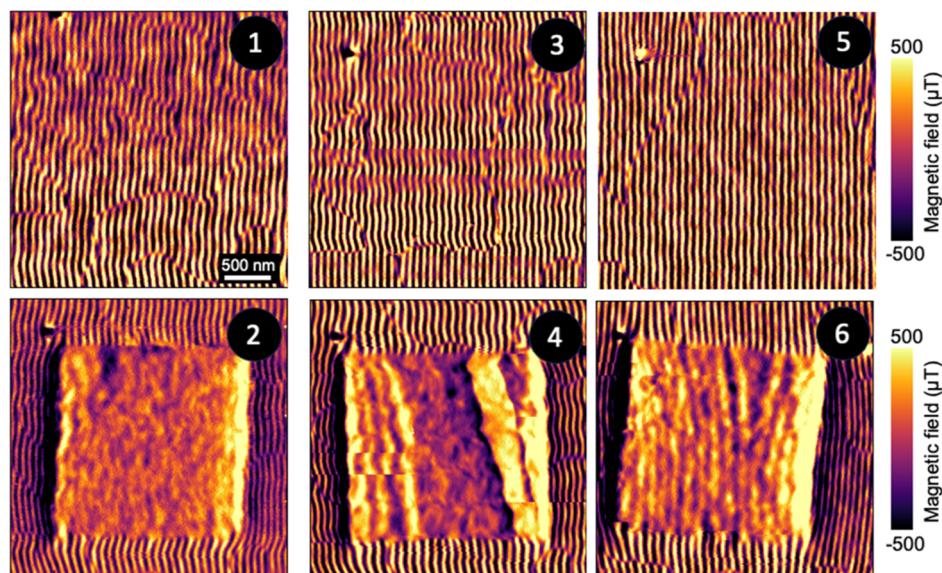


Figure 3. Nonvolatile deterministic control of the antiferromagnetic order. Scanning NV magnetometry images of the same area in the as-grown (1) and switched polarization states (2–6) in a $2 \times 2 \mu\text{m}^2$ square at the center of the images. Images (2), (4) and (6) correspond to the P_1^- polarization state, while images (1), (3), (5) correspond to the P_1^+ polarization state. The same area is testified by the small defect located of the top left corner of the images. It can be clearly seen that switching between the two polarization states leads to a reversible switching of the antiferromagnetic order from cycloidal to noncycloidal states.

