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Highly efficient deterministic voltage control of the perpendicular Néel vector in a magnetoelectric antiferromagnet

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Efficient manipulation of the antiferromagnetic (AFM) state is desirable for advanced spintronic devices with fast operation and robustness regarding magnetic field perturbation. In this article, we demonstrate deterministic bipolar AFM spin reversal induced by a low voltage below 0.8 V (51 mV/nm) based on the magnetoelectric (ME) effect in a Pt/Cr₂O₃/Ir epitaxial trilayer. A stable spin direction is determined depending on the sign of the electric field and magnetic field product, enabling deterministic access to four quadrants in E - H space. With the help of first-principles calculations, we show that the voltage effect on the magnetic anisotropy energy density is a minor effect, whereas the ME effect determines both the stable spin state and the switching behavior. Our findings not only advance voltage-based insulating AFM spintronics but also provide a basis for the device function, such as a deterministic logic operation by means of the field combination.

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I. INTRODUCTION

Manipulation of spin is crucial in spintronic devices since the device function relies on the spin direction of a magnetic layer in the devices. External fields to manipulate the spins have been used, from a conventional magnetic field to modern current-based stimuli. Although the spin-polarized current can apply a torque on the spins to switch the direction, known as the spin transfer torque [1–3] and the spin-orbit torque [4–6], it requires a huge current density typically greater than 10^{10} A/m² [7,8], giving rise to high energy consumption and a temperature rise due to Joule heating.

A voltage-driven mechanism is an alternative, energy-efficient manipulation principle, and it also benefits from complementary metal-oxide semiconductor (CMOS) compatibility. There are typically two distinct physical phenomena that couple voltage and the magnetic moment: the voltage-controlled magnetic anisotropy (VCMA) effect [9,10] and the magnetoelectric (ME) effect [11,12]. The VCMA effect refers to a change in the magnetic anisotropy energy density at a metallic magnetic layer/nonmagnetic insulator interface by E . Since the

discovery of the VCMA effect in an all-solid-state system [10], VCMA-based spin manipulation has been explored for various ferromagnetic (FM) metal/oxide interfaces [13–17]. The underlying mechanism of the VCMA effect is charge doping and/or charge redistribution causing changes in the orbital magnetic moment and the quadrupolar magnetic moment, respectively [13,18,19]. Since the VCMA effect only reduces the energy barrier and does not change the symmetry of the magnetic anisotropy energy landscape, the magnetization switching is not deterministic [20,21].

The ME effect is another mechanism for voltage-induced spin manipulation, and is attributed to the induction of a net magnetization M by an electric field E and the induction of a dielectric polarization P by a magnetic field H [11,12]. This effect typically occurs in an antiferromagnetic (AFM) insulator in both the bulk [11,12,22–24] and a thin film [25–28] and it is rather compatible with recently emerging AFM spintronics [29]. The ME effect has been developed into multiferroic effect [30,31], manifested in, for example, BiFeO₃ [32], where magnetism, dielectricity, and elasticity are coupled. In this system, the lattice constraint due to epitaxial growth degrades the multiferroicity, which becomes significant on lowering of the thickness. The linear ME effect can overcome this

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limitation since the lattice deformation is not involved for the ME effect to occur. It has been attempted to use Cr_2O_3 , a prototypical linear ME AFM material [11,12], in voltage-driven spintronic devices due to the possible occurrence of the ME effect above room temperature. The key for the achievement is control of the spin-polarized surface state called the “boundary magnetization” [33] or the “surface magnetization” [34] inherently existing in ME AFM materials, due to the built-in electric field in conjunction with the ME effect. The boundary magnetization also generates spin-dependent transport phenomena at the interface with a nonmagnetic heavy metal such as the anomalous Hall effect (AHE) [35–38] and spin Hall magnetoresistance [39], which can be used as the detection principle for the boundary magnetization and consequently the Néel vector $\mathbf{l}$. While the AHE itself emerges in bulk Cr_2O_3 [38], no hysteresis was observed in the bulk [38] or in thick films, e.g., 200-nm-thick, films [37]. This is because the bulk AFM nature dominates the total magnetic energy density in these systems. Recently, we reported external field switching of $\mathbf{l}$ by taking advantage of the enhanced contribution of the boundary magnetization by reducing the Cr_2O_3 thickness to 15 nm [40]. Consequently, we also found that the switching field was modulated by the electric field E , with high efficiency above 4.8 T nm/V, due to the ME effect in the Cr_2O_3 layer [28]. These previous reports led to the work presented in this paper reporting the voltage-induced reversible switching of $\mathbf{l}$.

In this article, we demonstrate the voltage-induced bipolar switching of $\mathbf{l}$. We reveal that the AFM spin direction is coupled with the sign of the EH product and it can be described by a checkerboardlike pattern in the neighboring four quadrants in E - H space. The results, together with the first-principles calculations, suggest the ME effect is responsible for spin switching.

