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# Giant magnetoelectric effect in an $L_{21}$ -ordered $\text{Co}_2\text{FeSi}/\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$ multiferroic heterostructure

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## ABSTRACT

We experimentally show a giant magnetoelectric (ME) effect at room temperature in an interfacial multiferroic heterostructure consisting of  $L_{21}$ -ordered  $\text{Co}_2\text{FeSi}$  and  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$  (PMN-PT). Molecular beam epitaxy growth at 400 °C enables us to obtain epitaxial and  $L_{21}$ -ordered  $\text{Co}_2\text{FeSi}$  films on PMN-PT(001). For the epitaxial  $\text{Co}_2\text{FeSi}/\text{PMN-PT}$  heterostructure, the remanent magnetization state can be largely modulated by varying electric fields. We note that the room-temperature ME coupling coefficient ( $\alpha$ ) is estimated to be  $6.0\text{--}6.3 \times 10^{-6}$  s/m, comparable to the highest  $\alpha$  value reported previously. Nonvolatile and repeatable magnetization changes in remanent states are also demonstrated. These results will pave the way for room-temperature electric-field control of the magnetization of half-metallic Heusler alloys in high-performance spintronic devices.

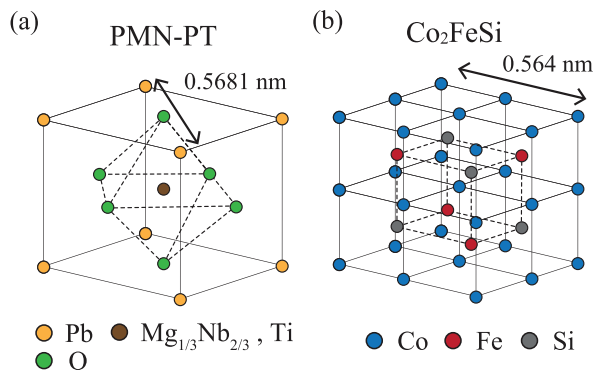
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As a next-generation memory for semiconductor integrated circuits with low power consumption, the magnetoresistive random access memory (MRAM) has been studied because of its high-speed, repeatable, and nonvolatile operation.<sup>1–3</sup> In general, writing data through the magnetization switching processes by applying electric currents consumes a large electric power in spin-transfer torque or spin-orbit torque MRAMs. To tackle the problem in these MRAMs, the magnetization switching induced by applying electric fields has been proposed, in which less power consumption is expected.<sup>4</sup>

To date, the electric field magnetization manipulation has been achieved in several special systems such as multiferroic insulator oxides,<sup>5,6</sup> ferromagnetic (FM) semiconductors,<sup>7,8</sup> and ultra-thin FM atomic layers.<sup>9,10</sup> In addition to these systems, artificial FM/ferroelectric (FE) multiferroic heterostructures have recently been explored because of potential advantages for room-temperature operation and a wide variety of material selections.<sup>11–14</sup> For the electric field magnetization manipulation in FM/FE multiferroic heterostructures, several mechanisms via magnetostriction, exchange bias, charge accumulation, and redox have been proposed.<sup>11</sup> Among

these mechanisms, the magnetostriction effect is particularly important to effectively modulate the magnetization states in the FM layer through the piezostains induced in the FE layer. Also, the magnetostriction from the FE layer is a long-range scale effect and induces a relatively large modulation of the magnetization state in the FM layer.<sup>11,12</sup>

For these reasons, an FE material,  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$  (PMN-PT), having a large piezoelectric constant ( $d_{33} \sim 2000$  pm/V),<sup>15,16</sup> has been explored, where a schematic for the crystal structure of PMN-PT is illustrated in Fig. 1(a). Actually, PMN-PT has been widely studied in FM/FE multiferroic heterostructures with a combination of many FM materials:  $(\text{La,Sr})\text{MnO}_3$ ,<sup>17,18</sup>  $\text{Fe}_3\text{O}_4$ ,<sup>19</sup>  $\text{FeGa}$ ,<sup>20</sup>  $\text{CoFeB}$ ,<sup>21,22</sup> and common ferromagnetic materials such as Fe,<sup>23</sup> Co,<sup>24,25</sup> Ni,<sup>26</sup> and their alloys.<sup>27–29</sup> Among a variety of FM materials, Co-based Heusler alloys have attracted interest from the viewpoint of MRAM applications because they have high spin polarization leading a high magnetoresistance ratio in tunnel magnetoresistance junctions.<sup>30–33</sup> However, only the experimental study of a  $\text{Co}_2\text{FeAl}$  (CFA)/PMN-PT multiferroic heterostructure has been reported because of



**FIG. 1.** Schematic crystal structures of (a)  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$  and (b)  $\text{Co}_2\text{FeSi}$ .

the growth difficulty.<sup>34,35</sup> In fact, although the formation of the  $L_{21}$ -ordered structure is required for the high-performance operation, it is limited that the reported CFA/PMN-PT was composed of  $B2$ -ordered or polycrystalline CFA films. As a result, the large magnetoelectric (ME) effect in the CFA/PMN-PT heterostructure at room temperature has not been reported yet.

