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Large converse magnetoelectric coupling in FeCoV/lead zinc niobate-lead titanate heterostructure
Strain-mediated converse magnetoelectric coupling strength manipulation by a thin titanium layer

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The manipulation of the strain-mediated magnetoelectric (ME) coupling strength is investigated by inserting a thin Ti layer (0–10 nm) between a 50 nm Co$_{50}$Fe$_{50}$ layer and a (011) oriented lead magnesium niobate-lead titanate (PMN-PT) substrate. A record high remanence ratio ($M_r/M_s$) tunability of 100% has been demonstrated in the 50 nm CoFe/8 nm Ti/PMN-PT heterostructure, when a total in-plane piezoelectric strain of $-1821$ ppm was applied at an electric field (E-field) of 16 kV/cm. The ME coupling strength is gradually optimized as the Ti layer thickness increases. Magnetic energy calculation showed that with increasing Ti layer thickness the uniaxial magnetic anisotropy energy ($E_{uni}$) was reduced from $43 \pm 1$ kJ/m$^3$ to $29.8 \pm 1$ kJ/m$^3$. The reduction of $E_{uni}$ makes the strain effect dominant in the total magnetic energy, thus gives an obvious enhanced ME coupling strength. © 2016 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4939111]

Magnetoelectric (ME) coupling in multiferroic (MF) composites has drawn intensive interest due to their important potential applications such as low-power spintronic devices. In recent years, many papers have reported the obvious strain-mediated converse ME coupling in a wide range of the MF layered heterostructure. Both magnetization and magnetic anisotropy of the magnetic film can be manipulated by an external electric field (E-field) induced piezoelectric strain, which provides great opportunities for E-field controlling magnetic devices. However, the manipulation of ME coupling strength is much less investigated. Kim et al. studied the manipulation of ME coupling by varying the composition and thickness of the Co$_x$Pd$_{1-x}$ film. A large ME coupling strength was found for 10 nm Co$_{0.25}$Pd$_{0.75}$ film due to a high magnetostriction. The effect of the underlayer on magnetic thin films has been widely studied in the past two decades. The reason can be attributed to the thin underlayer leading a change of the texture and the stress in the magnetic thin films. Jung and Doyle found that a thin Cu underlayer changed the preferred orientation from (200) to (110) in the CoFe film and thus improved the magnetic softness. In our work, the effect of the introduction of a thin Ti layer on the converse ME coupling strength was carried out. A record high remanence ratio ($M_r/M_s$) tunability of 100% has been demonstrated in the 50 nm CoFe/8 nm Ti/PMN-PT heterostructure. In addition, our previous results have demonstrated a promising ME coupling effect in the Co$_{50}$Fe$_{50}$ (CoFe)/(011) PMN-PT heterostructure. 50 nm thick CoFe/8 nm Ti films ($x = 0, 2, 4, 6, 8, 10$) were RF sputtered on the (011) oriented PMN-30%PT substrate with mean surface roughness $1.31 \pm 0.08$ nm at room temperature. The polished substrate was cleaned prior to use with n-Butyl acetate, acetone, and IPA. The sputtering power, working pressure, and base pressure were 75 W, 5.0 $\pm$ 0.1 mTorr, and 1.2 $\pm$ 0.2 $\times$ 10$^{-7}$ mTorr, respectively.

