Electric-Field-Enhanced Bulk Perpendicular Magnetic Anisotropy in GdFe/Pb(Mg_{1/3}Nb_{2/3})_{0.7}Ti_{0.3}O₃ Multiferroic Heterostructure

Aitian Chen, Senfu Zhang, Yan Wen, Haoliang Huang, Jürgen Kosel, Yalin Lu, and Xixiang Zhang

ACS Appl. Mater. Interfaces, **Just Accepted Manuscript** • DOI: 10.1021/acsami.9b16904 • Publication Date (Web): 18 Nov 2019

Downloaded from pubs.acs.org on November 27, 2019

Just Accepted

"Just Accepted" manuscripts have been peer-reviewed and accepted for publication. They are posted online prior to technical editing, formatting for publication and author proofing. The American Chemical Society provides "Just Accepted" as a service to the research community to expedite the dissemination of scientific material as soon as possible after acceptance. "Just Accepted" manuscripts appear in full in PDF format accompanied by an HTML abstract. "Just Accepted" manuscripts have been fully peer reviewed, but should not be considered the official version of record. They are citable by the Digital Object Identifier (DOI®). "Just Accepted" is an optional service offered to authors. Therefore, the "Just Accepted" Web site may not include all articles that will be published in the journal. After a manuscript is technically edited and formatted, it will be removed from the "Just Accepted" Web site and published as an ASAP article. Note that technical editing may introduce minor changes to the manuscript text and/or graphics which could affect content, and all legal disclaimers and ethical guidelines that apply to the journal pertain. ACS cannot be held responsible for errors or consequences arising from the use of information contained in these "Just Accepted" manuscripts.

**Electric-Field-Enhanced Bulk Perpendicular Magnetic Anisotropy in
GdFe/Pb(Mg_{1/3}Nb_{2/3})_{0.7}Ti_{0.3}O₃ Multiferroic Heterostructure**

*Aitian Chen,[†] Senfu Zhang,[†] Yan Wen,[†] Haoliang Huang,[‡] Jürgen Kosel,[§] Yalin Lu,[‡] and Xi-
Xiang Zhang^{*,†}*

[†]Physical Science and Engineering Division, King Abdullah University of Science and Technology, Thuwal 23955-6900, Saudi Arabia

[‡]Anhui Laboratory of Advanced Photon Science and Technology, National Synchrotron Radiation Laboratory, University of Science and Technology of China, Hefei 230026, China

[§]Computer, Electrical and Mathematical Sciences and Engineering Division, King Abdullah University of Science and Technology, Thuwal 23955-6900, Saudi Arabia

KEYWORDS: bulk perpendicular magnetic anisotropy, electric-field control of magnetism, multiferroic heterostructure, ferrimagnetic, ferroelectric, rare earth-transition metal alloys

ABSTRACT: Perpendicular magnetic anisotropy is important for increasing the information storage density in the perpendicular magnetic recording media, and for rare earth-transition metal alloys with bulk perpendicular magnetic anisotropy that generate great research interest due to their abundant interesting phenomena, such as fast domain wall motion and skyrmion. Here, we deposit amorphous GdFe ferrimagnetic films on Pb(Mg_{1/3}Nb_{2/3})_{0.7}Ti_{0.3}O₃ ferroelectric substrate and investigate the effect of electric-field-induced piezostress on its bulk perpendicular magnetic anisotropy. The anomalous Hall effect and polar Kerr image measurements suggest an enhanced bulk perpendicular magnetic anisotropy by electric field, which originates from a positive magnetoelastic anisotropy due to the positive magnetostriction coefficient of the GdFe film and the electric-field-induced tensile strain along the *z* axis in Pb(Mg_{1/3}Nb_{2/3})_{0.7}Ti_{0.3}O₃ ferroelectric substrate. Our results enrich the electrical control of perpendicular magnetic anisotropy and are useful for designing spintronic devices based on perpendicular magnetic anisotropy.

INTRODUCTION

In terms of the memory density, scaling behavior and thermal stability, perpendicularly magnetized magnets are superior to their in-plane magnetized counterparts for applications in scaled technology.^{1,2} As one of the key material parameters in modern spintronic devices, perpendicular magnetic anisotropy (PMA) has been extensively investigated. Perpendicular magnetic anisotropy can be roughly classified into two types: interface PMA and bulk PMA. Interface PMA is primarily based on heavy-metal/ferromagnet and heavy-metal/ferromagnet/oxide structures, such as Co/Pt and Ta/CoFeB/MgO, in which spin-orbit coupling plays an important role.³ Bulk PMA is often observed in Heusler alloys,⁴ $L1_0$ magnetic alloys (such as FePt, FePd, CoPt)^{5,6} and rare earth-transition metal (RE-TM) alloys.⁷⁻¹⁰

