Probing electric field control of magnetism using ferromagnetic resonances

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Exchange coupled CoFe/BiFeO\textsubscript{3} thin-film heterostructures show great promise for power-efficient electric field-induced 180° magnetization switching. However, the coupling mechanism and precise qualification of the exchange coupling in CoFe/BiFeO\textsubscript{3} heterostructures have been elusive. Here we show direct evidence for electric field control of the magnetic state in exchange coupled CoFe/BiFeO\textsubscript{3} through electric field-dependent ferromagnetic resonance spectroscopy and nanoscale spatially resolved magnetic imaging. Scanning electron microscopy with polarization analysis images reveal the coupling of the magnetization in the CoFe layer to the canted moment in the BiFeO\textsubscript{3} layer. Electric field-dependent ferromagnetic resonance measurements quantify the exchange coupling strength and reveal that the CoFe magnetization is directly and reversibly modulated by the applied electric field through a ~180° switching of the canted moment in BiFeO\textsubscript{3}. This constitutes an important step towards robust repeatable and non-volatile voltage-induced 180° magnetization switching in thin-film multiferroic heterostructures and tunable RF/microwave devices.

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FMR measurements of CoFe/BFO. Field-dependent FMR at X-band (9.5 GHz) under different applied voltages (across the BFO layer) was measured as schematically shown in Fig. 2a, with $\alpha$ being the angle between the applied magnetic field direction and the net magnetic easy axis of CoFe layer. The sample was rotated from 0° to 360° in the FMR measurement system. Voltage pulses (100 ms) with varying amplitudes from 0 to $-0.5$ V were applied across the BFO layer and all the FMR spectra at open circuit after each voltage pulse were recorded at a fixed $\alpha$. As shown in Fig. 2b, we observe a large voltage-induced FMR field shift (the difference between blue and red curve at zero crossing) of $0 \pm 1$ mT after applying $+0.5$ V (red) and $-0.5$ V (blue) voltage pulse amplitudes. By tracing the FMR field (that is, the zero crossing of the magnetic field of the FMR spectrum in Fig. 2b) after applying different voltage pulses, we obtain the dependence of the FMR field on the voltage pulse amplitude. The upper right inset of Fig. 2b shows a typical square, hysteretic dependence on voltage. Figure 2c shows the FMR field dependence on the applied voltage pulses along different magnetic field orientations, $\alpha = -45^\circ$, 0°, 45° and 90°, from the CoFe layer magnetic easy axis. The largest voltage-induced FMR field change is $0 \pm 1$ mT at $\alpha = 0^\circ$, that is, when the magnetic field was applied along the easy axis. At $\alpha = -45^\circ$ or 45°, we observe a relatively smaller FMR field shift with the maximum FMR field being $0 \pm 1$ mT. The hysteretic FMR field dependence on voltage pulse amplitude means that it is switched back and forth by voltage pulses between two states, identified as the +0.5 V state and -0.5 V state in Fig. 2c. However, there is no obvious hysteretic behaviour at $\alpha = 90^\circ$, and the maximum FMR shift is comparable to the measurement noise level ($\approx 0.5$ mT).

To test the repeatability of the voltage control of the FMR field shift, we then apply a sinusoidal voltage signal with an amplitude of 10 V (frequency= 0.167 Hz) across the thickness direction of the BFO layer. We measure the dependence of the FMR field on applied voltage pulse after switching the voltage 50 and 100 times by the sinusoidal as shown in Fig. 2d. Similar hysteresis loops of the FMR field shift at $\alpha = 0^\circ$ are observed after 50 and 100 ferroelectric switching cycles indicating a robust, non-volatile and repeatable voltage-induced FMR field shift.

Figure 3a represents the schematic of CoFe/BFO bilayer two-domain stripe-like structure, in which the magnetization of CoFe can be tuned by an OOP E-field. While Fig. 3b shows the corresponding IP (left) and OOP (right) piezoelectric microscopy (PFM) images with polarization orientations manipulated by an OOP E-field. Figure 3c shows the angular dependence of the magnetic coupling and illustrates the main result of our measurements. After the +5 V pulse was applied across the BFO layer, the FMR field dependence on $\alpha$ was switched from the blue curve (0 V state) to the red one (+5 V state) at a fixed $\alpha$ angle, corresponding to the +0 V state of FMR field. A subsequent -5 V voltage pulse switched the angular dependence of the FMR field back to the blue curve (-0 V state). The angular dependence of the blue curve indicates that the CoFe magnetization is parallel to the effective magnetic coupling field from BFO near 0° and 180°. On the other hand, the angular dependence of the red curve shows that by changing the electric polarization, the CoFe magnetization, which has not changed due to the large magnetic field applied for the FMR measurements, now lies in a direction that is antiparallel to the effective magnetic coupling field from BFO, leading to a higher FMR field.