An in-situ magnetic field (H-field) of 65.3 kA/m was applied along the [100] crystallographic direction of the PMN-PT substrate during the growth of the CoFe film for obtaining a maximum normalized remanence ($M_r/M_s$) ratio (~1) and thus maximum variation in $M_r/M_s$. The (011)PMN-PT (10 cm [01-1] $\times$ 5 cm [100] $\times$ 0.5 cm [011]) had anisotropic strains with in-plane piezoelectric coefficients of $d_{31} = -1200$ to $-1800$ pC/N reported by H.C. Materials Corporation. The piezoelectric constant $d_{33}$ was measured to be $400 \pm 10$ pC/N for all these substrates on a $d_{33}$ testing meter. The difference between all PMN-PT substrates used in this work was $\sim 2.5\%$. The polarization (P) and out of plane piezoelectric strain ($e_{33}$) of the (011) PMN-PT substrate as a function of the E-field were measured using triangular voltage waveforms by a ferroelectric hysteresis measurement tester (National Physics Laboratories, UK). The piezoelectric strain was converted to a voltage signal by a pair of NXB2-AL nanosensors and sent to a digital voltmeter for acquiring data. When the overall volume of the substrate is kept constant, the in-plane piezoelectric strain ($e_{xy}$) is $-e_{33}$. Magneto-Optical Kerr Effect (MOKE) measurements were used to investigate the magnetic properties when a large range dc E-fields (0–17 kV/cm) were applied to the MF heterostructures. The MOKE H-field was applied along the [100] or [01-1] direction of the PMN-PT, while the E-field was applied along [011] direction of the PMN-PT. More detailed measurement methods are found in Refs. 14 and 15. The CoFe film and the bottom Au on the PMN-PT surface were used as the electrodes. To test the CoFe electrode, the electric capacity was measured to be about 3000 pF using an impedance analyzer, which is comparable with a sample with two Au electrodes. The effective saturation magnetostriiction constants ($\chi_{eff}$) were measured using a technique based on the Villari effect. To investigate the microstructure of the heterostructure, X-ray diffraction (XRD) measurements were made on the samples using a Siemens D5000 X-ray diffractometer with CuK$_\alpha$ radiation.

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Cross-sectional transmission electron microscopy (TEM) specimens were prepared by Focused Ion Beam (FIB) in FEI Quanta 200 3D. The fabricated TEM specimens were observed on a JEOL R005 operating at 300 kV.

Figure 1(a) shows the XRD patterns of the 50 nm CoFe/x nm Ti/PMN-PT heterostructures at a scan speed of 0.23°/min and 65 nm CoFe/PMN-PT heterostructures at a scan speed of 0.5°/min. It shows strong (011) and (022) diffraction peaks for the PMN-PT substrate. The vertical lines indicate the 2θ position of the standard diffraction peaks (110) and (002) (cited from PDF#04-003-5514) for the CoFe film. The weak (110) diffraction peak of CoFe film at 2θ = 44.9° is only seen on a smaller intensity scale (inset), while the (002) diffraction peak at 2θ = 65.3° overlaps with the strong PMN-PT (022) peak at 2θ = 65.5° and thus it is hard to be observed. The (110) peak of CoFe is clearly observed for 50 nm CoFe without Ti; however, the peak disappears when the Ti layer was added, with all data being taken with the same scan conditions. This suggests that the Ti layer promotes random orientation of the CoFe grains, which is helpful to reduce the magnetocrystalline anisotropy energy contribution to total magnetic energy and thus to make the magnetoelastic energy dominant. In Fig. 1(c), the High Angle Annular Dark Field (HAADF) image shows a columns growth structure of CoFe film and an average columnar grain size of 3.6 ± 0.5 nm in the CoFe/8 nm Ti/PMN-PT heterostructure, which is almost same as that of 3.3 ± 0.5 nm in the CoFe/PMN-PT heterostructure. Selected area electron diffraction (SAED) pattern shows the polycrystalline structure of CoFe film. However, the crystalline plane of (110) in CoFe film for the CoFe/8 nm Ti/PMN-PT heterostructure shows weaker diffraction than that for the CoFe/PMN-PT heterostructure, which is consistent with XRD results of CoFe film.

Figure 1(b) shows the P and ε of (011) PMN-PT substrate as a function of the E-field. The ferroelectric and piezoelectric properties were studied at an alternating E-field with a maximum amplitude of 16 kV/cm and a frequency of 0.5 Hz. The maximum in-plane compressive strain ε_x+ε_y is ~1821 ppm at 16 kV/cm. The ferroelectric coercive field is ~3.5 kV/cm and slightly higher compared with ~2 kV/cm reported previously, which results in the larger operational E-field up to around 16 kV/cm compared with the reported 10 kV/cm for PMN-PT single crystal.