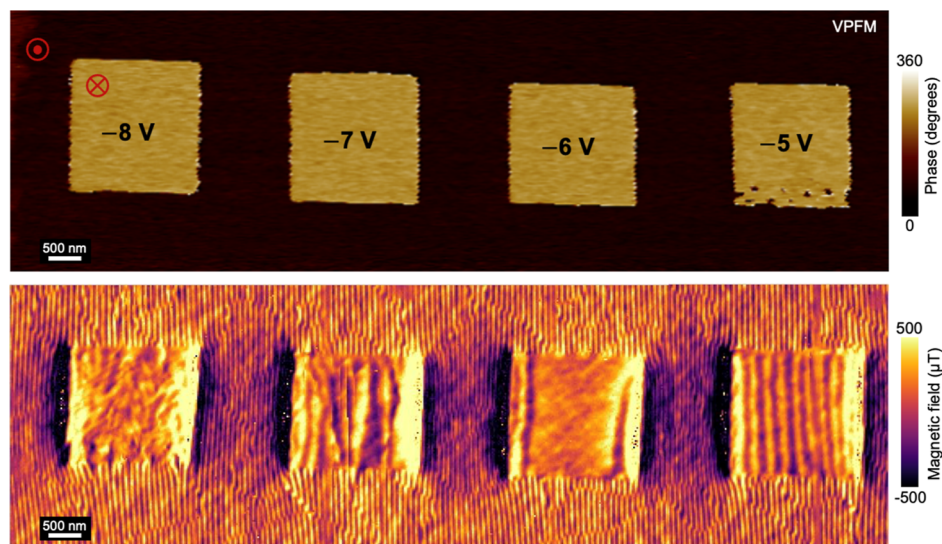


Figure 4. Electric field control of the antiferromagnetic order. Ferroelectric polarization is reversed by scanning the PFM tip in four different $2 \times 2 \mu\text{m}^2$ square areas using voltages of -8 V down to -5 V . The resulting pattern is visualized by vertical PFM (top row). The corresponding magnetic field pattern by scanning NV magnetometry (bottom row) clearly shows a periodic stray field only in the area written with the lowest voltage. This suggests that an incomplete switching of polarization through the thickness of the film in the -5 V area, gives rise to a cycloidal order from the unswitched part of the film.

the ground, we applied a negative dc voltage to the bottom electrode of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ to reverse the out-of-plane component of polarization from upward to downward. Figure 2 shows that the application of this electric field in a $2 \times 2 \mu\text{m}^2$ square results in a change of both the vertical and lateral components of PFM (Figure S3). As sketched in Figure 2a, this corresponds to a 180° reversal of the ferroelectric polarization from the as-grown P_1^+ to the written P_1^- . Hence, the ferroelastic domain favored by the mechanical boundary conditions of the vicinal substrate is preserved throughout the electric-field switching process. We systematically observe the same 180° reversal of polarization upon the application of the

electric field, in contrast to previous observations on thick BiFeO_3 films grown on miscut SrTiO_3 which showed 71° switching of polarization.²⁵ This 180° reversal does not depend on the direction of the trailing field while scanning the PFM tip on the surface with a dc voltage (Figure S3). Thus, the application of the electric field results in a deterministic control of the ferroelectric polarization, with a unique 180° switch being allowed in our heterostructures.

In order to examine the influence of such deterministic control of polarization on the magnetic textures, we used scanning NV magnetometry. Our protocol consists in sequentially writing and reading a $2 \times 2 \mu\text{m}^2$ square with