The above experimental observations cannot be explained by conventional magnetoelectric coupling mechanisms, such as charge-mediated magnetoelectric coupling or strain-mediated magnetoelectric coupling, for the following reasons. The IP anisotropic FMR field dependence on applied voltage along different magnetic field orientations is not likely to originate from interfacial charge-screening effects, even though the BFO layer has a high polarization. Charge-screening effect provides an effective OOP magnetic anisotropy to the magnetic thin film, therefore, the voltage-induced IP FMR field shift due to charge-screening effect should be isotropic with varying $\alpha$, which is in contrast to our measurements. The observed anisotropic voltage dependence of the FMR field is also not likely to be dominated by strain-mediated magnetoelectric coupling, as strain-mediated magnetoelectric coupling will be diminished by substrate clamping effects. Furthermore, it has been previously demonstrated that the coupling between the BFO and CoFe vanishes when a few nm thick, fully strained SrTiO$_3$ layer is interposed between them.
uch recent research has been devoted to realizing fast, compact and low-power magnetoelectric random access memories (MERAM) based on electric field (E-field) control of magnetization. Multiferroic materials with simultaneous ferroelectricity and magnetism provide a pathway to achieving strong magnetoelectric coupling with efficient voltage control of magnetism, and compact and power-efficient electric field-tunable magnetic devices. A variety of ‘magnetoelectric-multiferroics’ such as magnetic/ferroelectric and magnetic/multiferroic heterostructures have been investigated, which are providing pathways to novel electric field-tunable radio frequency (RF/microwave signal processing devices, magnetic field sensors, MERAM devices and voltage-tunable resistive memory devices.

Several magnetoelectric coupling mechanisms have been explored to achieve efficient electric field control of magnetism, for example, strain-mediated magnetoelectric coupling and spin-polarized charge-mediated magnetoelectric coupling and voltage control of carrier-mediated magnetism in dilute magnetic semiconductors. Strain-mediated magnetoelectric coupling in thin-film magnetic/ferroelectric heterostructures on substrates can be diminished due to substrate clamping effects, while spin-polarized charge-mediated magnetoelectric coupling can only be observed in ultra-thin (<1 nm) magnetic thin films, which puts a limit on its application in real magnetoelectric devices. Finally, voltage control of carrier-mediated magnetism in dilute magnetic semiconductors has been challenging at room temperature.

The room temperature single-phase multiferroic, BiFeO₃ (BFO), has attracted a lot of recent research interest due to the coexistence of robust ferroelectricity (P) and antiferromagnetism (M) and a weak canting magnetic moment (M₀). To bulk BFO, the weak moment results from the canting of the magnetic sublattices due to the Dzyaloshinskii–Moriya interaction as predicted by the density functional theory and confirmed experimentally. E-field control of magnetism, magnetoresistance, magnetic anisotropy and magnetization in a ferromagnetic layer exchange coupled to BFO layer has been most recently reported. Nevertheless, one fundamental question has remained open: does the canting moment in this system reverse direction on the application of an electric field? In a recent paper, we used a combination of ab initio computation, magnetotransport measurements and photoemission electron microscopy studies to demonstrate that the canting moment can indeed switch by 180° when the ferroelectric polarization is switched by 180°, particularly in a (001)-oriented epitaxial film that is comprised of a periodic ferroelectric domain pattern.

To emphasize and validate the results of these studies, we carried out a very detailed electric field-dependent ferromagnetic resonance (FMR) study of exactly the same heterostructure (CoFe/BFO). The FMR results directly validate, confirm and quantify the electric field-dependent magnetotransport results. Phase-field-based theoretical studies present complementary validation of the experimental results and are consistent with the ab initio theoretical results.

FMR of the CoFe layer is used to probe both the coupling between the CoFe and BFO and the electric field modulation of this coupling. We find that an out-of-plane (OOP) electric field on BFO induces an anisotropic in-plane (IP) change in the CoFe layer FMR field, which is ±1 mT along the magnetic easy axis of the CoFe layer and ±5 mT along the magnetic hard axis. The switching of the FMR field, which occurred at the same voltage as the switching of the BFO ferroelectric polarization, is repeatable over at least 100 reversals. This robust and repeatable switching in the CoFe/BFO heterostructure provides a potential mechanism for compact and power-efficient MERAM and voltage tunable RF/microwave devices. We provide a model for the modulation of the FMR field as a consequence of the $\approx 180°$ switching of the canted moment of the BFO layer that is exchanged coupled to the CoFe layer. Further, we extracted the effective coupling field ($\approx 5.7 \pm 0.4$ mT) and canted moment switching angle ($178° \pm 11°$) quantitatively by FMR measurements. Voltage tunable magnetic film/BFO heterostructures enable next generation of compact, fast, energy-efficient and voltage-controllable MERAM devices. Moreover, in this work, the tunability of CoFe at X-band RF/microwave frequency also provides the opportunity to realize voltage tunable RF/microwave devices, such as filters, phase shifters and antennas, which are widely used in aircraft, satellite, radar and mobile communication systems.