Figures 2(a) and 2(b) show the strong E-field dependence of the hysteresis loops measured along [01-1] and [100] in 50 nm CoFe/8 nm Ti/PMN-PT heterostructure, respectively. For the other samples, a strong E-field dependence of the hysteresis loops was also observed at the higher applied E-field, but is not shown in this paper. As we can see, the magnetization process along [01-1] becomes easy, while that along [100] becomes hard with the applied E-field. The results can be qualitatively analysed by the strain induced domain wall motion. The magnetoelastic energy can be described as.

![Figure 1](image1.png)

![Figure 2](image2.png)

FIG. 1. (a) XRD spectra of 50 nm CoFe/x nm Ti/PMN-PT and 65 nm CoFe/PMN-PT heterostructures. The inset shows an enlarged part around the (110) diffraction peak of the CoFe film. (b) Ferroelectric hysteresis loop and in-plane piezoelectric strain loop of (011) oriented PMN-PT. (c) The HAADF image of the CoFe/8 nm Ti/PMN-PT heterostructure.

FIG. 2. Electric dependence of magnetic hysteresis loops measured along [01-1] (a) and [100] (b) in 50 nm CoFe/8 nm Ti/PMN-PT heterostructure.
where $\lambda$ and $\sigma$ are magnetostrictive constant and stress, while $\theta$ is the angle between the saturation magnetisation ($M_s$) and $\sigma$. For minimizing the $E_{\text{stress}}$ in the system, the external E-field induces tensile strain (i.e., $\sigma > 0$) along $[01-1]$, causing more magnetic moments to align along this tensile stress direction due to the positive $\lambda$. When the H-field is applied along $[01-1]$, a lower H-field will be required to saturate the sample due to the relatively easy $180^\circ$ wall motion. In contrast, the compressive stress ($\sigma < 0$) along $[100]$ causes more magnetic moments to align perpendicular to $[100]$. Thus, when the H-field is applied along $[100]$, a higher H-field is required to rotate the magnetic moments $90^\circ$ into the $[100]$ direction. Here, the E-field induced stress field plays the same role to move the domain wall in the magnetization process as that of the H-field. However, the domain rotation has to be completed by the H-field. Furthermore, the stress field induced domain motion can make the domain rotation easier or harder by the H-field.

Figures 3(a) and 3(b) show the detailed variation in the $M_r/M_s$ along $[01-1]$ and $[100]$ at each applied E-field in these heterostructures. When the E-field is increased, the $M_r/M_s$ is increased along the $[01-1]$ direction and reduced along the $[100]$ direction in all the samples. As the E-field is increased further, the $M_r/M_s$ along $[01-1]$ is almost constant close to 1, while the $M_r/M_s$ along $[100]$ is decreased as the E-field is increased. At the E-field of 17 kV/cm, the maximum $M_r/M_s$ change along $[100]$ is 72% for the sample without Ti layer, while a larger $M_r/M_s$ change of 100% is reached at the E-field of 16 kV/cm for the sample with 8 nm Ti layer. This suggests that the addition of a Ti layer significantly improves the converse ME coupling strength.

The total magnetic energy ($E_{\text{total}}$) of the samples can be described as:

$$E_{\text{total}} = E_{\text{zeeman}} + E_{\text{shape}} + E_{\text{stress}} + E_{\text{charge}} + E_{\text{uni}},$$