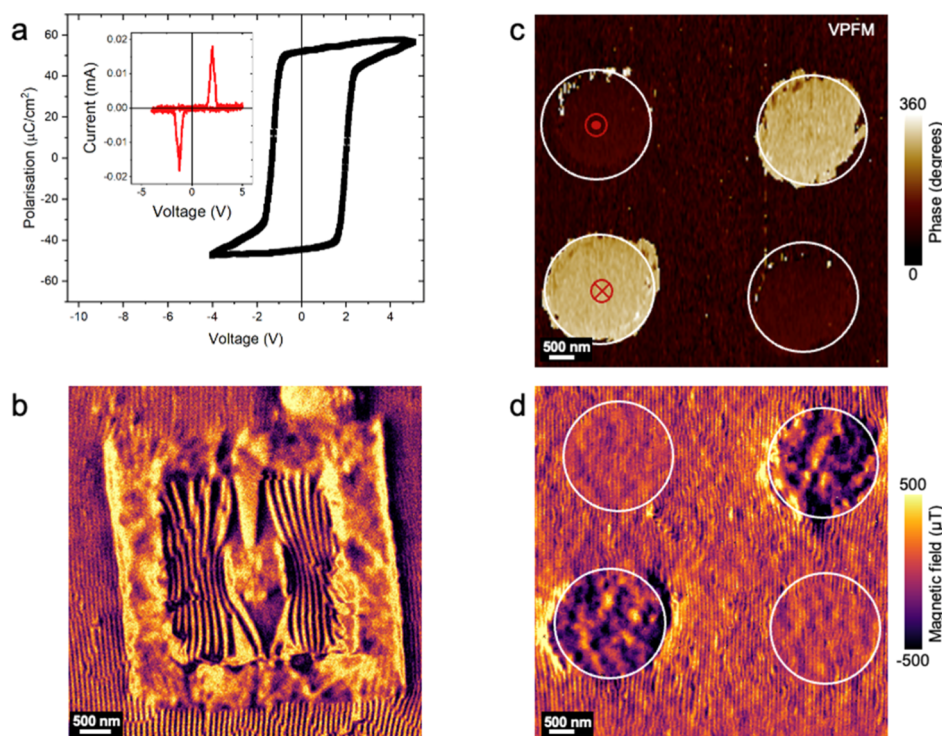


Figure 5. Antiferromagnetic phase transition: reconfigurability and solid-state devices. (a) Polarization vs voltage loop obtained from a Pt/BiFeO₃/La_{0.7}Sr_{0.3}MnO₃ capacitor grown on a SrTiO₃(001) substrate with 4° miscut. The diameter of the capacitor is 8 μm. The triangular wave voltage is applied at a frequency of 1 kHz. The inset shows the sharp peaks of the displacement current as a function of voltage. (b) Scanning NV magnetometry image of a box-in-a-box pattern in which the polarization was sequentially reversed downward (4.5 × 4.5 μm²), upward (3 × 3 μm²) and downward (1 × 1 μm²). The antiferromagnetic phase appears to be cycloidal (noncycloidal) when the polarization is upward (downward). (c) Vertical PFM image of four Pt/BiFeO₃/La_{0.7}Sr_{0.3}MnO₃ capacitors with diameters of 2 μm each. Two of the capacitors were electrically switched (Figure S8). (d) Scanning NV magnetometry image of the same area as in (c) showing an antiferromagnetic cycloid in capacitors with upward polarization and a nonperiodic magnetic stray field in capacitors with downward polarization.

PFM, transferring it to NV magnetometry, and then back to PFM to further reverse the ferroelectric domain state. Figure 3 summarizes the results of this sequence of measurements on the magnetic state of the films. State “1” corresponds to the virgin state of polarization in which a single antiferromagnetic cycloid is clearly displayed. However, upon the reversal of polarization in state “2”, the cycloid can no longer be observed. Instead, the absence of a periodic magnetic stray field, with field divergence at the written square edges, suggests the presence of a collinear canted antiferromagnetic state (Figure S4). Interestingly, reversing the polarization back in the same area with the electric field triggers the complete restoration of the spin cycloid, as visible from state “3”. The same observations can be established from states “4”, “5”, and “6”. We can make a few comments with respect to the magnetic switching behavior. First, it seems that some defects, that may be related to stacking faults of the spin density wave locked to the spin cycloid, are annihilated in the different writings restoring the P_1^+ state, as visible when comparing states “1”, “3”, and “5” in Figure 3. In addition, the magnetic stray field seems to vary in the switched P_1^- polarization, when comparing states “2”, “4”, and “6”. Indeed, different collinear antiferromagnetic domains may be expected within a single ferroelectric domain state, as permitted by the symmetry of BiFeO₃. State “6” may also reveal a periodic magnetic stray field with a larger period, as possibly caused by the presence of a destabilized spin cycloid. A similar, single domain ferroelectric state coupled to a single spin cycloid could be obtained

in epitaxial BiFeO₃(111) thin films grown under anisotropic strain on DyScO₃(011) orthorhombic substrates,¹⁹ but without showing any magnetic transition coupled to ferroelectric switching.

To assess the robustness of the observed magnetic states with respect to the writing conditions in BiFeO₃, we performed combined PFM and NV experiments in which the amplitude of the electric field was gradually decreased during the writing process. Figure 4 shows the PFM and NV images of a large area in which four 2 × 2 μm² square domains were switched with voltages of −8 V, −7 V, −6 V and −5 V, from left to right, respectively. At first, vertical PFM images did not show any striking differences between the domains written with different electric fields. However, scanning NV magnetometry shows that, while the first three domains with P_1^- indicate the absence of any periodic magnetic stray field related to a spin cycloid, the last written domain with the smallest electric field clearly displays a periodic magnetic stray field, albeit with a very large period (210 ± 5 nm). This suggests that an incomplete switching of ferroelectric polarization throughout the BiFeO₃ film thickness can lead to a periodic magnetic stray field linked to the spin cycloid of the remaining P_1^+ state. For instance, assuming that polarization reversal gives rise to a canted G-type antiferromagnetic state, a thinner unswitched layer with a cycloidal state would not be detected in the PFM response but would dominate the magnetic stray field detected by the NV sensor (Note S1). This would also be the case if the

ferroelectric polarization of the film is partially relaxing after switching.