**Results**

**Preparation and imaging of CoFe/BFO.** Ferromagnetic-multiferroic thin-film heterostructures of Pt (2.5 nm)/Co$_{80}$Fe$_{20}$(2.5 nm)/BiFeO$_3$ (200 nm)/SrTiO$_3$ (STO) were grown onto (110)-oriented DyScO$_3$ (DSO) substrates with a SRO layer as the bottom electrode using pulsed laser deposition. The resulting epitaxial BFO layer exhibits a characteristic, two-domain structure consisting of a one-dimensional, quasi-periodic array of $\pm 1\bar{1}$ domains, using this as a model system to probe the coupling to the ferromagnetic CoFe. The Pt (2.5 nm)/Co$_{80}$Fe$_{20}$(2.5 nm) bilayers were deposited on BFO films under a 20-nm in situ bias magnetic field along the DSO (1–10) direction, leading to a well-defined IP magnetic easy axis in the CoFe layer.

The ferroelectric domain structure of the BFO layer was imaged by measuring the intensity of the back-scattered electrons as shown in Fig. 1a. We image the magnetic structure of the CoFe layer at zero magnetic field using scanning electron microscopy with polarization analysis (SEMPA). By measuring the spin polarization of secondary electrons, SEMPA directly measures the local vector magnetization in the CoFe layer. An example of an IP magnetization image is shown in Fig. 1b. Comparison of the SEMPA and back-scattered electron images clearly shows the close correspondence between the magnetic structure of the CoFe film and the quasi-periodic, striped ferroelectric domain structure of the underlying BFO layer.

**Micromagnetic modelling of this structure.** Supplementary Fig. 1 and Fig. 2, confirms this interpretation and yields an effective interfacial coupling field of $\approx 7$ mT (ref. 17). The striped structure forms two types of magnetic domains in this as-prepared remnant state, one with the net magnetization parallel to the net IP polarization direction (yellow/red striped region in Fig. 1c) and the other with net IP magnetization antiparallel (blue/cyan striped region). The coexistence of parallel and antiparallel magnetization orientations, along with other measurements, demonstrates that the magnetic coupling to the BFO leads to a net uniaxial rather than unidirectional anisotropy in the CoFe layer.
Theoretical analysis of E-field control of CoFe/BFO. To understand the voltage-dependent FMR data, we consider a CoFe layer exchange coupled to the canted moments of a BFO layer within the two ferroelectric domains as shown in Fig. 3a\textsuperscript{13-17}. The canted magnetic moment orientations (green and orange) in the two ferroelectric domains (red and blue, respectively) of the BFO thin film are perpendicular to each other\textsuperscript{13}. Within a single BFO unit cell, the polarization (P) lies along a <111> direction. The canted moments (M\textsubscript{c}) and the antiferromagnetic axis (L) are perpendicular to each other in a (111) plane\textsuperscript{14,15}, see Fig. 3a and Supplementary Fig. 3, with L along <1-10> direction and M\textsubscript{c} along <11-2> direction. Thus, on this (001) surface of the BFO film, the total net IP polarization and the total net IP canted moment in BFO project parallel to each other along the (1-10) DSO direction, which is perpendicular to the stripes as shown in Figs 1c and 3a and Supplementary Fig. 3\textsuperscript{13}.

Under an OOP electric field, the polarization in each of the two BFO domains is rotated by 180° as illustrated on the right side of Supplementary Fig. 3 and confirmed experimentally by the FPM images shown in Fig. 3b. Both the IP (Fig. 3b, left, black arrows), and OOP (Fig. 3b, right), components of the polarization of BFO domains reverse from the as-grown (left side) to the switched state (right side). Therefore, the total polarization of BFO is rotated by 180° under an OOP E-field. These experimental results are confirmed by phase field modeling as shown in Supplementary Fig. 2, which clearly reveals the two-step switching of the polarization vector under an OOP applied electric field.

The measured angle dependence of the FMR field is shown in Fig. 3c. Within the picture presented in our model, M\textsubscript{c} and M\textsubscript{CoFe} are parallel on the blue curve so less FMR field is required to attain the resonance condition. On the red curve, M\textsubscript{c} and M\textsubscript{CoFe} are antiparallel so the FMR field must be larger. The FMR field at χ = 0° and 180° can be switched by a voltage pulse back and forth from the blue curve to the red curve, from 217 to 225 mT, that is, a tunable FMR field range of 8 mT. In contrast, the FMR fields at χ = 90° and 270° show nearly no change with voltage pulse. These results are consistent with the experimentally observed FMR field versus applied voltage loops at different χ angles shown in Fig. 2c. The difference between the FMR field measured at χ = 0° and χ = 90° of 5.1 ± 0.5 mT (the blue curve) and 2.9 ± 0.3 mT (the red curve) from Fig. 3c is the combination of the growth anisotropy field and the net IP magnetic anisotropy field induced by the exchange coupling\textsuperscript{13} between CoFe layer and BFO layer. The growth anisotropy field was found to be 1.1 ± 0.2 mT. The net IP exchange coupling field, H\textsubscript{ex}, was determined to be 4 ± 0.3 mT, which is scaled by 1/\cos 45° to give H\textsubscript{ex} = 5.7 ± 0.4 mT (see Supplementary Methods, modelling the FMR field and β angle dependence on applied voltage). This value is comparable to a previously reported value for the IP exchange coupling determined by a complementary method\textsuperscript{13}.