Here, we concentrate on  $\text{Co}_2\text{FeSi}$  (CFS), which has been expected to be a half metal with a high Curie temperature of 1100 K.<sup>36</sup> A schematic for the crystal structure of CFS is shown in Fig. 1(b). Notably, we have shown that the epitaxial and  $L_{21}$ -ordered CFS films can be experimentally grown on oxides<sup>37,38</sup> or semiconductor structures.<sup>39–41</sup> In this Letter, we experimentally demonstrate an epitaxial and  $L_{21}$ -ordered CFS/PMN-PT(001) multiferroic heterostructure using a molecular beam epitaxy (MBE) technique at 400 °C. For the epitaxial  $\text{Co}_2\text{FeSi}$ /PMN-PT heterostructure, the remanent magnetization state can be largely modulated by varying electric fields. The room-temperature ME coupling coefficient ( $\alpha$ ) is estimated to be  $(6.3 \pm 0.2) \times 10^{-6}$  s/m, comparable to the highest  $\alpha$  value reported previously.<sup>22</sup> In addition, nonvolatile and repeatable magnetization changes in remanent states are demonstrated. These results will pave the way for room-temperature electric-field control of the magnetization of half-metallic Heusler alloys in high-performance spintronic devices.

First, we explain the crystal structures of PMN-PT and CFS, shown in Figs. 1(a) and 1(b), respectively. PMN-PT has a perovskite structure, it can be considered as a pseudocubic, and the lattice constant of PMN-PT is 0.4017 nm with the 30% PT composition,<sup>16</sup> while CFS is a cubic and an  $L_{21}$  ordered structure with a lattice constant of 0.564 nm.<sup>42</sup> Thus, the mismatch between the diagonal length of the lattice constant of PMN-PT and the lattice constant of CFS is approximately 0.7%. As a result, the epitaxial growth of CFS films on PMN-PT(001) can be expected.

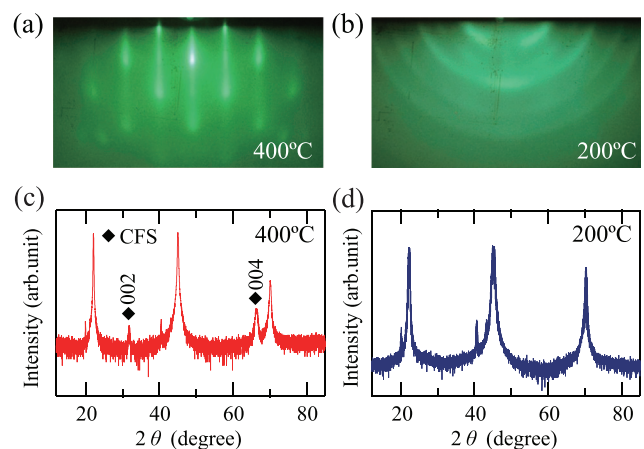
For the CFS/PMN-PT heterostructure, we next explain the growth procedure of CFS films on PMN-PT(001). 30-nm-thick CFS thin films were grown on single-crystal PMN-PT(001) substrates at 200 or 400 °C by MBE. Prior to the growth, the heat treatment was performed at 600 °C for 15 min to obtain a flat surface of PMN-PT(001). After cooling down to the growth temperature of 200 or 400 °C, the CFS film was grown by co-evaporating using Knudsen cells, where we set the supplied atomic composition ratio of Co:Fe:Si

to 2.0:1.0:1.0 during the growth.<sup>37,39</sup> After the growth, *in situ* reflection high-energy electron diffraction (RHEED) images are observed, as shown in Figs. 2(a) and 2(b). For the CFS film grown at 400 °C, symmetrical streak patterns are observed in the RHEED image [Fig. 2(a)], indicating two-dimensional epitaxial growth. In contrast, the CFS film grown at 200 °C shows the ring-like RHEED patterns [Fig. 2(b)], indicating the formation of polycrystalline films.  $\theta$ -2 $\theta$  x-ray diffraction (XRD) patterns for the CFS films grown at 400 and 200 °C are shown in Figs. 2(c) and 2(d), respectively. Only for the CFS film grown at 400 °C, evident 004 and 002 diffraction peaks are observed, indicating that the film is (001)-oriented epilayers. The out-of-plane lattice constant of the CFS film grown at 400 °C is 0.563 nm, slightly smaller than the bulk value of 0.564 nm.<sup>42</sup> Here, energy dispersive x-ray spectroscopy (EDX) measurements reveal that the grown CFS film has a slightly Co-rich composition (Co:Fe:Si = 2.27:0.91:0.82). We infer that the slight difference in the lattice constant between the grown film and bulk is due to the compositional deviation and/or the in-plane tensile strain from the PMN-PT(001) substrate. From these results, we can form the two-different CFS/PMN-PT heterostructures (epitaxial or polycrystalline) by varying the growth temperatures.