Electrical control of PMA in magnetic materials is drawing much attraction due to its potential to lower energy consumption. Much efforts have been focused on controlling PMA via current-induced spin-orbit torque,¹¹⁻¹⁶ voltage-induced ion migration,¹⁷⁻¹⁹ charge carrier doping²⁰ and so on. Alternatively, multiferroic heterostructures, hybrid ferromagnetic and ferroelectric, enable electric-field control of magnetism via strain-mediated magnetoelectric coupling,²¹⁻²³ though most studies have focused on how to control ferromagnetic films with in-plane magnetization.²¹⁻²⁵ Recently, electric-field control of PMA using multiferroic heterostructures has been reported.²⁶⁻³⁸ For example, Sun et al.³⁴ and Peng et al.³⁵ demonstrated electric-field control of spin reorientation transition (SRT) and interface PMA in (Co/Pt)₃/PMN-PT (Pb(Mg_{1/3}Nb_{2/3})_{0.7}Ti_{0.3}O₃) multiferroic heterostructure. In these studies, the observed remarkable change of interface PMA by electric field is limited to the sample around the SRT and depends strongly on the thickness of the ferromagnetic layer. Many of these works utilize electric-field-induced piezostress to modulate interface PMA and focus primarily on ferromagnetic

materials,³¹⁻³⁸ while the effect of strain on ferrimagnetic materials with bulk PMA has not been studied. Additionally, in these works, the electric field typically weakens the PMA^{26,27,31-38} (i.e., rotates the magnetic easy axis from the out-of-plane to the in-plane), while electrically enhanced PMA remains elusive.

Rare earth-transition metal alloys with bulk PMA have been widely used in magneto-optical recording.^{10,39} Since the spins of two inequivalent RE and TM sublattices are antiferromagnetically coupled, ferrimagnetic RE-TM alloys provide a framework for investigating antiferromagnetic spin dynamics, such as fast domain wall motion⁴⁰ and vanishing skyrmion Hall effect.^{7,41} Moreover, the RE-TM alloys have attracted great interest due to the recent observation of magnetic skyrmion,^{7,8,42} which is promising for high-speed, high-density spintronic devices. Up to now, the investigation of electric-field control of RE-TM alloys and ferrimagnetic materials with bulk PMA is still lacking, but it will pave a new way to manipulate these interesting phenomena in RE-TM alloys.

Here, we study the effect of electric fields on the bulk PMA of the amorphous GdFe ferrimagnetic films on PMN-PT ferroelectric substrate. Through the anomalous Hall effect (AHE) and polar Kerr image measurements with in situ electric fields, we observed electric-field-enhanced bulk PMA of the GdFe films. The results of the in situ X-ray diffractometer (XRD) indicate that electric fields extend the crystal lattice of the PMN-PT substrate, resulting in a tensile strain along the z direction. This tensile strain induces a positive magnetoelastic anisotropy along the z direction, which contributes to the enhancement of bulk PMA in the GdFe/PMN-PT multiferroic heterostructure. These results enrich the electric-field control of magnetism and PMA.

RESULTS AND DISCUSSION

The 20 nm amorphous RE-TM alloy GdFe (Gd₂₅Fe₇₅) films were deposited on one-side-polished PMN-PT (011) single crystals; afterward, a 5 nm Pt layer was deposited to protect the GdFe film from oxidation. The GdFe/PMN-PT multiferroic heterostructure was selected as our sample because the bulk PMA of the ferrimagnetic GdFe film is closely related to the magnetoelastic contribution,^{43,44} and the PMN-PT ferroelectric single crystal has been widely used as the piezoelectric substrate in multiferroic heterostructures^{21,31,34,35,37} to produce distinct, stable and electrically-reversible lattice strain due to its large piezoelectric coefficients. A shadow mask was used during the film deposition to pattern the samples into a Hall bar shape for magnetotransport measurements, as illustrated in Figure 1. The electric field was applied on the PMN-PT substrate to generate strain through the converse piezoelectric effect to modify the magnetic property of the GdFe film. In all measurements, a positive electric field was defined as pointing from PMN-PT to GdFe.

It is well-documented that the AHE is closely related to the out-of-plane magnetization, i. e., $R_{\text{Hall}} \propto M_z$;³² thus, we measured the AHE under in situ electric fields to study the effect of the electric field on the PMA. Figure 2a displays the normalized Hall resistance as a function of the out-of-plane magnetic field ($H_{\perp}$), which can also manifest the out-of-plane magnetic hysteresis (M-H) loop. It is evident that the squareness of the AHE loops was enhanced as a result of increasing the applied electric field, though the coercive field had a slight increase from approximately 44 Oe at $E = 0$ kV cm⁻¹ to approximately 47 Oe at $E = 20$ kV cm⁻¹. The normalized R_{Hall} values at zero magnetic field, which is proportional to the remnant magnetization, for different electric fields were extracted from the AHE curves in Figure 2a and plotted as a function of electric field, as illustrated in Figure 2b. The normalized R_{Hall} at $H_{\perp} = 0$ Oe increased almost linearly as the electric field increased. The normalized R_{Hall} at $H_{\perp} = 0$ Oe is

approximately 0.495 at $E = 0 \text{ kV cm}^{-1}$ and 0.585 at $E = 20 \text{ kV cm}^{-1}$, corresponding to an enhancement of about 18%. The enhancements of the squareness of the AHE loops and the normalized R_{Hall} at $H_{\perp} = 0 \text{ Oe}$ induced by the applied electric fields indicate electric-field-enhanced the squareness of the M-H loops and the out-of-plane remnant magnetization, which are features of PMA. Thus these results suggest an enhancement of the bulk PMA induced by the applied electric fields. It is notable that this electrically enhanced PMA is sharply distinguished from the previous electric-field-weakened PMA observed in multiferroic heterostructures of ferroelectric substrate with Co/Pt multilayers,^{34,35,37,38} CoPd alloy,^{26,27} Cu/Ni multilayers,³⁶ Co/Ni multilayers³³ and so on. Moreover, the magnetic materials in the literature are ferromagnetic, while the GdFe film in the current study is ferrimagnetic. The mechanism of this enhanced PMA by the electric field is discussed later.