To further our understanding of the coupling, we measure FMR fields as we vary χ without switching the voltage, which complements our usual measurements taken by switching voltage pulses at fixed χ angle. In particular, after a voltage pulse switched from point 1 to point 4 in Fig. 3c, we maintain the +0 V polarization state and rotate the applied magnetic field direction from 0° to 180°. The FMR field follows the red curve from point 4 to point 3 at χ = 90°, but then follows the blue curve to point 2 at χ = 180° instead of to point 5. When the applied magnetic field is rotated back from 180° to 0°, the FMR field remains on the blue curve, changing from point 2 to point 3, at χ = 90°, and back to point 1, instead of to point 4. This measurement demonstrates that the +0 V points (red curve) correspond to a metastable state that is created by applying an electric field pulse, and that this metastable state can be switched to a more stable -0 V ground state (blue curve) by rotating the bias field through χ = 90° or χ = 270° where the states are degenerate. This instability explains why a voltage-induced exchange bias is not observed in hysteresis measurements of this system\textsuperscript{13}. 

Figure 2 | FMR measurements of CoFe/BFO multiferroic heterostructure. (a) Schematic of CoFe (2.5 nm)/BFO (300 nm) multiferroic heterostructure for FMR measurements at varying angles between the magnetic field and the easy axis; (b) FMR spectra of CoFe/BFO multiferroic heterostructure measured at open circuit after applying +5 V and -5 V at χ = 0°. Upper right inset is the relative FMR field dependence on applied voltage; (c) relative FMR field dependence after applying voltage pulses along different orientations of the magnetic field; (d) relative FMR field versus applied voltage pulses after 50 and 100 cycles of a 10-V sinusoidal voltage. Error bars in b-d are defined as the s.d. of five FMR measurements for each FMR data.
Finally, to reaffirm the coupling between BFO and CoFe, it is critical to carry out a 'null' experiment. We deposited Cu (3 nm)/CoFe (1.2 nm, 2.5 nm) bilayers in a 20-mT in situ magnetic field on a 50-nm SrTiO$_3$ (STO) dielectric thin film. We measured the FMR signal on the CoFe/STO heterostructure at different voltages across STO following the same procedure as for the CoFe/BFO heterostructures. No FMR field shift was observed in CoFe (2.5 nm)/STO (50 nm) heterostructure, see Supplementary Fig. 4. The calculations and discussion are listed in the Supplementary Methods, experimental and theoretical study of FMR measurements in a CoFe/STO multiferroic heterostructure (Supplementary Fig. 4).

Discussion
With this compelling experimental data as the framework, we present a model that allows us to understand what happens to the canted moments in each of the BFO domains' exchange coupled to the CoFe. Unfortunately, little is known about the magnetic structure and energetics of the CoFe/BFO interface so several coupling models may be possible. However, a model must be consistent with our experimental observations: namely, the uniaxial anisotropy, the hysteretic switching of FMR fields, the voltage-induced metastable state and the degenerate nature of the structures at φ = 90° and φ = 270°. A model, which describes
the data well, is based on the assertion that the IP $M_z$ is either parallel or antiparallel to the IP $P$, and that $P$ reverses with voltage but does not change when the magnetic field is rotated. When $P$ is rotated $180^\circ$ by an OOP E-field, the voltage-induced system energy change overwhelms the CoFe/BFO cantilever moment interface exchange coupling energy, creating an energy barrier that allows the cantilever moment in BFO to be in a metastable state antiparallel to the magnetization direction of CoFe. Although the coupling between $M_{CoFe}$ and $M_z$ is unidirectional, it becomes unstable at $\varphi = 90^\circ$ and thereby approximates a uniaxial anisotropy (see Supplementary Methods, modelling the FMR field and $\beta$ angle dependence on applied voltage).

To calculate the BFO-cantilever magnetic moment rotation angle quantitatively from the FMR measurements and to simulate the voltage dependence of the angular variation of the FMR field, we used the coordinate system in Fig. 3a. As discussed in the Supplementary Methods section, the voltage-dependent FMR field of CoFe at different $\varphi$ angles can be quantitatively simulated as shown by the dashed line in Fig. 3c. Also, by substituting the experimentally determined angle-dependent FMR field shift into the energy functional, we can deduce the dependence of $\beta (V)$ on applied voltage pulse. The hysteretic $\beta (V)$ versus applied voltage curve (blue) obtained from the model closely matches the P-E polarization hysteresis loop (red) as seen in Fig. 3d. Suppose the two states $0^\circ$ and $180^\circ$ refer to two different remnant polarization states at $V$ where the $0^\circ$ is initial state. We can solve for the angle $\beta$ at both $0^\circ$ and $-180^\circ$ as

$$\beta(0^\circ) = 45^\circ \pm 8^\circ, \quad \beta(-180^\circ) = 129^\circ \pm 8^\circ, \quad \Delta \beta = 178^\circ \pm 11^\circ$$

The analytical results indicate that $\beta (V)$ in the single domain (red) was switched from $45^\circ$ to $-135^\circ$ with changing applied voltage pulses, which confirms the $180^\circ$ reversal of the cantilever magnetic moment by the E-field in agreement with the schematic of Supplementary fig. 3.