II. SAMPLE PREPARATION AND FILM QUALITY

A Pt(2 nm)/ Cr_2O_3 (15 nm)/Ir(20 nm) trilayer was fabricated on an α - Al_2O_3 (0001) substrate by the dc. magnetron

sputtering method. The base pressure of the deposition system was typically below 1×10^{-6} Pa. The substrate was ultrasonically cleaned with acetone, 2-propanol, distilled water, and ethanol before being placed into a vacuum chamber and subsequently heated for more than 7 h at 873 K in the deposition chamber. The Ir buffer layer was deposited at 873 K. On the Ir buffer layer, the Cr_2O_3 layer was deposited by means of the reactive magnetron sputtering technique for the metal Cr target with an Ar and O_2 gas mixture. The deposition temperature was 773 K. Subsequently, the top Pt layer was deposited at room temperature. Structural investigation was performed by *in situ* reflection high-energy electron diffraction and x-ray diffraction (XRD). The x-ray reflection (XRR) profile was measured to estimate the actual thickness of each layer and the interface roughness. We also measured the high-angle 2θ - ω profile with the scattering vector $\mathbf{q}$ along the film normal. The XRR and XRD measurements were done with Cu $K\alpha$ radiation. All structural investigations were done at room temperature. Detailed structural information is presented in Supplemental Material [41].

We used the AHE as the detection scheme for the interfacial AFM moments [35–37]. The Hall device with 5 μm width and 25 μm length was fabricated by means of photolithography and Ar ion milling. An optical microscope image with the equivalent circuit and a schematic drawing of the device are shown in Fig. 1(a). A gate voltage (an electric field) V_G (E) is applied across the Cr_2O_3 layer between the top and bottom electrodes. The I - V_G curves across the Cr_2O_3 layer are shown in Fig. 1(b). The nonlinear I - V_G curves indicate the insulating nature of the Cr_2O_3 layer, ensuring the above-mentioned prerequisite. The leakage current (the current density) at -0.2 V (-63 mV/nm) is about 20 nA (13 A/m²) at room temperature and it decreases with decreasing temperature. The low leakage current is associated with a good crystalline quality of the Cr_2O_3 layer (see also Supplemental Material [41]).

Upon the AHE measurements, the current essentially flows in the Pt layer and, therefore, the AHE signal is due to magnetism at the Pt/ Cr_2O_3 interface, such

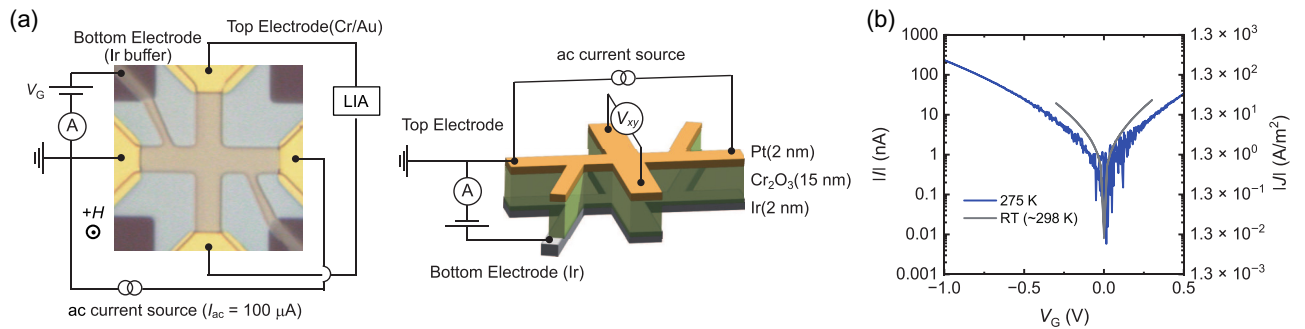


FIG. 1. (a) Optical microscope image of the fabricated Hall device with the equivalent electric circuit. A schematic drawing of the three-dimensional structure of the device is also shown. LIA denotes the Lock-in amplifier. (b) I - V curves across the Cr_2O_3 layer. Blue and gray curves represent the data measured at 275 K and room temperature (RT; approximately 298 K), respectively.

as the above-mentioned boundary magnetization [33,34]. While some other mechanisms for the AHE at the heavy metal/Cr₂O₃ interface have been proposed, such as AHE by spin-polarized Pt [35], spin Hall magnetoresistance [42], and interfacial spin texture in the momentum space [43], all mechanisms reasonably assume that the AHE is a fingerprint of **I**. The temperature dependence of the AHE response supports this assumption and gives the Néel temperature as 279 K [41].

The transverse voltage V_{xy} was measured with a lock-in amplifier under an alternating current I_{ac} at 1 kHz with an amplitude of 100 μ A. R_{xy} (V_{xy}/I_{ac}) was measured as a function of either H or E , which were applied in the direction perpendicular to the film plane [see Fig. 1(a)]. A constant V_G (E) was applied during the R_{xy} - H measurements and a constant H , H_{const} , was applied during the R_{xy} - V_G measurements. The background signal especially for the R_{xy} - V_G curve was collected at $\mu_0 H_{const} = 0$ T, and was subtracted from the raw data for other R_{xy} - V_G curves. A positive direction of the field was defined as the direction from the bottom to the top of the film. We varied the measurement temperature from 220 to 280 K.