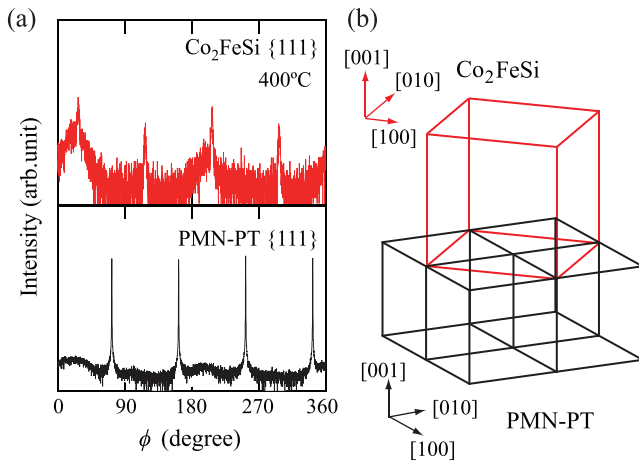
Figure 3(a) shows the {111} diffraction peaks of the 400 °C-grown CFS film (top) and the used PMN-PT(001) substrate (bottom) in XRD  $\phi$ -scan measurements. The fourfold {111} peaks in the top of Fig. 3(a) indicate the presence of the  $L_{21}$  structures in the 400 °C-grown CFS film. By a comparison of the {111} peaks between the CFS film and PMN-PT(001), we conclude the epitaxial relationship,  $\text{CFS}[100](001) \parallel \text{PMN-PT}[110](001)$  as shown in Fig. 3(b). Using the results in such XRD measurements, we estimate the degree of  $B2$  and  $L_{21}$  order parameters,  $S_{B2}$  and  $S_{L_{21}}$ , respectively, only for the 400 °C-grown CFS film. The values of  $S_{B2}$  and  $S_{L_{21}}$  are expressed as follows:<sup>43,44</sup>

$$S_{B2} = \sqrt{\frac{I_{002}/I_{004}}{I_{002}^R/I_{004}^R}}, \quad S_{L_{21}} = \frac{2}{3 - S_{B2}} \sqrt{\frac{I_{111}/I_{202}}{I_{111}^R/I_{202}^R}}, \quad (1)$$

where  $I_{hkl}$  and  $I_{hkl}^R$  are the experimental and theoretical diffraction peak intensities for the  $hkl$  plane. As a result, the values of  $S_{B2}$  and  $S_{L_{21}}$ ,



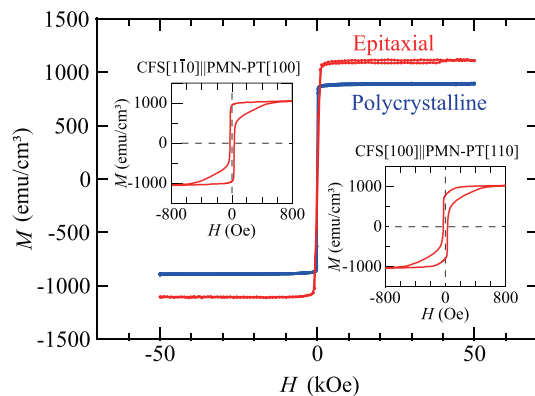
**FIG. 2.** RHEED images for the  $\text{Co}_2\text{FeSi}$  films grown at (a) 400 °C and (b) 200 °C with the incident electron beam along the PMN-PT[110] direction. XRD profiles in  $\theta$ -2 $\theta$  scan for  $\text{Co}_2\text{FeSi}$  films grown at (c) 400 °C and (d) 200 °C.



**FIG. 3.** (a) XRD  $\phi$ -scan profiles for {111} of the  $\text{Co}_2\text{FeSi}$  film grown at 400 °C (top) and the PMN-PT(001) substrate (bottom). (b) Schematic illustration of the epitaxial relationship between the  $\text{Co}_2\text{FeSi}$  film grown at 400 °C and the PMN-PT(001) substrate. The red and black arrows denote the crystallographic orientations for the  $\text{Co}_2\text{FeSi}$  film and the PMN-PT substrate, respectively.

are estimated to be 0.94 and 0.80, respectively. According to EDX measurements, since it was revealed that there is a slight deviation of the atomic compositions in CFS films, we should consider the presence of the structural disorder as a reason for  $S_{B2}$ ,  $S_{L2_1} < 1.0$  even in the epitaxial CFS/PMN-PT(001) heterostructure. However, we can regard  $L2_1$ -ordered structures as the dominant phase in the present CFS/PMN-PT(001) heterostructure.