To gain further insight into this electric-field-enhanced bulk PMA, we performed polar Kerr image measurements with in situ electric fields. For each electric field, a positive magnetic field of 300 Oe was first applied to saturate the GdFe film. Afterward, a series of negative magnetic field pulses having a width of 0.1 s (Figure 3a) were applied to reverse the magnetization, and the evolution of the domain structures was recorded at zero magnetic field after each pulse using a quasi-static technique (Figure S1). The domain structure experienced a remarkable change around the coercive field due to the magnetic domain reversal. Figure 3b presents the representative Kerr images recorded after applying the magnetic field pulses of -50 Oe and -60 Oe , which were around the coercive field, under different electric fields. The magnetic domains with black contrast were upward, since they had the same contrast as did the domains after applying a positive magnetic field of 300 Oe, and the magnetic domains with white contrast were downward and were generated and expanded by the negative magnetic field pulses. For each

electric field, the images after applying -50 Oe magnetic field pulse had primarily black contrast, while the images after applying -60 Oe magnetic field pulse had primarily white contrast. This occurred because the coercive field of the film is approximately 50 Oe (extracted from Fig. 2a) and is smaller than 60 Oe. The change of contrast indicates the magnetic domain reversal around the coercive field. In addition, it can be found that the electric fields had a significant effect on the domain structures, as displayed in Figure 3b. After applying the same magnetic pulse, the areas with white contrast under $E = 0$ kV cm $^{-1}$ were larger than those under $E = 20$ kV cm $^{-1}$, which is clearly observable in the areas encircled by the yellow dashed lines. After applying a magnetic field pulse of -60 Oe, for example, the entire encircled area for $E = 0$ kV cm $^{-1}$ nearly had downward magnetic domain with white contrast. However, the same area still presented black contrast when applying electric fields, especially under $E = 20$ kV cm $^{-1}$. The decrement of white contrast at large electric fields suggests hardness of the magnetic domain reversal. Furthermore, the magnetic domain reversal ratio for each magnetic field pulse and electric field was quantified by statistically analyzing the ratio of the areas with white contrast in the Kerr images (Figure S1, Supporting Information); the results are presented in Figure 3c. The magnetic domain for various electric fields mostly reversed around -60 Oe, since the electric field had little effect on the coercive field, as deduced from Figure 2a, and the coercive field (about 50 Oe) is slightly smaller than the applied field (-60 Oe). For the pulse magnetic field in the range of -30 to -70 Oe, the reversal ratio decreased when increasing the electric field, as guided by the arrow. This electric-field-induced decrease in the ratio of the reversed magnetic domains suggests the decrease of the reversed magnetic domain and the hardness to reverse magnetic domain, which indicates an electric-field-enhanced bulk PMA in the GdFe film and is consistent with the aforementioned AHE measurements.

Both the AHE and polar Kerr image measurements suggest the electric-field-enhanced bulk PMA of the GdFe film. Next, we explore the mechanism and physics underlying these observations. In multiferroic heterostructures, both strain- and charge-mediated magnetoelectric couplings are expected. In this work, we only applied positive electric fields and the interface charge should not change. So the charge effect, which affects only several nanometers (Figure S2, Supporting Information), can be eliminated for the 20 nm thickness of the GdFe film.⁴⁵ Thus, the aforementioned electric-field-enhanced bulk PMA of the GdFe film should originate from the strain-mediated magnetoelectric coupling. The strain-induced magnetoelastic anisotropy can be written simply as $K_{\text{strain}} = \frac{3}{2} \varepsilon_z Y \lambda / (1 - \nu)$,^{28,31} where ε_z is the strain along the z axis, Y , λ and ν represent the Young's modulus, magnetostriction coefficient and Poisson ratio of the magnetic layer, respectively. The ε_z is closely related to the out-of-plane lattice parameter of the PMN-PT substrate, which can be obtained from XRD. Figure 4a presents the XRD results with in situ electric fields. By increasing the electric field, both (011) and (022) diffraction peaks of the PMN-PT substrate shifted to smaller angles, indicating the expansion of the out-of-plane lattice. Based on the shift of the (022) peak in Figure 4a, the ε_z induced by the electric fields can be estimated. As illustrated in Figure 4b, the induced tensile strain along the z axis depends almost linearly on the electric field, and the largest strain at $E = 20 \text{ kV cm}^{-1}$ is approximately 0.14%. For the GdFe films, the magnetostriction coefficient is positive (approximately 7×10^{-6}),⁹ so the strain-induced magnetoelastic anisotropy under $E = 20 \text{ kV cm}^{-1}$ is approximately 1680 J m^{-3} , considering $Y = 8 \times 10^{10} \text{ Pa}$ and $\nu = 0.3$.⁹ This positive magnetoelastic anisotropy induced by the tensile strain contributes to the enhancement of the bulk PMA in the GdFe film. This simple model can explain also the previous results on electric-field-controlled PMA. In previous

literature, Co/Pt multilayers,^{34,35,37,38} CoPd alloy,^{26,27} Cu/Ni multilayers³⁶ and Co/Ni multilayers³³ are used in multiferroic heterostructures to modulate their PMA. Due to their negative magnetostriction coefficient, the electric field induces a negative magnetoelastic anisotropy that weakens their PMA such that the magnetization rotates from out of plane to in plane.^{36,38} Therefore, in our GdFe/PMN-PT multiferroic heterostructure, owing to the electric-field-induced tensile strain along the z axis in PMN-PT and the positive magnetostriction coefficient of the GdFe film, a positive strain-induced magnetoelastic anisotropy results in the enhancement of the PMA in the GdFe film.