In conclusion, we present angle-dependent FMR field shifts demonstrating a voltage pulse induced, robust, repeatable switching of the magnetic coupling in a CoFe/BFO thin-film heterostructure. The cantilever moment in BFO does reverse direction with the application of a bipolar electric field. This work demonstrates the significant potential for exchange coupled magnetic film/BFO heterostuctures in spintronics devices for compact, fast energy-efficient voltage-writable RF/microwave and spintronics/RFID devices.

**References**


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Author contributions

Z.Z. conducted and analyzed the data with help from T.N. and X.Y. R.R. and N.X.S. planned and supervised the study. M.T. and Ya Gu fabricated the films. M.T. measured the PFM images. S.R.B., D.T.P., and J.J. measured the SEMPA images. Yuan Guo performed the ion milling. M.J., B.M.H., and G.J.R. prepared the STO films. Z.Z. wrote the manuscript with modification from J.J., D.T.P., M.D.S., R.R. and N.X.S. Theoretical simulation of the method part and Supplementary figures 1 and 2 were done by D.Q., K.A. and S.S. All authors contributed to discussion of the results.
Supplementary Figures

(a) \hspace{1cm} (b)

Supplementary Figure 1 | Micromagnetic simulation. (a) Initial configuration of the magnetic moments (b) Configuration of the magnetic moments after a vertical electric field was applied to the ferroelectric.

Supplementary Figure 2 | Time evolution of the ferroelectric domains simulated using a three dimensional phase field model after a vertical electric field has been applied to the box region.
Supplementary Figure 3 | Schematic of the CoFe layer and angles defining $M_c$ and the BFO layer showing the in-plane components of $M_c$ (green and orange arrows) and $P$ in the two (red and blue) ferroelectric stripe domains as well as the net $M_c$ perpendicular to the stripes. The 3-d orientation of $P$, $M_c$, and the antiferromagnetic axis $L$ are shown below. The same is shown on the right after switching by application of an out-of-plane E field.
Supplementary Figure 4 | Ferromagnetic resonance measurements of CoFe/STO heterostructure under varied E-field. (a) Schematic of the CoFe (1.2 or 2.5 nm)/STO (50 nm) sample structure for the FMR field measurements; (b) Measured and calculated (dashed) angular dependence of FMR fields for the CoFe (1.2 nm)/STO (50 nm) multiferroic heterostructure measured after different voltage pulses; (c) Measured and calculated (dashed) angular dependence of FMR fields for the CoFe (2.5 nm)/STO (50 nm) multiferroic heterostructure; (d) FMR field shift dependence on applied voltage for different CoFe film thickness. Error bars (Supplementary Figure 4 (b), (c) and (d)) are defined as standard deviation of 5 FMR measurements of each FMR data.

Supplementary Methods:

Modeling the FMR field and $\beta$ angle dependence on applied voltage. Here we show the details of the analytical calculation of the voltage induced CoFe magnetization switching and the BFO canted moment rotation angle from the FMR measurements for the CoFe/BFO heterostructures following the coordinate system in Supplementary Fig. 3. The magnetization of CoFe forms an angle $\theta$ with the Z axis
and its in-plane (X-Y plane) projection forms an angle $\phi$ with the X axis (CoFe magnetic easy axis). The bias magnetic field $H$ forms an angle $\alpha$ with the easy axis. The canted magnetic moment in one BFO domain (Red) forms an angle $\beta$ with the magnetic easy axis, as shown in Supplementary Fig 3; $\beta$ is voltage dependent. Considering the total energy in the CoFe portion above one BFO ferroelectric domain (Red), we have:

$$E_{\text{tot}} = E_{\text{Zeeman}} + E_{\text{M}_s} + E_{\text{surf}} + E_{\text{EX}} + E_{\text{anisotropy}},$$

where $E_{\text{Zeeman}}$ is the Zeeman energy, $E_{\text{M}_s}$ is the demagnetizing energy, $E_{\text{surf}}$ is the surface anisotropy energy, $E_{\text{EX}}$ is the interfacial exchange coupling energy between the antiferromagnetic BFO and the ferromagnetic CoFe, and $E_{\text{anisotropy}}$ is the growth field induced anisotropy along the X axis. The energy $E_{\text{EX}}$ can be represented as:

$$E_{\text{EX}} = J_{\text{EX}} \vec{M}_{\text{Fe}} \cdot \vec{M}_{\text{CoFe}},$$

with $J_{\text{EX}}$ being an effective magnetic field originating from the exchange coupling of the BFO canted moment, $M_{\text{CoFe}}$, and the ferromagnetic moment in CoFe, $M_{\text{CoFe}}$. The total energy in the part of the CoFe film exchange coupled to the BFO domain (Red) can therefore be expressed as:

$$E = -\mu_s M_s \sin \theta (\cos \phi \cos \sigma + \sin \phi \sin \sigma) + \frac{1}{2} \mu_s M_s^2 \cos^2 \theta + K_u \sin^2 \theta \sin^2 \phi + \frac{K_{\text{CoFePt}} + K_{\text{CoFe/BFO}}}{d} \sin^2 \theta - \mu_s H_{\text{EX}} M \sin \theta (\cos \phi \cos \beta(V') + \sin \phi \sin \beta(V'))$$