In addition, we looked at the possible influence of the ultrathin $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ on the magnetic stray field observed from our heterostructures. Magnetic field dependent scanning NV magnetometry was performed in an area containing a reversed P_1^- state of BiFeO_3 , applying an external magnetic field along the hard and easy axes of the ultrathin $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ ferromagnet (Figure S5). It clearly appears that saturating the $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ magnetization does not have an influence on the magnetic texture emanating from this P_1^- state. Indeed, no signature of an antiferromagnetic cycloid is recovered while applying a magnetic field, confirming the existence of this electric-field induced antiferromagnetic phase transition from a cycloidal to a noncycloidal state in BiFeO_3 .

In principle, multiple mechanisms might explain these observations, as our P_1^+ and P_1^- states are by construction nonequivalent by symmetry, on account of the $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ bottom layer and the bare BiFeO_3 top surface. For example, a polarization-dependent interfacial coupling to $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ may impact BiFeO_3 's magnetic state,^{32,38} and its possible role cannot be excluded. However, we suggest that the observed switching between cycloidal and canted-antiferromagnetic states may be directly coupled to structural differences between the P_1^+ and P_1^- states. Indeed, the observed P_1^+ as-grown state is reminiscent of the theoretical discussion by Efe et al. on the interplay between BiFeO_3 surfaces and the orientation of the polarization.³⁹ These authors showed that a film starting with a BiO plane, which is positively charged, can be expected to have a polarization pointing up (like in our P_1^+ state) so that the electrostatic energy is minimized. By contrast, in such a film, the P_1^- is unstable, unless external compensating (and positive) charges are brought to the surface. This theoretical expectation matches what we observe in our films, where the P_1^- is field-induced and eventually unstable upon removal of the applied voltage. Nevertheless, for domains with P_1^- polarization written with a sufficiently large voltage, both the ferroelectric and antiferromagnetic states were not shown to relax even after 6 days (Figure S6).

Given all this, it seems reasonable to assume that the P_1^+ and P_1^- states in our films will be structurally different; in particular, the out-of-plane polarization can be expected to be larger for the P_1^+ state. Given the well-known coupling between polarization and octahedral tilts in BiFeO_3 , a smaller polarization should immediately result in larger tilts.⁴⁰ Critically, such structural differences can be expected to have a definite impact in the magnetic configuration. Indeed, as described in Note S2, one can discuss this question by introducing a minimal spin Hamiltonian consisting of the antiferromagnetic superexchange and two types of Dzyaloshinskii–Moriya interactions (DMI). The first DMI couples spins at first nearest-neighbors leading to their canting, and its magnitude is proportional to the octahedral tilts.⁴¹ The second DMI couples second-nearest-neighboring spins favoring the cycloidal modulation, and its magnitude is proportional to the polarization. As we show in Note S2, we can use experimental information^{11,42} to derive values for these three couplings in bulk BiFeO_3 ; the results are consistent with the known spin cycloid being the most stable magnetic state. At the same time, our approximate model suggests that a reduction of 30% to 40% in the polarization would be sufficient for the canted

solution to dominate. We also see that intermediate situations could yield a cycloid of extended period, as e.g., shown in Figure 4 for an applied voltage of -5 V.