Based on the different electric-field-modulated PMA with negative and positive magnetostriction coefficients, we can design a prototypal perpendicular magnetic tunnel junction (MTJ) to enable electrical control. As displayed in Figure 5a, the perpendicular MTJ consists of two magnetic layers with PMA, one magnetic layer with a positive magnetostriction coefficient (such as GdFe) and the other magnetic layer with a negative magnetostriction coefficient (such as Co/Pt multilayer). For the GdFe layer, electric fields enhance its PMA so that it can be treated as a pinned layer, whose magnetization always points out of plane. The magnetic layer with a negative magnetostriction coefficient, in contrast, serves as a free layer, since the electric field can switch its magnetization between out of plane and in plane.^{36,38} Thus, the magnetization configuration of this perpendicular MTJ can be electrically switched between parallel and perpendicular (Figure 5b), resulting in two reversible low and high resistance states without the assistance of a magnetic field, which is similar to the electrical control of magnetoresistance in the in-plane MTJ.^{46,47} The perpendicular MTJ with RE-TM alloys has been reported^{48,49} though its tunnel magnetoresistance is lower than CoFeB/MgO/CoFeB. It is also notable that although the electric-field modulation of PMA in GdFe film is volatile, the nonvolatile electric-field

modulation of PMA can be achieved by applying asymmetric electric fields.^{35,46}

CONCLUSIONS

In summary, we have demonstrated electrically enhanced bulk PMA in GdFe/PMN-PT multiferroic heterostructure through AHE measurements and Kerr imaging at room temperature. This enhancement of bulk PMA in a ferrimagnetic GdFe film was ascribed to a positive magnetoelastic anisotropy induced by the electric-field-generated tensile strain, which was confirmed by the XRD measurements. This electric-field-enhanced bulk PMA in the GdFe film enriches the electric-field control of PMA and provides a new method to modulate RE-TM alloys. Therefore, we expect our finding will stimulate future investigations into electric-field control of perpendicular MTJ and RE-TM alloys to explore electric-field modulation of domain wall motion and skyrmions.

EXPERIMENTAL SECTION

The 20 nm GdFe films were deposited on one-side-polished PMN-PT (011) single crystals (5 mm×6 mm×0.2 mm) using a magnetic sputtering system with a base pressure less than 10^{-7} Torr. A Gd₂₅Fe₇₅ target was mounted in the sputtering system with a 5 mTorr Ar gas pressure and 80 W DC sputtering power. Then, a 5 nm Pt layer was deposited to protect the GdFe film from oxidation. A rotating substrate stage was used for deposition, and a shadow mask was used to pattern the samples into a Hall bar shape for AHE measurements. Ti (10 nm) and Au (150 nm) layers were sputtered on the bottom of PMN-PT as an electrode. The in situ AHE measurements with various electric fields were performed by a homemade electromagnet system using a Keithley 6221 current source and a Keithley 2182 nanovolt meter. The voltage applied to the PMN-PT substrate was generated by a Keithley 6517 electrometer. The magnetic domains were imaged using a Kerr microscope in polar geometry. The XRD with in situ electric fields was

characterized by a Rigaku SmartLab 9 kW X-ray diffractometer with a Cu K α radiation. All the measurements were performed at room temperature.

ASSOCIATED CONTENT

Supporting Information.

The Supporting Information is available free of charge on the ACS Publications website at DOI: XXXXXX

Kerr images measured with varying electric fields after applying a series of magnetic field pulses, the AHE curves under in situ electric fields for 10 nm GdFe film (PDF)

AUTHOR INFORMATION

Corresponding Author

*E-mail: xixiang.zhang@kaust.edu.sa

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENT

This work was supported by King Abdullah University of Science and Technology (KAUST), Office of Sponsored Research (OSR) under Award No. CRF-2017-3427-CRG6. The authors acknowledge the Nanofabrication Core Lab at KAUST for their excellent assistance. The authors also acknowledge partial support from the Bureau of Facility Support and Budget, CAS, and the Anhui Initiative in Quantum Information Technologies (AHY100000).

REFERENCES

- (1) Ikeda, S.; Miura, K.; Yamamoto, H.; Mizunuma, K.; Gan, H. D.; Endo, M.; Kanai, S.; Hayakawa, J.; Matsukura, F.; Ohno, H. A Perpendicular-Anisotropy CoFeB-MgO Magnetic

Tunnel Junction. *Nat. Mater.* **2010**, *9*, 721-724.

(2) Watanabe, K.; Jinnai, B.; Fukami, S.; Sato, H.; Ohno, H. Shape Anisotropy Revisited in Single-Digit Nanometer Magnetic Tunnel Junctions. *Nat. Commun.* **2018**, *9*, 663.