III. MAGNETOELECTRIC SWITCHING OF THE PERPENDICULAR NÉEL VECTOR

Figure 2 shows a series of R_{xy} - H loops under V_G application measured at 275 K. Rectangular hysteresis loops are observed in agreement with our previous report [40]. The reason why the hysteresis in the R_{xy} - H curve appears can

be found in Ref. [40]. With $V_G > -0.3$ V [Fig. 2(a)], up-to-down switching of R_{xy} was observed in the positive H direction and the opposite switching was observed in the negative H direction. The switching field monotonically decreases with increasing V_G . Notably, when $V_G < -0.6$ V [Fig. 2(b)], the switching direction was reversed: down-to-up switching in the positive H direction and up-to-down switching in the negative H direction. Besides, the V_G dependence of the switching field shows the opposite trend: the switching field decreases with decreasing V_G , i.e., increasing the absolute value of V_G .

Figure 3 shows the switching magnetic field $\mu_0 H_{sw}$ as a function of V_G as solid symbols, which resembles an asteroid curve. Outside the asteroid, the spin state is monostable, either up or down, and inside it is bistable, corresponding to the bistable region of the hysteresis shown in Fig. 2. The presence of the monostable region indicates that the switching occurs deterministically. The spin direction of the monostable region in each quadrant is assigned in a checkerboard manner, i.e., the spin state was determined by the sign of the EH product. A similar deterministic four-quadrant assignment of the spin state was reported in our previous work [28], where 40-nm-thick Cr₂O₃ was used. The important advance from the previous work is that we are able to significantly reduce the switching voltage by reducing the Cr₂O₃ thickness while maintaining a good insulating property.

The results in Figs. 2 and 3 suggest that V_G -induced switching at a constant $\mu_0 H_{const}$ is also possible. Figure 4 shows R_{xy} as a function of V_G with $\mu_0 H_{const}$ ranging

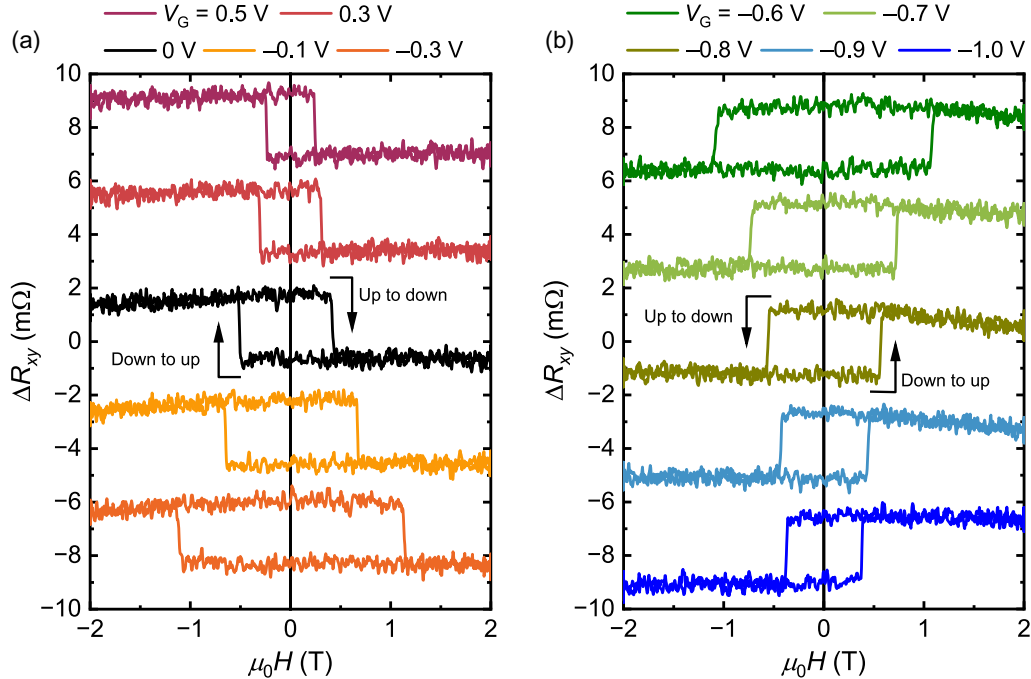


FIG. 2. R_{xy} - H curves under various values of constant V_G . (a),(b) Curves for $V_G \geq -0.3$ V and $V_G \leq -0.6$ V, respectively. The arrows represent the switching direction of R_{xy} . The measurement temperature was 275 K.