where $E_{\text{zeeman}}$, $E_{\text{shape}}$, $E_{\text{stress}}$, $E_{\text{charge}}$, and $E_{\text{uni}}$ are the Zeeman energy, demagnetizing energy, magnetoelastic energy, charging induced surface anisotropy energy, and uniaxial magnetic anisotropy energy, respectively. In the present work, only the last three energy terms are changed. First, when the conductive Ti layer is introduced into the interface between the CoFe and the PMN-PT substrate, the charge accumulation is hindered on the interface due to the metallic short charge screening length. Therefore, $E_{\text{charge}}$ is reduced in the sample with the Ti layer. Next, the $E_{\text{uni}}$ can be calculated as: $E_{\text{uni}} = (1/2)H_kM_s$, where the anisotropy field $H_k$ is taken from the measured hysteresis loop at the $M_r/M_s = 1$ and shown in Fig. 3(d) and $M_s$ is 1900 emu/cm$^3$ taken from Ref. 24. As shown in Fig. 3(d), when the Ti thickness increases, the $E_{\text{uni}}$ is gradually reduced. For the sample without a Ti layer, the $E_{\text{uni}}$ is 43 $\pm$ 1 kJ/m$^3$, while the $E_{\text{uni}}$ is 29.8 $\pm$ 1 kJ/m$^3$ in the sample with 8 nm Ti. Therefore, the $E_{\text{uni}}$ is reduced by 13.2 kJ/m$^3$ in the 8 nm Ti sample. The anisotropy change also can be clearly observed in the inset of Fig. 3(a), which shows the $M_r/M_s$ measured along a series of angles for the 0 nm and 8 nm Ti samples.

The other contribution to the ME coupling strength change by the Ti layer is the increasing $E_{\text{stress}}$ due to the varying $\lambda$. In a (110) textured film, the $\lambda$ is given by $\lambda_{(110)} = (1/5)\lambda_{100} + (4/5)\lambda_{111} = 54$ ppm, while in a randomly oriented polycrystalline film, $\lambda$ is given by $\lambda_{\text{isotropic}} = (2/5)\lambda_{100} + (3/5)\lambda_{111} = 78$ ppm, where $\lambda_{100}$ and $\lambda_{111}$ are magnetostriction constant along the [100] and [111] of a single crystal, 150 ppm and 30 ppm, respectively. As discussed in the XRD results (Fig. 1(a)), the (110) texture exists in the sample without Ti layer, while the texture disappears in the sample with a Ti layer. Therefore, the $\lambda$ can be altered due to the texture change in the CoFe film. The $\lambda$ is estimated by measuring the same grown films on the Si substrate. As shown in Fig. 3(c), the effective saturation magnetostriction constant $\lambda_{\text{eff}}$ increases from 37 $\pm$ 2 ppm to 48 $\pm$ 2 ppm, as the Ti layer thickness increases from 0 nm to 4 nm.
However, when the Ti thickness increases from 4 to 10 nm, the $\lambda_{\text{eff}}$ is almost constant. Based on the measured $\lambda$, the $E_{\text{stress}}$ can be deduced to be increase as $E_{\text{stress}} = 3/2 \lambda Y$, where $Y$ is Young’s modulus of CoFe. Therefore, the energy reduction of $E_{\text{charge}}$ and $E_{\text{uni}}$ and meanwhile the increase of $E_{\text{stress}}$ make the $E_{\text{stress}}$ contribution to the $E_{\text{total}}$ dominant, thus produces the stronger converse ME coupling.

To investigate the repeatability of the ME coupling, Figs. 4(a)–4(d) show the magnetic properties change induced by E-field, when the E-field is applied at 12 kV/cm, then at 14 kV/cm and repeated for 30 measurements. Each magnetic hysteresis loop measurement takes about 30 s in the high H-field MOKE. The heterostructure shows a stable magnetization, thus produces the stronger converse ME coupling.

In summary, the effect of the introduction of a Ti layer on the converse ME coupling strength was investigated. Both the interface structure and the film structure were altered by the Ti layer. The addition of the Ti layer reduced the contribution of the original magnetic anisotropy energy to the total magnetic energy, which resulted in the enhanced converse ME coupling strength. In addition, these heterostructures also show good high-voltage stability suggesting potential applications in high voltage handling capability and high output power devices.

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