(3) Dieny, B.; Chshiev, M. Perpendicular Magnetic Anisotropy at Transition Metal/Oxide Interfaces and Applications. *Rev. Mod. Phys.* **2017**, *89*, 025008.

(4) Kurt, H.; Rode, K.; Venkatesan, M.; Stamenov, P.; Coey, J. M. D. High Spin Polarization in Epitaxial Films of Ferrimagnetic Mn_3Ga . *Phys. Rev. B* **2011**, *83*, 020405(R).

(5) He, P.; Ma, L.; Shi, Z.; Guo, G. Y.; Zheng, J. G.; Xin, Y.; Zhou, S. M. Chemical Composition Tuning of the Anomalous Hall Effect in Isoelectronic $\text{L}_{10}\text{FePd}_{1-x}\text{Pt}_x$ Alloy Films. *Phys. Rev. Lett.* **2012**, *109*, 066402.

(6) Seemann, K. M.; Mokrousov, Y.; Aziz, A.; Miguel, J.; Kronast, F.; Kuch, W.; Blamire, M. G.; Hindmarch, A. T.; Hickey, B. J.; Souza, I.; Marrows, C. H. Spin-Orbit Strength Driven Crossover Between Intrinsic and Extrinsic Mechanisms of the Anomalous Hall Effect in the Epitaxial L_{10} -Ordered Ferromagnets FePd and FePt. *Phys. Rev. Lett.* **2010**, *104*, 076402.

(7) Woo, S.; Song, K. M.; Zhang, X.; Zhou, Y.; Ezawa, M.; Liu, X.; Finizio, S.; Raabe, J.; Lee, N. J.; Kim, S.; Park, S.; Kim, Y.; Kim, J.; Lee, D.; Lee, O.; Choi, J. W.; Min, B.; Koo, H. C.; Chang, J. Current-Driven Dynamics and Inhibition of the Skyrmion Hall Effect of Ferrimagnetic Skyrmions in GdFeCo Films. *Nat. Commun.* **2018**, *9*, 959.

(8) Woo, S.; Song, K. M.; Zhang, X.; Ezawa, M.; Zhou, Y.; Liu, X.; Weigand, M.; Finizio, S.; Raabe, J.; Park, M.; Lee, K.; Choi, J. W.; Min, B.; Koo, H. C.; Chang, J. Deterministic Creation and Deletion of a Single Magnetic Skyrmion Observed by Direct Time-Resolved X-Ray Microscopy. *Nat. Electron.* **2018**, *1*, 288-296.

(9) Yoshino, S.; Tsunashima, S.; Masuda, M.; Uchiyama, S. Magnetostriction and

Perpendicular Magnetic Anisotropy of Amorphous GdFeCo Thin Films. *Jpn. J. Appl. Phys.* **1988**, 27, 1247.

(10) Stanciu, C. D.; Hansteen, F.; Kimel, A. V.; Kirilyuk, A.; Tsukamoto, A.; Itoh, A.; Rasing, T. All-Optical Magnetic Recording with Circularly Polarized Light. *Phys. Rev. Lett.* **2007**, 99, 047601.

(11) Oh, Y.; Baek, S. C.; Kim, Y. M.; Lee, H. Y.; Lee, K.; Yang, C.; Park, E.; Lee, K.; Kim, K.; Go, G.; Jeong, J.; Min, B.; Lee, H.; Lee, K.; Park, B. Field-Free Switching of Perpendicular Magnetization through Spin-Orbit Torque in Antiferromagnet/Ferromagnet/Oxide Structures. *Nat. Nanotechnol.* **2016**, 11, 878-884.

(12) Fukami, S.; Anekawa, T.; Zhang, C.; Ohno, H. A Spin-Orbit Torque Switching Scheme with Collinear Magnetic Easy Axis and Current Configuration. *Nat. Nanotechnol.* **2016**, 11, 621-625.

(13) Fukami, S.; Zhang, C.; DuttaGupta, S.; Kurenkov, A.; Ohno, H. Magnetization Switching by Spin-Orbit Torque in an Antiferromagnet-Ferromagnet Bilayer System. *Nat. Mater.* **2016**, 15, 535-541.

(14) Lau, Y.; Betto, D.; Rode, K.; Coey, J. M. D.; Stamenov, P. Spin-Orbit Torque Switching without an External Field Using Interlayer Exchange Coupling. *Nat. Nanotechnol.* **2016**, 11, 758-762.

(15) Yu, G.; Upadhyaya, P.; Fan, Y.; Alzate, J. G.; Jiang, W.; Wong, K. L.; Takei, S.; Bender, S. A.; Chang, L.; Jiang, Y.; Lang, M.; Tang, J.; Wang, Y.; Tserkovnyak, Y.; Amiri, P. K.; Wang, K. L. Switching of Perpendicular Magnetization by Spin-Orbit Torques in the Absence of External Magnetic Fields. *Nat. Nanotechnol.* **2014**, 9, 548-554.

(16) Cai, K.; Yang, M.; Ju, H.; Wang, S.; Ji, Y.; Li, B.; Edmonds, K. W.; Sheng, Y.; Zhang,

B.; Zhang, N.; Liu, S.; Zheng, H.; Wang, K. Electric Field Control of Deterministic Current-Induced Magnetization Switching in a Hybrid Ferromagnetic/Ferroelectric Structure. *Nat. Mater.* **2017**, *16*, 712-716.