Magnetic properties of the CFS/PMN-PT(001) heterostructures are examined using a vibrating sample magnetometer (VSM) at room temperature (27 °C). Figure 4 shows in-plane magnetization curves for the CFS/PMN-PT(001) heterostructures formed at 400 °C (epitaxial) and at 200 °C (polycrystalline), where the direction of the applied external magnetic field ( $H$ ) is along PMN-PT[100]. For the epitaxial CFS/PMN-PT(001) heterostructures, the saturation magnetization

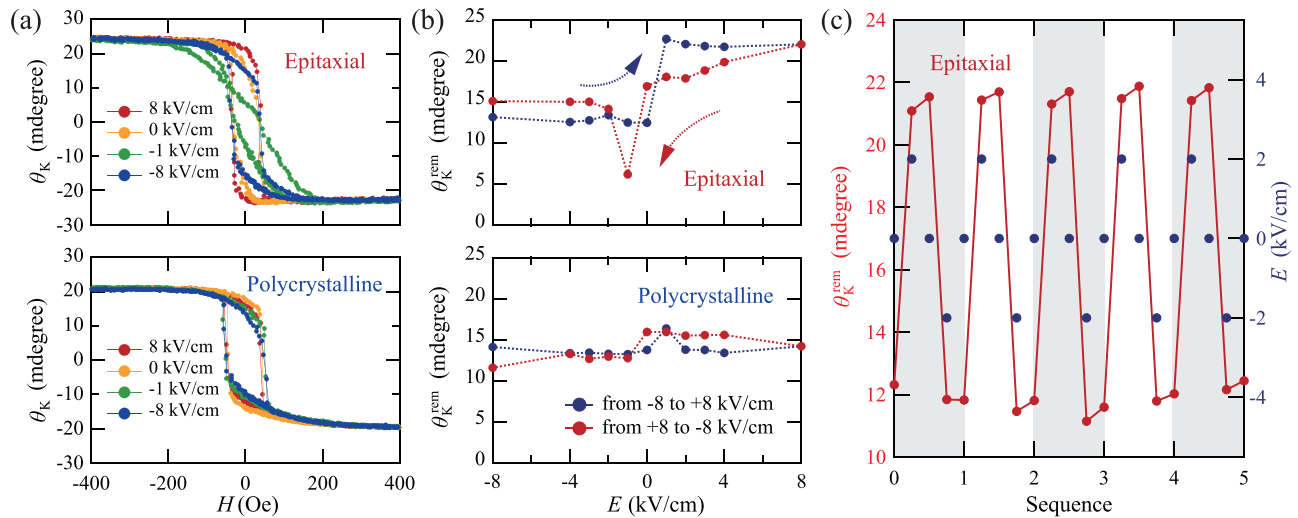


**FIG. 4.** Magnetization curves of the epitaxial (red) and polycrystalline (blue)  $\text{Co}_2\text{FeSi}$  films. The external magnetic field is applied along the PMN-PT[100] direction. The insets show expanded views of the magnetization curves of the epitaxial film under the magnetic fields along  $\text{Co}_2\text{FeSi}[110] \parallel \text{PMN-PT}[100]$  and  $\text{Co}_2\text{FeSi}[100] \parallel \text{PMN-PT}[110]$ . All the measurements were performed at 27 °C.

( $M_S$ ) is estimated to be  $1110 \pm 40 \text{ emu/cm}^3$  ( $5.4 \pm 0.2 \mu_B/\text{f.u.}$ ), close to  $M_S$  for the CFS epilayers with a high spin polarization of 0.7–0.8.<sup>45</sup> On the other hand, the value of  $M_S$  for the polycrystalline CFS/PMN-PT(001) heterostructure is  $890 \pm 30 \text{ emu/cm}^3$  ( $4.3 \pm 0.1 \mu_B/\text{f.u.}$ ), relatively low compared to that for the epitaxial CFS/PMN-PT(001) heterostructure. We infer that the above difference in  $M_S$  is derived from the difference in the crystal quality because there is almost no sharp diffraction peak in Fig. 2(d).<sup>46</sup> The insets of Fig. 4 show the enlarged magnetization curves for the epitaxial CFS/PMN-PT(001) heterostructure, measured under the magnetic fields along CFS[110]  $\parallel$  PMN-PT[100] (left) and CFS[100]  $\parallel$  PMN-PT[110] (right). The magnetization reversal behavior of the epitaxial CFS/PMN-PT(001) heterostructure is slightly changed by applying the different magnetic-field directions. In contrast, the polycrystalline CFS/PMN-PT(001) heterostructure showed an isotropic feature (not shown here). From these structural and magnetic characterization studies, we regard the epitaxial CFS/PMN-PT(001) heterostructure as a high-quality FM/FE multiferroic system.