In order to demonstrate the reconfigurability of the system after multiple switching, we designed a box-in-a-box pattern (Figure S5b). The polarization was sequentially reversed downward ($4.5 \times 4.5 \mu\text{m}^2$), upward ($3 \times 3 \mu\text{m}^2$) and downward ($1 \times 1 \mu\text{m}^2$), so that in the inner square the polarization has been switched three times. We observe that we can destroy and restore the antiferromagnetic spin cycloid on demand, opening a path for reconfigurable devices. Furthermore, the period of the spin cycloid appears to increase when it is confined between noncycloidal domains (Figure S7). To further expand the potential of this deterministic switching, we designed more application compatible solid-state capacitors. This may also provide more favorable electrical boundary conditions to stabilize both upward and downward polarization states because of the more efficient screening of BiFeO_3 surfaces charges. Pt top electrodes were defined by optical lithography and lift-off on top of the continuous BiFeO_3 layer with typical sizes ranging from 1 to 10 μm . Figure 5a shows a polarization vs voltage loop measured in an 8 μm capacitor, in which displacement currents are associated with the polarization reversal from P_1^+ and P_1^- and vice versa. Hence, while the individual P_1^+ and P_1^- polarization values cannot be established from these measurements, the average ferroelectric polarization of $50 \mu\text{C}/\text{cm}^2$ is in agreement with polarization values obtained for (001)-oriented films.⁴³ If such top electrodes are thin enough, i.e. few nanometers in thickness, we are still able to probe both ferroelectric and antiferromagnetic landscapes through the electrode, after switching back and forth a given capacitor device. Figure 5c shows a 2×2 capacitor array with 2 μm diameters, imaged by vertical PFM. In all the capacitors, the ferroelectric polarization is upward as for the free surface. This is illustrated by the two, up-left and down-right, capacitors displaying dark VPFM phase. In contrast, the two others, up-right and down-left, display a bright VPFM phase contrast as they were electrically switched with a dc voltage pulse. In this capacitor geometry, the ferroelectric reversal is still deterministic with a polarization switching by 180° (Figure S8). Scanning NV magnetometry was performed on this 2×2 capacitor array (Figure Sd). Here again, we observe the destruction of the spin cycloidal order upon the reversal of ferroelectric polarization, while this spin cycloid appears unaffected by the covering capacitors. Switching back the ferroelectric polarization to the upward state fully restores the pristine cycloidal state (Figure S8).

CONCLUSIONS

To summarize, we observe that in single-domain epitaxial BiFeO_3 thin films grown on $\text{SrTiO}_3(001)$ miscut substrates, the application of an electric field results in a fully deterministic and reversible switching of polarization. Using scanning NV magnetometry and resonant elastic X-ray scattering, we observe that the as-grown state corresponds to a single domain antiferromagnetic spin cycloid. Reversing the ferroelectric polarization results in the destruction of this spin cycloid and the stabilization of a canted collinear antiferromagnetic state. This antiferromagnetic phase transition is further observed in solid-state capacitors, opening the path to functional devices based on magnetoelectric antiferromagnetic elements. This

result constitutes a key building block for the realization of low-power magnetoelectric spin–orbit devices.

We have demonstrated an electric-field-driven magnetic phase transition in BiFeO₃ heterostructures engineered through step-induced multiferroic domain selectivity. The toggling between two distinct antiferromagnetic states is associated with a 180° switching of ferroelectric polarization. The effect may originate from the reversible modulation of polarization magnitude and oxygen octahedral tilts,³⁴ coupled to the spin cycloid. From an applications perspective, this switching pathway is particularly attractive for magnetoelectric memory concepts because of the absence of ferroelastic lattice distortion and domain-wall formation during the switching process. In particular, such control has shown long retention and strong fatigue endurance both essential for memory application.⁴⁴ Our results therefore point to a magnetoelectric switching pathway that offers a more favorable route toward low-power and energy-efficient memory operation.