(17) Bi, C.; Liu, Y.; Newhouse-Illige, T.; Xu, M.; Rosales, M.; Freeland, J. W.; Mryasov, O.; Zhang, S.; Te Velthuis, S. G. E.; Wang, W. G. Reversible Control of Co Magnetism by Voltage-Induced Oxidation. *Phys. Rev. Lett.* **2014**, *113*, 267202.

(18) Bauer, U.; Yao, L.; Tan, A. J.; Agrawal, P.; Emori, S.; Tuller, H. L.; van Dijken, S.; Beach, G. S. D. Magneto-Ionic Control of Interfacial Magnetism. *Nat. Mater.* **2015**, *14*, 174-181.

(19) Tan, A. J.; Huang, M.; Avci, C. O.; Buttner, F.; Mann, M.; Hu, W.; Mazzoli, C.; Wilkins, S.; Tuller, H. L.; Beach, G. S. D. Magneto-Ionic Control of Magnetism Using a Solid-State Proton Pump. *Nat. Mater.* **2019**, *18*, 35-41.

(20) Ohno, H.; Chiba, D.; Matsukura, F.; Omiya, T.; Abe, E.; Dietl, T.; Ohno, Y.; Ohtani, K. Electric-Field Control of Ferromagnetism. *Nature* **2000**, *408*, 944-946.

(21) Chen, A. T.; Zhao, Y. G. Electrical Manipulation of Magnetism through Strain-Mediated Magnetoelectric Coupling in Multiferroic Heterostructures. *APL Mater.* **2016**, *4*, 032303.

(22) Hu, J.; Chen, L.; Nan, C. Multiferroic Heterostructures Integrating Ferroelectric and Magnetic Materials. *Adv. Mater.* **2016**, *28*, 15-39.

(23) Vaz, C. A. F. Electric Field Control of Magnetism in Multiferroic Heterostructures. *J. Phys.: Condens. Matter* **2012**, *24*, 333201.

(24) Heron, J. T.; Bosse, J. L.; He, Q.; Gao, Y.; Trassin, M.; Ye, L.; Clarkson, J. D.; Wang, C.; Liu, J.; Salahuddin, S.; Ralph, D. C.; Schlom, D. G.; Iniguez, J.; Huey, B. D.; Ramesh, R. Deterministic Switching of Ferromagnetism at Room Temperature Using an Electric Field. *Nature* **2014**, *516*, 370-373.

- (25) Chen, A.; Zhao, Y.; Li, P.; Zhang, X.; Peng, R.; Huang, H.; Zou, L.; Zheng, X.; Zhang, S.; Miao, P.; Lu, Y.; Cai, J.; Nan, C. Angular Dependence of Exchange Bias and Magnetization Reversal Controlled by Electric-Field-Induced Competing Anisotropies. *Adv. Mater.* **2016**, *28*, 363-369.
- (26) Lee, J. W.; Shin, S. C.; Kim, S. K. Spin Engineering of CoPd Alloy Films Via the Inverse Piezoelectric Effect. *Appl. Phys. Lett.* **2003**, *82*, 2458-2460.
- (27) Kim, J.; Ryu, K.; Jeong, J.; Shin, S. Large Converse Magnetoelectric Coupling Effect at Room Temperature in CoPd/PMN-PT (001) Heterostructure. *Appl. Phys. Lett.* **2010**, *97*, 252508.
- (28) Lei, N.; Park, S.; Lecoeur, P.; Ravelosona, D.; Chappert, C.; Stelmakhovich, O.; Holy, V. Magnetization Reversal Assisted by the Inverse Piezoelectric Effect in Co-Fe-B/Ferroelectric Multilayers. *Phys. Rev. B* **2011**, *84*, 012404.
- (29) Tsai, W.; Liao, S.; Huang, K.; Wang, D.; Lai, C. Nonvolatile Electric-Field Modulation of Magnetic Anisotropy in Perpendicularly Magnetized $L1_0$ -FePt/(001)[Pb(Mg $_{1/3}$ Nb $_{2/3}$)] $_{0.7}$ -(PbTiO $_3$) $_{0.3}$ Heterostructures. *Appl. Phys. Lett.* **2013**, *103*, 252405.
- (30) Yang, Y. T.; Zhang, Q. M.; Wang, D. H.; Song, Y. Q.; Wang, L. Y.; Lv, L. Y.; Cao, Q. Q.; Du, Y. W. Electric Field Control of Magnetic Properties in CoPt/Pb(Mg $_{1/3}$ Nb $_{2/3}$)O $_3$ -PbTiO $_3$ Heterostructure at Room Temperature. *Appl. Phys. Lett.* **2013**, *103*, 082404.
- (31) Yu, G.; Wang, Z.; Abolfath-Beygi, M.; He, C.; Li, X.; Wong, K. L.; Nordeen, P.; Wu, H.; Carman, G. P.; Han, X.; Alhomoudi, I. A.; Amiri, P. K.; Wang, K. L. Strain-Induced Modulation of Perpendicular Magnetic Anisotropy in Ta/CoFeB/MgO Structures Investigated by Ferromagnetic Resonance. *Appl. Phys. Lett.* **2015**, *106*, 072402.
- (32) Shepley, P. M.; Rushforth, A. W.; Wang, M.; Burnell, G.; Moore, T. A. Modification of Perpendicular Magnetic Anisotropy and Domain Wall Velocity in Pt/Co/Pt by Voltage-Induced

Strain. *Sci. Rep.* **2015**, *5*, 7921.