in which $d$ is the CoFe layer thickness, $K_u$, $K_{\text{CoFePt}}$ and $K_{\text{CoFe/BFO}}$ are the induced anisotropy of CoFe thin film and the magnetic surface anisotropy at the two interfaces of CoFe/Pt, and CoFe/BFO layer, respectively. For another BFO domain (Blue), the direction of canted magnetic moment is $90^\circ$ away from that of the Red domain, and we have the interfacial exchange coupling energy term as:

$$-\mu_s H_{\text{EX}} M \sin \theta (\cos \phi \cos (\beta(V') - \frac{\pi}{2}) + \sin \phi \sin (\beta(V') - \frac{\pi}{2}))$$ \n
$$-\mu_s H_{\text{EX}} M \sin \theta (\cos \phi \sin \beta(V') - \sin \phi \cos \beta(V')).$$

Suppose the two kinds (Red and Blue) of BFO domains have equal surface areas, which are exchange coupled to CoFe full film, we then have the total energy for the complete CoFe film as:
\[ E = -\mu_0 MH \sin \theta \cos (\phi - \alpha) - \left( \frac{1}{2} \mu_0 M^2 - \frac{K_{\text{ex}}}{d} \cos^2 \theta + \frac{K_{\text{sub}}}{d} \sin^2 \phi \right) \sin \theta \sin \beta \]

\[ + K_{ij} \sin^2 \theta \sin^2 \phi - \mu_0 M \frac{\sqrt{F}}{2} H_{ex} \sin \theta \cos (\phi - (\beta - 45^\circ)) \]

(2)

The last term is the exchange coupling energy of the two-domain state of the CoFe/BFO heterostructure as shown in Fig. 1, for example the orange and yellow domains in the upper right or the blue and cyan domains in the lower left of Fig. 1c. We distinguish between the net in-plane \( P \) and \( M_C \) perpendicular to the stripes and the in-plane \( P \) and \( M_C \) at \( \pm45^\circ \) in each stripe domain as seen in Supplementary Fig. 3. The net CoFe magnetization in Fig. 1c has the same energy whether it is parallel (orange/yellow region) or antiparallel (blue/cyan region) to the net polarization.

To model the data of Fig. 3c, we express the exchange coupling energy term as a piecewise function which minimizes the exchange energy term. For the blue data points, where the in plane \( M_{\text{CoFe}} \) is in approximately the same direction as \( M_C \), we have:

\[ E_{\text{ex}} = -\frac{\sqrt{F}}{2} \mu_0 H_{ex} M_{\text{CoFe}} \sin \theta \cos (\phi - (\beta - 45^\circ)) \quad (90^\circ < \alpha < 90^\circ, \alpha \approx \alpha, \beta \approx 45^\circ) \]

\[ E_{\text{ex}} = \frac{\sqrt{F}}{2} \mu_0 H_{ex} M_{\text{CoFe}} \sin \theta \cos (\phi - (\beta - 45^\circ)) \quad (90^\circ < \alpha < 270^\circ, \alpha \approx \alpha, \beta \approx 225^\circ) \]

This establishes the in-plane uniaxial anisotropy behavior of the CoFe film as seen in Fig. 3c. The sign of the exchange coupling energy term is reversed on the red curve where the direction of \( M_C \) is reversed.

For \(-90^\circ < \alpha < 90^\circ\) case, we can derive the total energy equation as:

\[ E = \left( \frac{1}{2} \mu_0 M^2 - \frac{K_{\text{ex}}}{d} \cos^2 \theta + \frac{K_{\text{sub}}}{d} \sin^2 \phi \right) \cos \theta \sin \beta \]

\[ -\mu_0 M \sin \theta (\cos \phi - \frac{1}{2} H_{ex} (\cos \beta \sin \phi + H \cos \alpha) - \sin \phi (\frac{1}{2} H_{ex} (\sin \beta \cos \phi + H \sin \alpha)) \]

We put the energy equation (3) into the FMR equation (4):

\[ \left( \frac{2\pi \beta}{\gamma} \right)^2 = \left( E_{\theta} E_{\phi} - E_{\theta \phi} \right)^2 - \frac{1}{\mu_0 M^2 \sin^2 \theta_0} \]

(4)

where:
\[ \frac{\delta E}{\delta \varphi} \bigg|_{\varphi_0} = 0; \quad \frac{\delta^2 E}{\delta \varphi^2} \bigg|_{\varphi_0} = 0; \]
\[ E_{\text{mag}} = \frac{\delta^2 E}{\delta \varphi^2} \bigg|_{\varphi_0, \omega_0, \theta_0} = E_{\text{mag}} = \frac{\delta^2 E}{\delta \varphi^2} \bigg|_{\varphi_0, \omega_0, \theta_0} \]
\[ \tan \varphi_0 = \frac{1}{2} \frac{H_{\text{ex}} (\sin \beta (V) - \cos \beta (V)) + H \sin \alpha + H \sin \alpha}{H_{\text{ex}} (\cos \beta (V) + \sin \beta (V)) + H \cos \alpha + H \cos \alpha} \]