To characterize the electric-field effect on magnetic properties for the CFS/PMN-PT(001) heterostructures, we perform magneto-optical Kerr effect measurements at room temperature with and without applying electric fields ( $E$ ). Here, Au(100 nm)/Ti(3 nm) backside electrodes are deposited on the bottom of the PMN-PT(001) substrate, which enables us to apply  $E$  to the CFS/PMN-PT(001) heterostructures along the PMN-PT[001] direction. Figure 5(a) shows room-temperature hysteresis loops of the Kerr rotation angle ( $\theta_K$ ) for the epitaxial (top) and polycrystalline (bottom) CFS/PMN-PT(001) heterostructures by applying  $E$ , where  $H$  is applied along the in-plane [100] direction of the PMN-PT substrate. Notably, we can clearly see that the value of the saturation Kerr-rotation angle ( $\theta_K^{\text{sat}}$ ) for the epitaxial CFS/PMN-PT(001) heterostructure is larger than that for the polycrystalline CFS one. In addition, the shape of the Kerr hysteresis loop for the epitaxial CFS/PMN-PT(001) heterostructure is markedly modulated by changing  $E$  from +8.0 to −8.0 kV/cm, while the shape of the Kerr loops is slightly varied by changing  $E$  for the polycrystalline CFS/PMN-PT(001) heterostructure.

To explore the electric field modulation in detail, we also measure the remanent Kerr rotation ( $\theta_K^{\text{rem}}$ ) vs  $E$  for both the CFS/PMN-PT(001) heterostructures, as shown in Fig. 5(b). For the measurement, we first apply an  $E$  value of −8 kV/cm and vary the amplitude of  $E$  from −8.0 to +8.0 kV/cm and then back to  $E = -8.0 \text{ kV/cm}$ . The sweep rate of  $E$  is 20 V/cm/s. For the epitaxial CFS/PMN-PT(001) heterostructure,  $\theta_K^{\text{rem}}$  sharply increases or decreases at  $E \sim \pm 1 \text{ kV/cm}$  upon increasing or decreasing  $E$  from −8.0 kV/cm or +8.0 kV/cm, respectively. We find that the value of  $E$  showing the abrupt change in  $\theta_K^{\text{rem}}$  is close to a reported coercive field of PMN-PT(001).<sup>47</sup> Also, the  $E$  dependence shows a hysteretic behavior, implying the nonvolatile variation in remanent magnetization states. In general, there are two types of piezoelectric polarization vector switchings along  $\langle 111 \rangle$  in the pseudocubic unit cell of PMN-PT,<sup>16,21,47</sup> in which one is the 109° polarization switching and the other is the 71/180° one. The former causes an in-plane strain variation rotated by 90° along  $\langle 110 \rangle$  in PMN-PT(001) although the latter does not induce the rotation of the strain.<sup>21,47</sup> Thus, for an application of  $E$  to PMN-PT[001], a hysteric feature seen in Fig. 5(b) is related to the 109° switching inducing the in-plane-strain rotation. In addition to this, since the value of  $\theta_K^{\text{sat}}$  for the epitaxial CFS/PMN-PT(001) heterostructure is nearly constant for all the Kerr



**FIG. 5.** (a) Magneto-optical Kerr loops for the epitaxial (top) and polycrystalline (bottom) CFS/PMN-PT(001) heterostructures at various  $E$  values at room temperature. The external magnetic field ( $H$ ) is applied along the PMN-PT[100] direction. (b)  $E$  dependence of the remanent Kerr rotation  $\theta_K^{\text{rem}}$  for the epitaxial (top) and polycrystalline (bottom) CFS/PMN-PT(001) heterostructures at room temperature. (c) Demonstration of the switching of the remanent Kerr rotation (red) for the epitaxial CFS/PMN-PT(001) heterostructure at room temperature. The magnitude of  $E$  is applied with the bipolar sequence (blue).

hysteresis loops in Fig. 5(a), we can conclude that the  $E$ -field-dependent  $\theta_K^{\text{rem}}$  originates from the modulation of the in-plane magnetic anisotropy induced by the in-plane-strain variation, as reported in many FM/PMN-PT systems.<sup>21,35,48,49</sup> Contrary to this, the polycrystalline CFS/PMN-PT(001) heterostructure does not show such a large variation in  $\theta_K^{\text{rem}}$  with increasing and decreasing  $E$ . Taking into account these facts, we can regard the large  $E$  effect on the remanent magnetization ( $M_{\text{rem}}$ ) as a consequence of the piezoelectric induced strain change at the epitaxial CFS/PMN-PT(001) interface.