Our results extend a possible path for direct readout of the memory state using spin–orbit materials integrated with the BiFeO₃ heterostructure. In this picture, the antiferromagnetic state written in the magnetoelectric layer could be transduced into an electrical signal by strong spin–orbit materials or topological insulators, such as Pt, W, SrIrO₃ or Bi₂Se₃, thus enabling a MESO-like architecture without the need for an additional ferromagnetic layer.⁴⁴ Such a strategy would shift the origin of nonvolatility from ferromagnetism to electrically controlled antiferromagnetic order. Beyond simplifying the device stack, the use of heavy metals or oxides with strong spin–orbit coupling could promote high-quality interfaces that are essential for efficient interfacial spin to charge conversion. Given that both the antiferromagnetic⁴⁵ and the ferroelectric⁴⁶ orders can be modulated at the picosecond time scale in BiFeO₃, this device concept could be further extended to the ultrafast regime.

From a more fundamental perspective, this phase transition to a uniform (noncycloidal) antiferromagnetic state may offer a suitable platform for studying electrically induced magnetic states relevant to emerging altermagnetic physics.⁴⁷ While signatures for altermagnetism have been recently reported in ultrathin BiFeO₃ films,^{48–50} the reversible electric-field control of distinct antiferromagnetic states is per se an appealing foundation.

METHODS

Growth of Heterostructures and Device Fabrication

The oxide thin films were grown by RHEED-assisted pulsed laser deposition, using a KrF excimer laser, on single crystal SrTiO₃ (001) substrates with 4° miscut toward [110]. Prior to the deposition, SrTiO₃ (001) was acid-etched by buffered HF for 30 s and heat treated for 2 h at 1000 °C. 5.5 nm (15 unit-cell) thick La_{0.7}Sr_{0.3}MnO₃ was first grown on SrTiO₃ substrates at 750 °C and 0.4 mbar before BiFeO₃ was deposited at 650 °C and 0.36 mbar. Repetition rates were set at 2 and 10 Hz for La_{0.7}Sr_{0.3}MnO₃ and BiFeO₃, respectively. The BiFeO₃ thickness was kept at 18 nm unless otherwise mentioned in the main text. The BiFeO₃ film thickness was estimated from scanning transmission electron microscopy experiments in cross section specimens. In order to prepare top electrodes and define alignment markers for the magnetoelectric investigations, a combined photolithography (Smart Print) and lift-off process were adopted. In all cases, a 20 nm-thick layer of platinum was deposited by magnetron sputtering.

X-ray Diffraction

Conventional 2θ-ω scans and reciprocal space maps were performed using Cu K_{α-1} radiation in a 9 kW rotating anode Rigaku SmartLab diffractometer.

Scanning Transmission Electron Microscopy

Experiments were performed using a Cs-corrected NION STEM operated at 200 keV, with a typical probe size of about 80 pm. High-angle annular dark-field (HAADF-STEM) images were used to map the polar distortion, as estimated by the displacement of the B-cation with respect to the average position of the four neighboring A-type cations. Samples were prepared for STEM measurements using FIB thinning, with progressively decreasing Ga-ion beam voltages from 30 to 2 keV to minimize artifacts.

Scanning Probe Microscopy

Piezoresponse force microscopy (PFM) experiments were carried out on a Bruker Nanoscope V Multimode atomic force microscope equipped with two external SR830 lock-in amplifiers (Stanford Research) for simultaneous detection of the vertical and lateral responses. AC voltage was applied to the La_{0.7}Sr_{0.3}MnO₃ bottom electrode using a DS360 source (Stanford Research) at 35 kHz, with the conductive Pt-coated tip grounded. In order to reverse the ferroelectric polarization of the BiFeO₃ layer, DC voltage was applied to La_{0.7}Sr_{0.3}MnO₃ while scanning the grounded Pt-coated tip.