(33) Gopman, D. B.; Dennis, C. L.; Chen, P. J.; Iunin, Y. L.; Finkel, P.; Staruch, M.; Shull, R. D. Strain-Assisted Magnetization Reversal in Co/Ni Multilayers with Perpendicular Magnetic Anisotropy. *Sci. Rep.* **2016**, *6*, 27774.

(34) Sun, Y.; Ba, Y.; Chen, A.; He, W.; Wang, W.; Zheng, X.; Zou, L.; Zhang, Y.; Yang, Q.; Yan, L.; Feng, C.; Zhang, Q.; Ca, J.; Wu, W.; Liu, M.; Gu, L.; Cheng, Z.; Nan, C.; Qiu, Z.; Wu, Y.; Li, J.; Zhao, Y. Electric-Field Modulation of Interface Magnetic Anisotropy and Spin Reorientation Transition in (Co/Pt)₃/PMN-PT Heterostructure. *ACS Appl. Mater. Interfaces* **2017**, *9*, 10855-10864.

(35) Peng, B.; Zhou, Z.; Nan, T.; Dong, G.; Feng, M.; Yang, Q.; Wang, X.; Zhao, S.; Xian, D.; Zhuang-De, J.; Ren, W.; Ye, Z.; Sung, N. X.; Liu, M. Deterministic Switching of Perpendicular Magnetic Anisotropy by Voltage Control of Spin Reorientation Transition in (Co/Pt)₃/Pb(Mg_{1/3}Nb_{2/3})O₃-PbTiO₃ Multiferroic Heterostructures. *ACS Nano* **2017**, *11*, 4337-4345.

(36) Shirahata, Y.; Shiina, R.; Gonzalez, D. L.; Franke, K. J. A.; Wada, E.; Itoh, M.; Pertsev, N. A.; van Dijken, S.; Taniyama, T. Electric-Field Switching of Perpendicularly Magnetized Multilayers. *NPG Asia Mater.* **2015**, *7*, e198.

(37) Yang, Q.; Nan, T.; Zhang, Y.; Zhou, Z.; Peng, B.; Ren, W.; Ye, Z.; Sun, N. X.; Liu, M. Voltage Control of Perpendicular Magnetic Anisotropy in Multiferroic (Co/Pt)₃/PbMg_{1/3}Nb_{2/3}O₃-PbTiO₃ Heterostructures. *Phys. Rev. Applied* **2017**, *8*, 044006.

(38) Peng, B.; Feng, M.; Zhang, Y.; Zhou, Z.; Hu, Z.; Liu, M. Voltage Control of Perpendicular Magnetic Anisotropy in (Co/Pt)₃/Pb(Zn_{1/3}Nb_{2/3})O₃-PbTiO₃ Multiferroic Heterostructures at Room Temperature. *Appl. Phys. Lett.* **2018**, *113*, 142901.

- (39) Hassdenteufel, A.; Hebler, B.; Schubert, C.; Liebig, A.; Teich, M.; Helm, M.; Aeschlimann, M.; Albrecht, M.; Bratschitsch, R. Thermally Assisted All-Optical Helicity Dependent Magnetic Switching in Amorphous $\text{Fe}_{100-x}\text{Tb}_x$ Alloy Films. *Adv. Mater.* **2013**, *25*, 3122-3128.
- (40) Kim, K.; Kim, S. K.; Hirata, Y.; Oh, S.; Tono, T.; Kim, D.; Okuno, T.; Ham, W. S.; Kim, S.; Go, G.; Tserkovnyak, Y.; Tsukamoto, A.; Moriyama, T.; Lee, K.; Ono, T. Fast Domain Wall Motion in the Vicinity of the Angular Momentum Compensation Temperature of Ferrimagnets. *Nat. Mater.* **2017**, *16*, 1187-1192.
- (41) Hirata, Y.; Kim, D.; Kim, S. K.; Lee, D.; Oh, S.; Kim, D.; Nishimura, T.; Okuno, T.; Futakawa, Y.; Yoshikawa, H.; Tsukamoto, A.; Tserkovnyak, Y.; Shiota, Y.; Moriyama, T.; Choe, S.; Lee, K.; Ono, T. Vanishing Skyrmion Hall Effect at the Angular Momentum Compensation Temperature of a Ferrimagnet. *Nat. Nanotechnol.* **2019**, *14*, 232-236.
- (42) Caretta, L.; Mann, M.; Büttner, F.; Ueda, K.; Pfau, B.; Günther, C. M.; Hessing, P.; Churikova, A.; Klose, C.; Schneider, M.; Engel, D.; Marcus, C.; Bono, D.; Bagschik, K.; Eisebitt, S.; Beach, G. S. D. Fast Current-Driven Domain Walls and Small Skyrmions in a Compensated Ferrimagnet. *Nat. Nanotechnol.* **2018**, *13*, 1154-1160.
- (43) Katayama, T.; Hirano, M.; Koizumi, Y.; Kawanishi, K.; Tsushima, T. Different Origin of the Perpendicular Anisotropy in Amorphous Gd-Fe From Gd-Co Films. *IEEE T. Magn.* **1977**, *13*, 1603.
- (44) Tsunashima, S.; Takagi, H.; Kamegaki, K.; Fujii, T.; Uchiyama, S. Magnetoelastic Contribution to Perpendicular Anisotropy in Amorphous Gd-Co and Gd-Fe Films. *IEEE T. Magn.* **1978**, *14*, 844.
- (45) Heidler, J.; Fechner, M.; Chopdekar, R. V.; Piamonteze, C.; Dreiser, J.; Jenkins, C. A.;