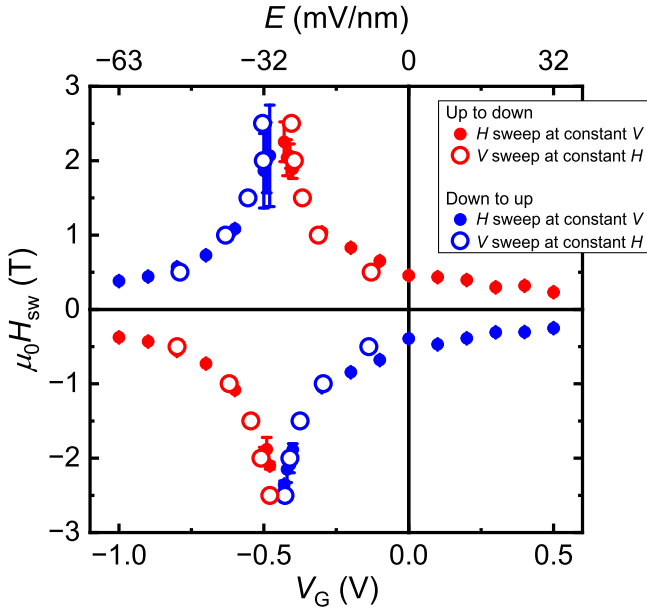


FIG. 3. Switching field in the E - H plane. Red and blue symbols represent the data for the up-to-down and down-to-up switching, respectively. Closed and open symbols denote the data collected from the R_{xy} - H curves (Fig. 2) and the R_{xy} - V_G curves (Fig. 4), respectively. “Up to down” and “down to up” are defined in Figs. 2 and 4.

from -5.0 to 5.0 T. The curve shows a hysteresis behavior for $|\mu_0 H_{\text{const}}| \leq 2.5$ T, indicating that voltage-induced bipolar switching was successfully achieved. Remarkably, the switching voltage was below 0.8 V with a relatively small $\mu_0 H_{\text{const}}$ of approximately 0.5 T. For both $\mu_0 H_{\text{const}}$

polarities, the switching voltage monotonically decreases with increasing absolute value of $\mu_0 H_{\text{const}}$, as plotted by open symbols in Fig. 3. The switching direction is reversed by the polarity of $\mu_0 H_{\text{const}}$, in agreement with what is seen in Fig. 2. The asteroid feature strongly suggests the ME effect denoted by αEH (α is the ME susceptibility) is dominant in the present spin reversal [44]. It is also found that the asteroid shifts along a horizontal axis toward negative V_G . The actual switching condition is roughly expressed as $\alpha(E - E_0)H$, where E_0 denotes the offset electric field corresponding to the magnitude of the uncompensated AFM moment at 0 V [28,40].

Here we discuss the switching condition at a high magnetic field effect based on the ME effect. For the ME effect, the magnetization M induced by the electric field E and the dielectric polarization P induced by the magnetic field H are coupled with each other [24]. The coupling induces the H -induced polarization flop [45,46], which is relevant to the spin-flop transition [47]. In our experiments, a similar effect manifests itself in the disappearance of the hysteresis in the R_{xy} - V_G curve. According to Fig. 4, the hysteresis disappears for $|\mu_0 H_{\text{const}}| \geq 3.5$ T, while the S-shaped curve barely remains. The decrease of the AHE amplitude and the disappearance of the R_{xy} - V_G loop suggest that the spin state in the Cr_2O_3 layer is disturbed by the high magnetic field. The disappearance of hysteresis in the R_{xy} - V_G curve manifests itself as the intersection of two hyperbolic curves in Fig. 3. Our observation seems to be consistent with the H dependence of the ME coefficient along the c axis, α_{33} : the ME coefficient α_{33} decreases with increasing H and becomes zero at the spin-flop field [48]. However, $|\mu_0 H_{\text{const}}| \sim 4$ T is quite small compared with the spin-flop

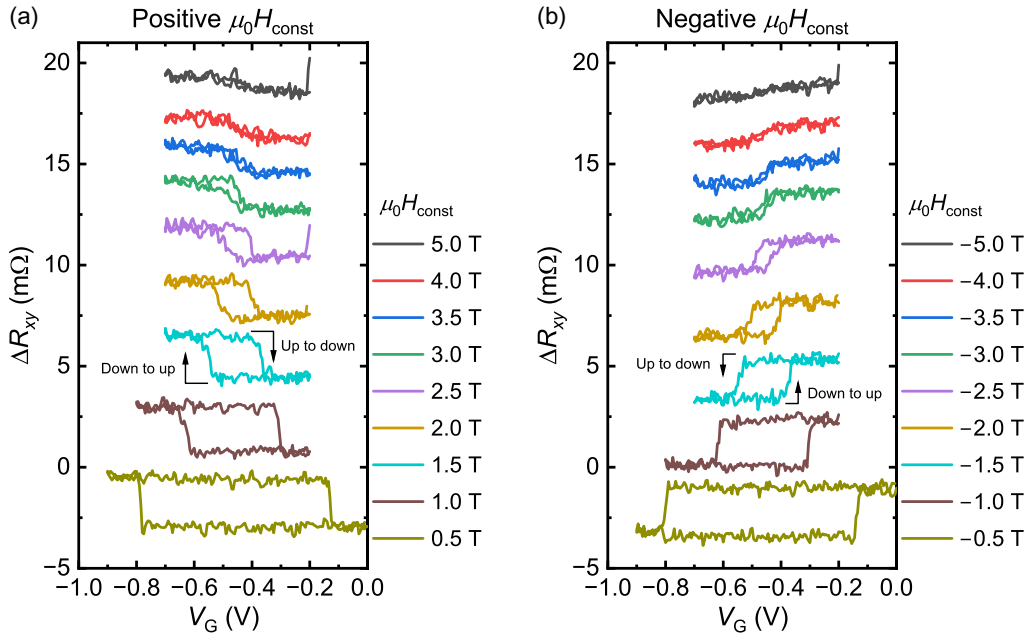


FIG. 4. R_{xy} - V_G curves under constant H . (a), (b) R_{xy} - V_G curves with positive and negative $\mu_0 H_{\text{const}}$, respectively.