From the energy equation above, since \( H = 220 \text{ mT} \), \( H_{\text{ex}} = -5.7 \text{ mT} \), the angle \( \phi \) is determined by \( \alpha \), we have:
\[ \tan \phi_0 = \tan \alpha, \phi_0 = \alpha \]

Solving the FMR Eq (4) at fixed frequency and at varying \( \alpha \), we have:
\[ f(\alpha, V) = \frac{1}{2 \pi} \sqrt{\frac{H(\alpha, V) + \frac{1}{2} H_{\text{ex}} (\cos \beta (V) + \sin \beta (V)) \cos \alpha + \sin \beta (V) \sin \alpha + H \cos \alpha}{H(\alpha, V) + \frac{1}{2} H_{\text{ex}} (\cos \beta (V) + \sin \beta (V)) \cos \alpha + \sin \beta (V) \sin \alpha + H \cos \alpha + M^*}} \]
\[ M^* = M \cdot \frac{2(K_{1, \text{CoFeB}} + K_{3, \text{CoFeB}})}{\mu \omega dM} \] (5)

At fixed \( f(\alpha, V) \), the term:
\[ H(\alpha, V) + \frac{1}{2} H_{\text{ex}} (\cos \beta (V) + \sin \beta (V)) \cos \alpha - \sin \beta (V) \sin \alpha + H \cos \alpha \] (6)
is constant. The net anisotropy field, \( H_{\text{net}}(\phi=90^\circ) - H(\phi=0^\circ) \) results from both the net effective exchange coupling perpendicular to the ferroelectric domain stripes \( H_{\text{Exeff}} \) and the growth induced anisotropy field \( H_\alpha \). As shown in Supplementary Fig. S3, \( H_{\text{Exeff}} = H_{\text{Ex}} \cos \beta_0 \) where \( \beta_0=45^\circ \) is the initial \( \beta \) angle in red domain. We have \( H_{\text{eff}} = H_{\text{Ex}} \cos \beta_0 + H_\alpha = 5.1 \text{ mT} \pm 0.4 \text{ mT} \) for the blue curve and \( H_{\text{eff}} = H_{\text{Ex}} \cos \beta_0 + H_\alpha = 2.9 \text{ mT} \pm 0.3 \text{ mT} \) for the red curve. The \( H_{\text{Ex}} \) and \( H_\alpha \) can be solved as \( H_{\text{Ex}} = 5.7 \text{ mT} \pm 0.4 \text{ mT} \) and \( H_\alpha = 1.1 \text{ mT} \pm 0.2 \text{ mT} \), respectively. The uncertainty of FMR field is determined by the multi-measurements of FMR field spectra after same voltage impulse. By putting \( H_{\text{Ex}} = 5.7 \text{ mT} \), \( H_\alpha = 1.1 \text{ mT} \) and \( \beta (V) = 45^\circ \) (Blue dashed line) or \( 225^\circ \) (Red dashed line) into Eq. (6), we obtain the dashed line fit in Fig. 3c. Using Eq. (6) and the experimental data at different \( \alpha \) we have:
\[ \Delta H(\alpha = 0) = \frac{\sqrt{3}}{2} H_{\text{ex}} \cos(\beta - 45^\circ) \] \[ \Delta H(\alpha = 45) = \frac{\sqrt{3}}{2} H_{\text{ex}} \sin \beta \] \[ \Delta H(\alpha = 90) = \frac{\sqrt{3}}{2} H_{\text{ex}} \sin(\beta - 45^\circ) \] \[ \Delta H(\alpha = -45) = \frac{\sqrt{3}}{2} H_{\text{ex}} \cos \beta \] 

The notation \( \alpha \) and \( \beta \) refers to two different polarization states at 0 V after plus or minus applied voltage pulses in the hysteresis loops of Fig. 3c, where 0- is the initial state. We obtain:

\[ \beta(0-) = 49^\circ \pm 8^\circ, \beta(0+) = -129^\circ \pm 8^\circ, \Delta \beta = 178^\circ \pm 11^\circ \]

We can easily solve for the angle \( \beta \) from Eq. (7) at other voltages, from data like Fig. 3c at other voltages, to obtain Fig. 3d.