To quantitatively evaluate the ME effect for the CFS/PMN-PT(001) heterostructures, we define the ME coefficient  $\alpha$  as follows:

$$\alpha = \mu_0 \frac{dM}{dE} = \mu_0 \frac{M_{\text{rem}}(E) - M_{\text{rem}}(E=0)}{E}, \quad (2)$$

where  $\mu_0$  is the vacuum permeability. Here, we define  $dM$  as the difference in the remanent magnetizations between before and after the application of  $E$ , where this method has been established in previous studies of FM/FE systems.<sup>21,22,50</sup> Note that  $dM$  in Eq. (2) is different from that for conventional ME substances such as  $\text{Cr}_2\text{O}_3$  for which  $dM$  is defined as the induction of spontaneous magnetization by  $E$ .<sup>51</sup> To indirectly deduce  $M_{\text{rem}}$ , we assume that the value of  $\theta_K^{\text{sat}}$  ( $\sim 23.9 \pm 0.1$  mdeg) corresponds to the value of  $M_s$  ( $1110 \pm 40$  emu/ $\text{cm}^3$ ) measured by VSM. On the basis of the above assumption,  $M_{\text{rem}}$  is estimated as  $M_{\text{rem}} = M_s \times (\theta_K^{\text{rem}} / \theta_K^{\text{sat}})$ . As a result, the  $\alpha$  values of  $(6.0 \pm 0.2) \times 10^{-6}$  s/m and  $(6.3 \pm 0.2) \times 10^{-6}$  s/m are obtained at  $E$  changing from 0 to +1 kV/cm and at  $E$  changing from 0 to -1 kV/cm, respectively, for the epitaxial CFS/PMN-PT(001) heterostructure. On the other hand, the polycrystalline CFS/PMN-PT(001) heterostructure gives smaller  $\alpha$  values of  $1.5\text{--}1.9 \times 10^{-6}$  s/m at the same  $E$  value. Notably, the  $\alpha$  values estimated here mean the magnitude of the strain-mediated ME effect at the formed CFS/PMN-PT heterointerfaces. Compared to the previous reports in such systems, the present values of  $\alpha$  for the epitaxial CFS/PMN-PT(001) heterostructure are comparable

to those of amorphous CoFeB/PMN-PT(011) ( $8 \times 10^{-6}$  s/m)<sup>22</sup> and CoFeB/PMN-PT(001) ( $2 \times 10^{-6}$  s/m),<sup>21</sup> the highest  $\alpha$  value in reported FM/FE interfacial multiferroic systems. In the case of the previous amorphous CoFeB,<sup>22</sup> the  $\alpha$  value increased upon changing the PMN-PT crystal orientations from (001) to (011) where the largest change in the piezostress was expected.<sup>29</sup> If we can form the epitaxial CFS/PMN-PT(011) heterostructure, further enhancement in the  $\alpha$  value can be expected.

Finally, repeatable switchings of the remanent magnetization states with nonvolatile nature are shown in Fig. 5(c), where the sequence of the  $E$  application to PMN-PT[001] is also displayed in the right vertical axis. Evident variations in two magnetic states with high/low  $\theta_K^{\text{rem}}$  can be repeatably demonstrated at room temperature. Because the magnetization vector switching does not need an application of  $H$ , the repeatable and nonvolatile magnetization control at the remanent states can be utilized as a part of the technology for voltage-induced magnetization switching in storage and/or memory devices with high-quality Co-based Heusler alloys.<sup>33,52–54</sup>

In summary, we demonstrated an epitaxial and  $L2_1$ -ordered  $\text{Co}_2\text{FeSi}/\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$  multiferroic heterostructure with giant  $\alpha$  values of  $6.0\text{--}6.3 \times 10^{-6}$  s/m. Nonvolatile and repeatable magnetization changes in remanent states were also achieved, meaning the utilization for a part of the technology for voltage-induced magnetization switching in storage and/or memory devices with high-performance Co-based Heusler alloys.

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## DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