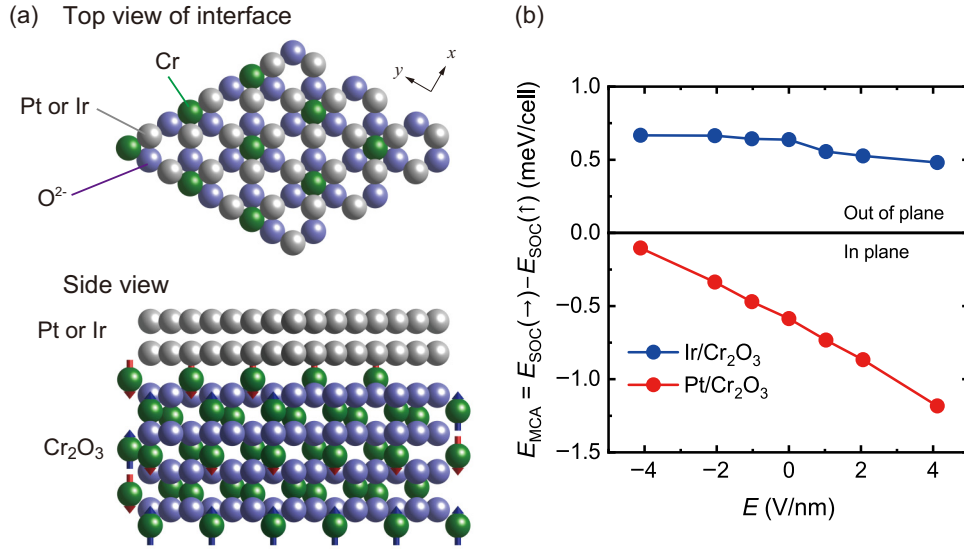


FIG. 5. (a) Film structure used for the density functional theory calculations. The top and bottom images represent the top and side views of the film, respectively. Green circles represent Cr³⁺ ions, purple circles represent O²⁻ ions, and light gray circles represent heavy metal atoms, e.g., Pt or Ir. Red and blue arrows in the side view represent magnetic moments. (b) MCA energy density as a function of electric field. Red and blue circles represent the values for Pt/Cr₂O₃ and Ir/Cr₂O₃, respectively. The results for Pt/Cr₂O₃ are reproduced from Ref. [28]. SOC, spin-orbit coupling.

field of bulk Cr₂O₃ (approximately 12 T) [49]. Although the details of the polarization flop need further investigation, the low critical field implies that the exchange field and/or the anisotropy field at the interface is distinct from that in the bulk [41].

To theoretically scrutinize the physical mechanism of the present observation, we performed first-principles calculations to compare the ME effect with another possibility, the VCMA effect. Figure 5(a) shows the calculation model and how E can modify the magnetocrystalline anisotropy (MCA) of the Pt(Ir)/Cr₂O₃ bilayer structure [41]. Figure 5(b) shows the changes in the MCA energy density as a function of E . The MCA energy density depends on the heavy metal layer, i.e., Pt or Ir, indicating that the interface magnetic anisotropy K_i is the main origin. As in the case of metallic FM ultrathin films, the MCA energy density monotonically decreases with increasing E . The slope of this change $\Delta K_i/\Delta E$ gives the VCMA efficiency as -19 fJ/V m for the Ir/Cr₂O₃ interface and -98 fJ/V m for the Pt/Cr₂O₃ interface [28], which are comparable to or lower than the values for metallic FM ultrathin films [13].

Since the spontaneous magnetization cannot be defined in our film due to the AFM nature of Cr₂O₃, the VCMA efficiency is difficult to evaluate experimentally. Besides, the domain wall motion, which dominates the switching process [39,41], complicates the quantitative analysis of the switching energy. Alternatively, we compare the V_G -induced and E -induced modulation efficiency of the switching magnetic field, $\Delta\mu_0 H_{\text{sw}}/\Delta V_G$, around 0 V with the efficiency in the VCMA-driven case. In the case of a

metallic ultrathin FM/insulator stacking structure such as Co-Fe-B/MgO, $|\Delta\mu_0 H_{\text{sw}}/\Delta V_G|$ is at most about 21 mT/V [21], whereas the value estimated from Fig. 3 is about 1.05 T/V, about 50 times higher than in the VCMA case. We confirmed that $|\Delta\mu_0 H_{\text{sw}}/\Delta V_G|$ is increased below 270 K and reaches a maximum value of 4.2 T/V (see also Supplemental Material [41]). The first-principles calculation and this modulation efficiency of the switching field suggest that the VCMA effect has a minor role, and that the ME effect dominates the voltage effect on the switching field.