Arenholz, E.; Rusponi, S.; Brune, H.; Spaldin, N. A.; Nolting, F. Magnetoelectroelastic Control of Magnetism in an Artificial Multiferroic. *Phys. Rev. B* **2016**, *94*, 014401.

(46) Chen, A.; Wen, Y.; Fang, B.; Zhao, Y.; Zhang, Q.; Chang, Y.; Li, P.; Wu, H.; Huang, H.; Lu, Y.; Zeng, Z.; Cai, J.; Han, X.; Wu, T.; Zhang, X.; Zhao, Y. Giant Nonvolatile Manipulation of Magnetoresistance in Magnetic Tunnel Junctions by Electric Fields Via Magnetoelectric Coupling. *Nat. Commun.* **2019**, *10*, 243.

(47) Li, P.; Chen, A.; Li, D.; Zhao, Y.; Zhang, S.; Yang, L.; Liu, Y.; Zhu, M.; Zhang, H.; Han, X. Electric Field Manipulation of Magnetization Rotation and Tunneling Magnetoresistance of Magnetic Tunnel Junctions at Room Temperature. *Adv. Mater.* **2014**, *26*, 4320-4325.

(48) Nishimura, N.; Hirai, T.; Koganei, A.; Ikeda, T.; Okano, K.; Sekiguchi, Y.; Osada, Y. Magnetic Tunnel Junction Device with Perpendicular Magnetization Films for High-Density Magnetic Random Access Memory. *J. Appl. Phys.* **2002**, *91*, 5246-5249.

(49) Kinjo, H.; Machida, K.; Matsui, K.; Aoshima, K.; Kato, D.; Kuga, K.; Kikuchi, H.; Shimidzu, N. Low-Current-Density Spin-Transfer Switching in Gd₂₂Fe₇₈-MgO Magnetic Tunnel Junction. *J. Appl. Phys.* **2014**, *115*, 203903.

Figure 1. Schematic of the GdFe/PMN-PT multiferroic heterostructure. The external electric field was applied on the PMN-PT substrate to generate strain, which can modulate the bulk PMA of the GdFe film.

Figure 2. (a) The AHE curves measured with in situ electric fields. The guided arrow reveals the increasing trend of the AHE curves in conjunction with increasing electric fields. (b) Electric-field dependence of the normalized remnant R_{Hall} at $H_{\perp} = 0$ Oe as extracted from the AHE curves in (a). The increment of the normalized remnant R_{Hall} at $H_{\perp} = 0$ Oe indicates the enhancement of bulk PMA in the GdFe film. The arrows represent the electric-field sweeping directions.

Figure 3. (a) Illustration of the sequential negative magnetic field pulses with a width of 0.1 s that were applied to reverse the magnetic domain for Kerr image measurements. Prior to applying the negative magnetic field pulses, the sample was first saturated by applying a positive magnetic field of 300 Oe. (b) The Kerr images taken after applying magnetic field pulses of -50 Oe and -60 Oe under $E = 0 \text{ kV cm}^{-1}$, 10 kV cm^{-1} and 20 kV cm^{-1} . The electric fields had a significant effect on the magnetic domain structures, especially in the areas encircled by the yellow dashed lines. The scale bar is $100 \text{ }\mu\text{m}$. (c) The magnetic domain reversal ratio as a function of the amplitude of the magnetic field pulses under various electric fields. Around the coercive field, the electric-field-induced decrement of the domain reversal ratio suggests an enhancement of the PMA as directed by the arrow.

Figure 4. (a) The XRD curves of the PMN-PT substrate with various electric fields. Both (011) and (022) diffraction peaks shifted to smaller angles, indicating the expansion of the out-of-plane lattice. (b) A nearly linear tensile strain along the z axis induced by electric fields, which was estimated based on the shift of the (022) peak in (a). This tensile strain results in the electrically enhanced bulk PMA due to the positive magnetostriction coefficient of the GdFe film.

Figure 5. (a) Schematic of the proposed prototypal perpendicular MTJ on the ferroelectric substrate. The magnetic layer with a positive magnetostriction coefficient (such as GdFe) serves as the pinned layer, while the magnetic layer with a negative magnetostriction coefficient (such as Co/Pt) serves as the free layer; a thin insulating layer (such as MgO) is inserted between the two PMA layers. (b) The schematic illustration of the electric-field manipulation of the perpendicular MTJ. The magnetizations of the two magnetic layers are parallel without an electric field, which indicates low resistance. Applying an electric field rotates the magnetization of the free layer to in plane but does not affect magnetization in the pinned layer, resulting in a perpendicular magnetization configuration of the MTJ with a high resistance. This low/high bistable resistance states can be reversibly switched by electric fields.

TOC