## REFERENCES

- <sup>1</sup>A. D. Kent and D. C. Worledge, *Nat. Nanotechnol.* **10**, 187 (2015).
- <sup>2</sup>S. Bhatti, R. Sbiaa, A. Hirohata, H. Ohno, S. Fukami, and S. Piramanayagam, *Mater. Today* **20**, 530 (2017).
- <sup>3</sup>S. Yuasa and D. D. Djayaprawira, *J. Phys. D: Appl. Phys.* **40**, R337 (2007).
- <sup>4</sup>J.-M. Hu, Z. Li, L.-Q. Chen, and C.-W. Nan, *Nat. Commun.* **2**, 553 (2011).
- <sup>5</sup>H. Schmid, *Ferroelectrics* **162**, 317 (1994).
- <sup>6</sup>N. A. Spaldin and R. Ramesh, *Nat. Mater.* **18**, 203 (2019).
- <sup>7</sup>H. Ohno, D. Chiba, F. Matsukura, T. Omiya, E. Abe, T. Dietl, Y. Ohno, and K. Ohtani, *Nature* **408**, 944 (2000).
- <sup>8</sup>F. Matsukura, Y. Tokura, and H. Ohno, *Nat. Nanotechnol.* **10**, 209 (2015).
- <sup>9</sup>T. Maruyama, Y. Shiota, T. Nozaki, K. Ohta, N. Toda, M. Mizuguchi, A. A. Tulapurkar, T. Shinjo, M. Shiraishi, S. Mizukami, Y. Ando, and Y. Suzuki, *Nat. Nanotechnol.* **4**, 158 (2009).
- <sup>10</sup>B. Dieny and M. Chshiev, *Rev. Mod. Phys.* **89**, 025008 (2017).
- <sup>11</sup>J.-M. Hu and C.-W. Nan, *APL Mater.* **7**, 080905 (2019).
- <sup>12</sup>J.-M. Hu, C.-G. Duan, C.-W. Nan, and L.-Q. Chen, *npj Comput. Mater.* **3**, 18 (2017).
- <sup>13</sup>T. Taniyama, *J. Phys. Condens. Matter* **27**, 504001 (2015).
- <sup>14</sup>C. A. F. Vaz, *J. Phys. Condens. Matter* **24**, 333201 (2012).
- <sup>15</sup>Y. Guo, H. Luo, D. Ling, H. Xu, T. He, and Z. Yin, *J. Phys. Condens. Matter* **15**, L77 (2003).
- <sup>16</sup>B. Noheda, D. E. Cox, G. Shirane, J. Gao, and Z.-G. Ye, *Phys. Rev. B* **66**, 054104 (2002).
- <sup>17</sup>S. P. Pati and T. Taniyama, *J. Phys. D: Appl. Phys.* **53**, 054003 (2020).
- <sup>18</sup>C. Thiele, K. Dörr, S. Fähler, L. Schultz, D. C. Meyer, A. A. Levin, and P. Paufler, *Appl. Phys. Lett.* **87**, 262502 (2005).
- <sup>19</sup>M. Liu, J. Hoffman, J. Wang, J. Zhang, B. Nelson-Cheeseman, and A. Bhattacharya, *Sci. Rep.* **3**, 1876 (2013).
- <sup>20</sup>W. Jahjah, J.-P. Jay, Y. L. Grand, A. Fessant, A. Prinsloo, C. Sheppard, D. Dekadjevi, and D. Spenato, *Phys. Rev. Appl.* **13**, 034015 (2020).
- <sup>21</sup>S. Zhang, Y. G. Zhao, P. S. Li, J. J. Yang, S. Rizwan, J. X. Zhang, J. Seidel, T. L. Qu, Y. J. Yang, Z. L. Luo, Q. He, T. Zou, Q. P. Chen, J. W. Wang, L. F. Yang, Y. Sun, Y. Z. Wu, X. Xiao, X. F. Jin, J. Huang, C. Gao, X. F. Han, and R. Ramesh, *Phys. Rev. Lett.* **108**, 137203 (2012).
- <sup>22</sup>J. Wang, D. Pesquera, R. Mansell, S. van Dijken, R. P. Cowburn, M. Ghidini, and N. D. Mathur, *Appl. Phys. Lett.* **114**, 092401 (2019).
- <sup>23</sup>C. Zhang, F. Wang, C. Dong, C. Gao, C. Jia, C. Jiang, and D. Xue, *Nanoscale* **7**, 4187 (2015).
- <sup>24</sup>W. Zhou, C. Ma, Z. Gan, Z. Zhang, X. Wang, W. Tan, and D. Wang, *Appl. Phys. Lett.* **111**, 052401 (2017).
- <sup>25</sup>C. Zhou, C. Zhang, J. Yao, and C. Jiang, *Appl. Phys. Lett.* **109**, 112404 (2016).
- <sup>26</sup>T. Wu, A. Bur, P. Zhao, K. P. Mohanchandra, K. Wong, K. L. Wang, C. S. Lynch, and G. P. Carman, *Appl. Phys. Lett.* **98**, 012504 (2011).
- <sup>27</sup>X. Han, L. Xi, Y. Li, X. Guo, D. Li, Z. Wang, Y. Zuo, and D. Xue, *Appl. Phys. Lett.* **105**, 122402 (2014).
- <sup>28</sup>M. Liu, W. Du, H. Su, B. Liu, H. Meng, and X. Tang, *Appl. Phys. Lett.* **116**, 152401 (2020).