IV. SUMMARY

In summary, we showed voltage-induced bipolar switching of the Néel vector in a Pt/Cr₂O₃/Ir trilayer with a small gate voltage of approximately 0.8 V (51 mV/nm). It is notable that the switching voltage could be below 0.8 V, which is comparable to the driving voltage of modern CMOS devices. The dominant mechanism allowing bipolar and energy-efficient switching is the ME effect of the Cr₂O₃, which inherently avoids endurance and stability issues. The AFM spin switching has a great advantage for high-speed operation in such applications. We also found that the switching condition was beyond the simple expression of the ME pressure, especially in the high- H regime, which implies that rich physics for the ME AFM spin reversal is hidden. The nonequivalence of the effects of E and H on the AFM spin state is related to the distinct nature of magnetic and electric charges. As the energy-efficient isothermal deterministic switching

of magnetic states is of particular interest for ultralow-power memory and logic applications, our demonstration paves an avenue toward low-energy-consumption spin devices.

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The authors declare no nonfinancial interest but declare a competing financial interest. A patent application related to this research has been filed.

H.S. and K.U. prepared the samples, performed the structural characterizations, and conducted the AHE measurements with the help of N.M., K.T., and R.N. H.S., N.M., and Y.S. analyzed the data with the help of K.T. and T.M. K.N. performed the density functional theory calculations. Y.S. and T.M. prepared the manuscript using input from all coauthors. All authors discussed the results and wrote the manuscript.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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- [1] J. C. Slonczewski, Current-driven excitation of magnetic multilayers, *J. Magn. Magn. Mater.* **159**, L1 (1996).
 - [2] L. Berger, Emission of spin waves by a magnetic multilayer traversed by a current, *Phys. Rev. B* **54**, 9353 (1996).
 - [3] D. C. Ralph and M. S. Stiles, Spin transfer torques, *J. Magn. Magn. Mater.* **320**, 1190 (2008).
 - [4] J. Sinova, S. O. Valenzuela, J. Wunderlich, C. H. Back, and T. Jungwirth, Spin Hall effects, *Rev. Mod. Phys.* **87**, 1213 (2015).
 - [5] I. M. Miron, T. Moore, H. Szabolcs, L. D. Buda-Prejbeanu, S. Auffret, B. Rodmacq, S. Pizzini, J. Vogel, M. Bonfim, A. Schuhl, and G. Gaudin, Fast current-induced domain-wall motion controlled by the Rashba effect, *Nat. Mater.* **10**, 419 (2011).
 - [6] A. Manchon, J. Železný, I. M. Miron, T. Jungwirth, J. Sinova, A. Thiaville, K. Garello, and P. Gambardella, Current-induced spin-orbit torques in ferromagnetic and antiferromagnetic systems, *Rev. Mod. Phys.* **91**, 035004 (2019).
 - [7] S. Wang, K. He, Y. Xu, Z. Li, J. Wang, C. Li, X. Dai, J. Du, Y.-L. Wang, R. Liu, X. Lu, Y. Xu, and L. He, Lower switching-current density in Ta/(Pt/X)*n*/Pt/Co/Ta (X = Ta, Mn, Cu, V, Zr, Bi; *n* = 3, 4) multilayers based on a domain-wall-depinning model, *Phys. Rev. Appl.* **22**, L021002 (2024).
 - [8] Q. Yang, D. Han, S. Zhao, J. Kang, F. Wang, S.-C. Lee, J. Lei, K.-J. Lee, B.-G. Park, and H. Yang, Field-free spin-orbit torque switching in ferromagnetic trilayers at sub-ns timescales, *Nat. Commun.* **15**, 1814 (2024).
 - [9] M. Weisheit, S. Fähler, A. Marty, Y. Souche, C. Poinsignon, and D. Givord, Electric field-induced modification of magnetism in thin-film ferromagnets, *Science* **315**, 349 (2007).
 - [10] T. Maruyama, Y. Shiota, T. Nozaki, K. Ohta, N. Toda, M. Mizuguchi, A. A. Tulapurkar, T. Shinjo, M. Shiraishi, M. Mizukami, Y. Ando, and Y. Suzuki, Large voltage-induced magnetic anisotropy change in a few atomic layers of iron, *Nat. Nanotechnol.* **4**, 158 (2009).
 - [11] D. N. Astrov, Magnetoelectric effect in chromium oxide, *J. Exp. Theor. Phys.* **37**, 881 (1959).
 - [12] G. T. Rado, Mechanism of the magnetoelectric effect in an antiferromagnet, *Phys. Rev. Lett.* **6**, 609 (1961).
 - [13] S. Miwa, M. Suzuki, M. Tsujikawa, T. Nozaki, T. Nakamura, M. Shirai, S. Yuasa, and Y. Suzuki, Perpendicular magnetic anisotropy and its electric-field-induced change at metal-dielectric interfaces, *J. Phys. D: Appl. Phys.* **52**, 063001 (2019).
 - [14] H. Onoda, T. Nozaki, S. Tamaru, T. Nozaki, and S. Yuasa, Enhancing voltage-controlled magnetic anisotropy in Fe₈₀B₂₀/MgO/HfO₂ thin films by dielectric constant modulation, *Phys. Rev. Mater.* **6**, 104406 (2022).
 - [15] H. Nakayama, T. Nozaki, T. Nozaki, and S. Yuasa, Strong impact of underlayers on the voltage-controlled magnetic anisotropy in interface engineered Co/MgO junctions with heavy metals, *Adv. Mater. Interfaces* **10**, 2300131 (2023).
 - [16] L. Saravanan, N. Kumar Gupta, V. Mishra, C. Garcia, and S. Chaudhary, Impact of annealing on perpendicular magnetic anisotropy in W/MgAl₂O₄/CoFeMnSi/W/CoFeMnSi/MgAl₂O₄/W double storage layers for upcoming MTJs, *J. Magn. Magn. Mater.* **603**, 172225 (2024).
 - [17] T. Nozaki, T. Ichinose, T. Yamamoto, K. Yakushiji, and S. Yuasa, Improvement of voltage-controlled magnetic anisotropy effect by inserting an ultrathin metal capping layer, *APL Mater.* **12**, 091109 (2024).
 - [18] T. Kawabe, K. Yoshikawa, M. Tsujikawa, T. Tsukahara, K. Nawaoka, Y. Kotani, K. Toyoki, M. Goto, M. Suzuki, T. Nakamura, M. Shirai, Y. Suzuki, and S. Miwa, Electric-field-induced changes of magnetic moments and magnetocrystalline anisotropy in ultrathin cobalt films, *Phys. Rev. B* **96**, 220412(R) (2017).
 - [19] S. Miwa, M. Suzuki, M. Tsujikawa, K. Matsuda, T. Nozaki, K. Tanaka, T. Tsukahara, K. Nawaoka, M. Goto, Y. Kotani, T. Ohkubo, F. Bonell, E. Tamura, K. Hono, T. Nakamura, M. Shirai, S. Yuasa, and Y. Suzuki, Voltage controlled interfacial magnetism through platinum orbits, *Nat. Commun.* **8**, 15848 (2017).