Scanning NV Magnetometry

Experiments were performed using a commercial scanning NV magnetometer (Qnami, ProteusQ) operated under ambient conditions, with commercial all-diamond scanning-probe tips containing single NV defects (Qnami, Quantilever MX). The tip was integrated into a tuning-fork based atomic force microscope combined with a confocal microscope optimized for single NV defect spectroscopy. Magnetic fields radiated by the sample are detected by recording the Zeeman shift of the NV defect's electronic spin sublevels through optical detection of the electron spin resonance. The scanning NV magnetometer was operated in the full-B imaging mode by continuously monitoring the photoluminescence (PL) intensity and sweeping the applied microwave frequency (ν) at each point of the scan. A Lorentzian fit on PL(ν) then provides the aforementioned local resonance frequency. Experiments were performed with a NV-to-sample distance of approximately 50 nm and a bias magnetic field of 1 mT applied along the NV quantization axis. Additional experiments were performed with a Quantum Scanning Microscope from Qzabre to look at the influence of an external magnetic field onto the magnetic stray field of the structures (Figure S5).

Resonant Elastic X-ray Scattering

The experiment was performed at SOLEIL synchrotron on the SEXTANTS beamline using the RESOXS diffractometer. The diffractometer is equipped with a charge-coupled device (CCD) camera (2048 × 2048 pixels of 13.5 μm) located on the detector arm at 26 cm from the sample. The solid angle probed by the camera alone is 6.3°. A set of beamstops placed a few millimeters before the CCD chip could be precisely aligned with few 10 s of nm precision in order to block the specular beam. Data were collected at the Fe L edge using circularly polarized X-rays delivered from an Apple 2 undulator located at the I14 M straight section of the SOLEIL storage ring. A typical photon flux of 10¹³ ph s⁻¹ with 100 meV energy resolution was used for the experiment. Topological structures display a dichroic signal that is of opposite sign in +q and -q. This depends on the sign of the chirality and it is therefore linked to the antiferromagnetic winding in the sample. This provides us with another means to further confirm that the dichroism stems from the antiferromagnetic cycloids.

Ferroelectric Measurements

Polarization vs electric field hysteresis loops were recorded at room temperature with a Radiant Multiferroic tester.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsnano.6c09046>.

Note S1 (with Table S1): influence of the film thickness on the cycloid period. Note S2: analytical model used to describe the transitions between cycloid and canted antiferromagnetic states. Figure S1: structural characterization of the films using X-ray diffraction. Figure S2: results from resonant elastic X-ray scattering. Figure S3: electric-field writing and PFM reading of ferroelectric domains. Figure S4: results from simulations of the magnetic stray field induced by a single-domain canted antiferromagnetic state. Figure S5: influence of the magnetic field on scanning NV images. Figure S6: relaxation of domains depending on the electric field used to write them and their influence on the scanning NV magnetometry images. Figure S7: influence of the size of written ferroelectric domains on the shape of the magnetic stray field. Figure S8: PFM and scanning NV magnetometry images on solid-state devices written in different ferroelectric states (PDF)

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Author Contributions

♦A.A. and N.R. contributed equally to this work. A.A. conceived the idea and proposed the experiments. V.G. coordinated the project and supervised the experiments. A.A. grew the epitaxial thin films. A.A., C.C., and V.G. performed the X-ray diffraction experiments and analyzed the structure of the films. A.A., S.F. and V.G. performed the piezoresponse force microscopy experiments. N.R., A.C. and K.B. performed the scanning NV magnetometry experiments. A.F. performed the simulations of the magnetic stray field. N.J., J.-Y.C., M.V., V.G. and S.F. performed the resonant elastic X-ray scattering experiments. O.C., J.-Y.D., X.L. and A.G. prepared the cross-section specimen by focused ion beam, examined the samples by scanning transmission electron microscopy, and calculated the polar displacements. S.F., A.A., and F.G. fabricated the devices and defined the markers by photolithography and sputtering. C.A.M.S. and J.I.-G. derived the analytic model to investigate the stability of the different antiferromagnetic states. All the authors, including E.W., R.M., B.D., and V.J. participated to the analysis, the discussion and interpretation of the results. V.G. wrote the manuscript together with A.A. and S.F.

Notes

The authors declare no competing financial interest.

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