- <sup>29</sup>W. Du, M. Liu, H. Su, H. Zhang, B. Liu, H. Meng, G. Xu, R. Peng, and X. Tang, *Appl. Phys. Lett.* **117**, 222401 (2020).
- <sup>30</sup>S. Trudel, O. Gaier, J. Hamrle, and B. Hillebrands, *J. Phys. D: Appl. Phys.* **43**, 193001 (2010).
- <sup>31</sup>Y. Sakuraba, M. Hattori, M. Oogane, Y. Ando, H. Kato, A. Sakuma, T. Miyazaki, and H. Kubota, *Appl. Phys. Lett.* **88**, 192508 (2006).
- <sup>32</sup>W. Wang, H. Sukegawa, R. Shan, S. Mitani, and K. Inomata, *Appl. Phys. Lett.* **95**, 182502 (2009).
- <sup>33</sup>Z. Wen, H. Sukegawa, S. Mitani, and K. Inomata, *Appl. Phys. Lett.* **98**, 192505 (2011).
- <sup>34</sup>G. Dunzhu, F. Liu, Y. Wang, C. Zhou, and C. Jiang, *Mater. Res. Express* **6**, 066114 (2019).
- <sup>35</sup>C. Zhou, G. Dunzhu, J. Yao, and C. Jiang, *J. Alloys Compd.* **710**, 680 (2017).
- <sup>36</sup>S. Wurmehl, G. H. Fecher, H. C. Kandpal, V. Ksenofontov, C. Felser, H.-J. Lin, and J. Morais, *Phys. Rev. B* **72**, 184434 (2005).
- <sup>37</sup>K. Kudo, Y. Hamazaki, S. Yamada, S. Abo, Y. Gohda, and K. Hamaya, *ACS Appl. Electron. Mater.* **1**, 2371 (2019).
- <sup>38</sup>S. Yamada, Y. Teramoto, D. Matsumi, D. Kepaptsoglou, I. Azaceta, T. Murata, K. Kudo, V. K. Lazarov, T. Taniyama, and K. Hamaya, *Phys. Rev. Mater.* **5**, 014412 (2021).
- <sup>39</sup>S. Yamada, K. Hamaya, K. Yamamoto, T. Murakami, K. Mibu, and M. Miyao, *Appl. Phys. Lett.* **96**, 082511 (2010).
- <sup>40</sup>K. Kasahara, K. Yamamoto, S. Yamada, T. Murakami, K. Hamaya, K. Mibu, and M. Miyao, *J. Appl. Phys.* **107**, 09B105 (2010).
- <sup>41</sup>K. Kasahara, Y. Fujita, S. Yamada, K. Sawano, M. Miyao, and K. Hamaya, *Appl. Phys. Express* **7**, 033002 (2014).
- <sup>42</sup>B. Balke, G. H. Fecher, H. C. Kandpal, C. Felser, K. Kobayashi, E. Ikenaga, J.-J. Kim, and S. Ueda, *Phys. Rev. B* **74**, 104405 (2006).
- <sup>43</sup>Y. Takamura, R. Nakane, and S. Sugahara, *J. Appl. Phys.* **105**, 07B109 (2009).
- <sup>44</sup>Y. Takamura, T. Suzuki, Y. Fujino, and S. Nakagawa, *J. Appl. Phys.* **115**, 17C732 (2014).
- <sup>45</sup>K. Hamaya, N. Hashimoto, S. Oki, S. Yamada, M. Miyao, and T. Kimura, *Phys. Rev. B* **85**, 100404(R) (2012).
- <sup>46</sup>Z. Gercsi and K. Hono, *J. Phys. Condens. Matter* **19**, 326216 (2007).
- <sup>47</sup>L. Yang, Y. Zhao, S. Zhang, P. Li, Y. Gao, Y. Yang, H. Huang, P. Miao, Y. Liu, A. Chen, C. W. Nan, and C. Gao, *Sci. Rep.* **4**, 4591 (2014).
- <sup>48</sup>J. J. Yang, Y. G. Zhao, H. F. Tian, L. B. Luo, H. Y. Zhang, Y. J. He, and H. S. Luo, *Appl. Phys. Lett.* **94**, 212504 (2009).
- <sup>49</sup>S. Zhang, Q. Chen, Y. Liu, A. Chen, L. Yang, P. Li, Z. S. Ming, Y. Yu, W. Sun, X. Zhang, Y. Zhao, Y. Sun, and Y. Zhao, *ACS Appl. Mater. Interfaces* **9**, 20637 (2017).
- <sup>50</sup>Y. Wei, C. Gao, Z. Chen, S. Xi, W. Shao, P. Zhang, G. Chen, and J. Li, *Sci. Rep.* **6**, 30002 (2016).
- <sup>51</sup>M. Fiebig, *J. Phys. D: Appl. Phys.* **38**, R123 (2005).
- <sup>52</sup>T. M. Nakatani, T. Furubayashi, S. Kasai, H. Sukegawa, Y. K. Takahashi, S. Mitani, and K. Hono, *Appl. Phys. Lett.* **96**, 212501 (2010).
- <sup>53</sup>S. Li, Y. Sakuraba, T. Sasaki, J. Chen, S. Bosu, and K. Hono, *AIP Adv.* **8**, 075230 (2018).
- <sup>54</sup>M. S. Gabor, T. Petrisor, M. Nasui, M. A. Nsibi, J. Nath, and I. M. Miron, *Phys. Rev. Appl.* **13**, 054039 (2020).