- [20] Y. Shiota, T. Nozaki, F. Bonell, S. Murakami, T. Shinjo, and Y. Suzuki, Induction of coherent magnetization switching in a few atomic layers of FeCo using voltage pulses, *Nat. Mater.* **11**, 39 (2012).
- [21] T. Yamamoto, T. Ichinose, T. Nozaki, S. Tamaru, K. Yakushiji, H. Kubota, and S. Yuasa, Write-error reduction in voltage-driven magnetization switching using a recording layer with low magnetic damping, *Phys. Rev. Appl.* **21**, 054008 (2024).
- [22] P. Borisov, A. Hochstrat, X. Chen, W. Kleemann, and C. Binek, Magnetoelectric switching of exchange bias, *Phys. Rev. Lett.* **94**, 117203 (2005).
- [23] X. He, Y. Wang, N. Wu, A. N. Caruso, E. Vescovo, K. D. Belashchenko, P. A. Dowben, and C. Binek, Robust isothermal electric control of exchange bias at room temperature, *Nat. Mater.* **9**, 579 (2010).
- [24] A. Iyama and T. Kimura, Magnetoelectric hysteresis loops in Cr_2O_3 at room temperature, *Phys. Rev. B* **87**, 180408(R) (2013).
- [25] K. Toyoki, Y. Shiratsuchi, T. Nakamura, C. Mitsumata, S. Harimoto, Y. Takechi, T. Nishimura, H. Nomura, and R. Nakatani, Equilibrium surface magnetization of α - Cr_2O_3 studied through interfacial chromium magnetization in Co/α - Cr_2O_3 layered structures, *Appl. Phys. Express* **7**, 114201 (2014).
- [26] T. Ashida, M. Oida, N. Shimomura, T. Nozaki, T. Shibata, and M. Sahashi, Observation of magnetoelectric effect in $\text{Cr}_2\text{O}_3/\text{Pt}/\text{Co}$ thin film system, *Appl. Phys. Lett.* **104**, 152409 (2014).
- [27] Y. Shiratsuchi, K. Toyoki, and R. Nakatani, Magnetoelectric control of antiferromagnetic domain state in Cr_2O_3 thin film, *J. Phys.: Condens. Mater.* **33**, 243001 (2021).
- [28] K. Ujimoto, H. Sameshima, K. Toyoki, T. Moriyama, K. Nakamura, Y. Kotani, M. Suzuki, I. Iino, N. Kawamura, R. Nakatani, and Y. Shiratsuchi, Giant gate modulation of antiferromagnetic spin reversal by the magnetoelectric effect, *NPG Asia Mater.* **16**, 20 (2024).
- [29] V. Baltz, A. Manchon, M. Tsoi, T. Moriyama, T. Ono, and Y. Tserkovnyak, Antiferromagnetic spintronics, *Rev. Mod. Phys.* **90**, 015005 (2018).
- [30] R. Ramesh and N. A. Spaldin, Multiferroics: Progress and prospects in thin films, *Nat. Mater.* **6**, 21 (2007).
- [31] T. Arima, Spin-driven ferroelectricity and magneto-electric effects in frustrated magnetic systems, *J. Phys. Soc. Jpn.* **80**, 052001 (2011).
- [32] J. Wang, J. B. Neaton, J. Zheng, V. Nagarajan, S. B. Ogale, B. Liu, D. Viehland, V. Vaithyanathan, D. G. Schlom, U. V. Waghmare, N. A. Spaldin, K. M. Rabe, H. Wuttig, and R. Ramesh, Epitaxial BiFeO_3 multiferroic thin film heterostructures, *Science* **299**, 1719 (2003).
- [33] K. D. Belashchenko, Equilibrium magnetization at the boundary of a magnetoelectric antiferromagnet, *Phys. Rev. Lett.* **105**, 147204 (2010).
- [34] A. F. Andreev, Macroscopic magnetic fields of antiferromagnets, *JETP Lett.* **63**, 758 (1996).
- [35] T. Kosub, M. Kopte, F. Radu, O. G. Schmidt, and D. Makarov, All-electric access to the magnetic-field-invariant magnetization of antiferromagnets, *Phys. Rev. Lett.* **115**, 097201 (2015).