**Experimental and theoretical study of FMR measurements in a CoFe/STO multiferroic heterostructure.** Cu (3 nm)/CoFe (1.2 nm, 2.5 nm)/STO (50 nm)/Pt (10 nm) magnetic/dielectric heterostructures were prepared by sputtering for the purpose of comparison with the CoFe/BFO heterostructures. A ~20 mT in-situ bias magnetic field was applied during CoFe deposition, establishing a uniaxial anisotropy in the CoFe thin film. FMR measurements on the CoFe/STO heterostructures at different voltages were carried out following the same procedure as for the CoFe/BFO heterostructures, as shown in Supplementary Fig. 4. The angular dependence of the FMR fields measured after different voltage pulses applied across the STO layer with different CoFe film thicknesses are shown in Supplementary Fig. 4b and c. An in-plane, isotropic voltage induced FMR field shift behavior is observed for CoFe (1.2 nm)/STO (50 nm) at all applied voltages, which is in sharp contrast to the in-plane anisotropic voltage induced FMR field shift in CoFe/BFO (Fig. 3c). This in-plane isotropic voltage induced FMR field shift observed in CoFe/STO thin film heterostructure is due to the spin polarized charge mediated magnetoelectric coupling, which leads to an E-field induced out-of-plane magnetic anisotropy change23-24. The
FMR field shifts of the CoFe (1.2 nm)/STO (50 nm) heterostructure are linearly dependent on applied voltages\textsuperscript{25}, as shown in Supplementary Fig. 4d. In the CoFe (2.5 nm)/STO (50 nm) heterostructure, no FMR field shift was obtained in our experiment, see Supplementary Fig. 4c and d. The result is consistent with charge-screening effects being negligibly small\textsuperscript{24} in the higher thickness (~0.5 nm) conducting CoFe magnetic thin film due to the fact that the Thomas-Fermi/Debye screening length is in the range of 0.05 nm to 0.08 nm for typical metals\textsuperscript{40}.

The coordinate (Supplementary Fig. 4a) used for the CoFe/STO thin film heterostructure system to calculate the voltage induced FMR field of the CoFe layer was similar to Supplementary Fig. 3. The total energy of the CoFe layer in the CoFe/STO heterostructure includes several terms:

$$E_{\text{total}} = E_{\text{Zeeman}} + E_{\text{M}} + E_{\text{surf}} + E_{\text{anisotropy}} + E_{\text{Charge}}$$  \hspace{1cm} (8)

Where $E_{\text{Zeeman}}$ is the Zeeman energy, $E_{\text{M}}$ is demagnetizing energy, $E_{\text{surf}}$ is the surface anisotropy energy, $E_{\text{anisotropy}}$ is the magnetic anisotropy induced by the growth magnetic field along the X direction during deposition, and $E_{\text{Charge}}$ is the spin-polarized surface charge mediated anisotropy. The total energy $E_{\text{total}}$ can be written as:

$$E = -\mu_0 M H \sin \theta (\cos \varphi \cos \alpha + \sin \varphi \sin \alpha) + \frac{1}{2} \mu_0 M^2 + \frac{E_{\text{CoFe/STO}} + E_{\text{CoFe/Cu}} + \Delta E_s(V)}{d} \cos^2 \theta$$  \hspace{1cm} (9)

$$-K_u \sin^2 \varphi + \frac{E_{\text{anisotropy}} + E_{\text{magnetic}} + \Delta E_s(V)}{d}$$

in which $d$ is the CoFe layer thickness and $K_{\text{CoFe/STO}}$ and $K_{\text{CoFe/Cu}}$ are the magnetic surface anisotropy at the two interfaces of CoFe/STO, and CoFe/Cu, respectively. $K_u$ is the growth induced anisotropy, and $\Delta E_s(V)$ is surface charge induced anisotropy change. We can put the total energy equation (9) into the FMR equation (10) below:

$$\left(\frac{2\pi f}{\gamma}\right)^2 = \left(E_{\text{anisotropy}} - E_{\text{surf}}\right)^2 \frac{1}{\mu_0 M^2 \sin^2 \theta_0}$$  \hspace{1cm} (10)

where:
\[ \frac{\partial E}{\partial \theta} \bigg|_{\theta=\theta_0} - \partial E \bigg|_{\theta=\theta_0} = 0; \]

\[ E_{\theta\theta} = \frac{\partial^2 E}{\partial \theta^2} \bigg|_{\theta=\theta_0}, \quad E_{\phi\phi} = \frac{\partial^2 E}{\partial \phi^2} \bigg|_{\phi=\phi_0}, \quad E_{\theta\phi} = \frac{\partial^2 E}{\partial \theta \partial \phi} \bigg|_{\theta=\theta_0, \phi=\phi_0}. \]

We solve the FMR equation at fixed frequency at varying \( \alpha \) and obtain:

\[ f(\alpha, V) = \frac{\gamma}{2 \pi} \sqrt{\left( H + \frac{2K_{\perp}}{\mu_0 M} \cos^2 \alpha \right) \left( H + \frac{2K_{\perp}}{\mu_0 M} \cos^2 \alpha + M' \right)} \]  \hspace{1cm} (11)

\[ M' = M - 2 \frac{(K_{v,CoFe/STO} + K_{c,CoFe/STO} + \Delta K_{v}(V))}{\mu_0 M}. \]

The anisotropy field \( H = 2K_{\perp}M \) is determined to be 4.3 mT±0.3 mT from the difference of FMR field between \( \alpha=0^\circ \) and \( \alpha=90^\circ \) at the same voltage, for example, on blue dashed line in Supplementary Fig. 4 after applied a -5V pulse. The effective surface charge induced anisotropy field, gives the FMR field shift and can be represented at fixed \( f(\alpha, V) \) as:

\[ \Delta H = \frac{K_{v}(V)}{\mu_0 M}. \]  \hspace{1cm} (12)

It is independent of the applied magnetic field orientation, \( \alpha \). The theoretical model is shown by the dashed curve in Supplementary Fig. 4a fits our CoFe/STO experimental data well.