- [36] J.-L. Wang, W. Echtenkamp, A. Mahmood, and C. Binek, Voltage controlled magnetism in Cr_2O_3 based all-thin-film systems, *J. Magn. Magn. Mater.* **486**, 165262 (2019).
- [37] X. Wang, J. Ujimoto, K. Toyoki, R. Nakatani, and Y. Shiratsuchi, Increase of Néel temperature of magnetoelectric Cr_2O_3 thin film by epitaxial lattice matching, *Appl. Phys. Lett.* **121**, 182402 (2022).
- [38] W.-C. Liao, H. Liu, W. Tan, J. Keagy, J.-M. Chen, and J. Shi, Electrical detection of single-domain Néel vector reorientation across the spin-flop transition in Cr_2O_3 crystals, *Nano Lett.* **25**, 9794 (2025).
- [39] T. Iino, T. Moriyama, H. Iwaki, H. Aono, Y. Shiratsuchi, and T. Ono, Resistive detection of the Néel temperature of Cr_2O_3 thin films, *Appl. Phys. Lett.* **114**, 022402 (2019).
- [40] K. Ujimoto, H. Sameshima, K. Toyoki, Y. Kotani, T. Moriyama, K. Nakamura, R. Nakatani, and Y. Shiratsuchi, Direct observation of antiferromagnetic domains and field-induced reversal in $\text{Pt}/\text{Cr}_2\text{O}_3/\text{Pt}$ epitaxial trilayers, *Appl. Phys. Lett.* **123**, 022407 (2023).
- [41] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/2k8x-pryw> for detailed structural information, characterization of the Néel temperature based on the AHE measurements, the angular dependence of the switching field, the temperature dependence of the switching field and the modulation efficiency, the effect of a high magnetic field on the ME switching behavior at 270 K, and the method for the first-principles calculations. The Supplemental Material also includes Refs. [21,35,37,40,49].
- [42] Y. Cheng, S. Yu, M. Zhu, J. Hwang, and F. Yang, Evidence of the topological Hall effect in $\text{Pt}/\text{antiferromagnetic insulator bilayers}$, *Phys. Rev. Lett.* **123**, 237206 (2019).
- [43] T. Moriyama, Y. Shiratsuchi, T. Iino, H. Aono, M. Suzuki, T. Nakamura, Y. Kotani, R. Nakatani, K. Nakamura, and T. Ono, Giant anomalous Hall conductivity at the $\text{Pt}/\text{Cr}_2\text{O}_3$ interface, *Phys. Rev. Appl.* **13**, 034052 (2020).
- [44] T. V. A. Nguyen, Y. Shiratsuchi, A. Kobane, S. Yoshida, and R. Nakatani, Magnetic field dependence of threshold electric field for magnetoelectric switching of exchange-bias polarity, *J. Appl. Phys.* **122**, 073905 (2017).
- [45] T. Kimura, T. Goto, H. Shintani, K. Ishizaka, T. Arima, and Y. Tokura, Magnetic control of ferroelectric polarization, *Nature* **426**, 55 (2003).
- [46] K. Taniguchi, N. Abe, T. Takenobu, Y. Iwasa, and T. Arima, Ferroelectric polarization flop in a frustrated magnet MnWO_4 induced by a magnetic field, *Phys. Rev. Lett.* **97**, 097203 (2006).
- [47] H. Wang, T. Li, Y. Feng, X. Shi, X. Dong, M. Yang, C. Lu, and J. Wang, High magnetic field phase diagrams of the multiferroic manganite YMn_2O_5 single crystal, *Phys. Rev. B* **110**, 014430 (2024).
- [48] J. Ohtani and K. Kohn, Magnetoelectric effect and spin direction in a spin flopped Cr_2O_3 single crystal, *J. Phys. Soc. Jpn.* **53**, 3744 (1984).
- [49] R. D. Yacovitch and Y. Shapira, Magnetostriction near the magnetic-field-induced phase transitions of Cr_2O_3 , *Phys. B + C* **86-88**, 1126 (1977).