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Study and Simulation of the Electric Field-Induced Spin Switching in PZT/NiFe/CoFe Nanostructured Composites

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ABSTRACT

In this work, we have studied the electric field-induced spin switching in the PZT/NiFe/CoFe nanostructured composites by sputtering ferromagnetic layers on a horizontal polarized piezoelectric PZT substrate. The electric field-induced change in the magnetization orientation was investigated systematically using a vibrating sample magnetometer and analytical simulations. The results revealed that electric field applications could indirectly control the magnetic spin orientations. Moreover, the magnetization change depends not only on the electric field but also on the direction of the electric field applying against the magnetic field. The images of magnetic moment orientations under various electric field applications are modeled by the Monte Carlo and NMAG simulations. In particular, a critical electric field of $E_{cr} \approx 300$ kV/cm, which makes a 90° spin switching, was determined. These results are proposed to offer an opportunity for random access memory applications.

Keywords: Electric field controlled magnetic anisotropy, Monte Carlo and NMAG simulations, nanostructured composites

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INTRODUCTION

Magnetoelectric (ME) effect-induction of magnetization by an electric field or of electric-polarization by a magnetic field can be realized in multiferroic composites that exhibit a linear coupling between the applied electric voltage and magnetic field (Cheong & Mostovoy, 2007; Liang et al., 2021; Palneedi et al., 2016). A representative example of this idea is the ME random access memory (MERAM) device which permits an electric field-induced magnetization switching in a ferromagnetic (FM) layer based on the combination of ME coupling and interfacial exchange coupling between ferroelectric (FE) and FM phases. In particular, the electric-field control of spin switching, known as the converse ME effect, has attracted the attention of a large number of scientists from areas such as physics, computer science, and mathematics due to its potential applications in electrically tunable microwave magnetic devices (Eerenstein et al., 2006; Hong, Doan et al., 2013; Lin et al., 2016) and electric-write magnetic memories (Cheng et al., 2018; Kumar et al., 2021; Hu et al., 2010; Liu et al., 2009; Palneedi et al., 2016; Popov et al., 2020).

In the research topic of the electric field-induced spin-switching effect, we have demonstrated the switchability of voltage control to the magnetic anisotropy in PZT/NiFe/CoFe heterostructured nanocomposites (Hong, Doan et al., 2013). In this work, the material characterizations of the nanostructured composites have also been reported. Then, the converse magnetoelectric effect (Hong, Duc, et al., 2013) and the electrical field-induced magnetization switching in the nanocomposites (Hong et al., 2014) were systematically investigated under applied voltages up to \pm 700 V and \pm 400 V, respectively. Particularly, a change in magnetization up to 245% and above 100%, respectively, at low-bias magnetic fields could be reached thanks to elastic stress transfers from the PZT piezoelectric phase to NiFe/CoFe magnetic phase and/or the changes of NiFe thickness. However, higher bias voltages could not be increased due to the limitation of our experimental instruments. In order to further investigate the electric field-induced spin-switching behavior in the nanostructured composites and find out the critical electric field (E_{cr}), the magnetic characterizations combined with analytical Monte Carlo simulations will be done in this work with inheriting characterized results from our previous studies.

The Monte Carlo simulation has been an effective method to model the magnetic properties of ferromagnetic layers such as antiferromagnetic Fe/Cr layers (Masrour et al., 2014), multilayered magnetic structures with giant magnetoresistance (GMR) effect (Prudnikov et al., 2016), fcc Ni_cFe_{1-c} alloy (Taylor & Gyorffy, 1992), and Layered Ising Nanocube Fe/Co/Fe (Kadiri et al., 2022). The method has also been used for investigating the ferroelectric properties of PZT material (Tamerd et al., 2020; Cornelissen et al., 2019; Wei et al., 2021). However, few reports regarding Monte Carlo simulation for ferromagnetic (FM) and ferroelectric (FE) heterostructured nanocomposites exist. The work of Dung and Long (2016) should be a rare example. We have calculated the critical electric field E_c and determined electric field E_d for different ferromagnetic layers: Fe, Fe₃O₄, and CoFe₂O₄ (CFO) grown on ferroelectric PZN-PT substrate and proposed the structures for the MERAM application. Using Monte Carlo simulation to investigate the electric field induced spin switching effect for PZT/NiFe/CoFe heterostructured nanocomposites, to the best of our knowledge, is the first trial.

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Herein, the electric field-induced changes in the magnetization orientation of nanostructured PZT/NiFe/CoFe multiferroics have been investigated to determine the conformable parameters of the external electric field for upcoming experimental research on MERAM (Figure 1). The particular 90° spin switching behavior of the nanostructured composites, which may correspond to the switching between the "0" and "1" bits in MERAM, will be discussed. Furthermore, we observed that the applied electric field could control the spin configuration in the FM layer. Particularly, in the case of isotropic coupling, a small critical electric field (E_{er}) at which the spin started to rotate has been demonstrated.



Figure 1. (a) Schematic of PZT/NiFe/CoFe nanostructured composites with the proposed MERAM element. M_h and M_f are initial orientations of magnetization of the CoFe and NiFe layers, respectively. An electric field E is applied to the FE layer (PZT) to generate a 90° in-plane magnetization switching; and (b) Bits "0" and "1" switching in MERAM

MATERIALS AND METHODS

The FM layers of NiFe and CoFe were deposited in sequence on the horizontal polarized 500 μ m thick PZT (100) substrate at room temperature using a radio frequency (RF) magnetron sputtering device (2000-F, AJA International). The polarization of the PZT substrate was aligned along its original (100) plane. During the deposition process, an argon pressure and RF sputtering power of 2.2 × 10⁻³ Torr and 50 W, respectively, were set in the main deposited chamber. Finally, a thin Ta layer was sputtered onto the film surface to prevent the oxidization of FM layers. All sample areas were similar, but the thickness of the CoFe layer was fixed at 190 nm, while the thicknesses of the FM thin films were 10, 25, 50, and 90 nm for samples N1, N2, N3, and N4, respectively. The area of all samples was fixed at 5 mm × 5 mm. The electric field-induced change in the magnetization orientation was measured using a vibrating sample magnetometer (VSM 7400, Lakeshore). The switching of the magnetization vector driven by an electric field applied to the FE layer was investigated systematically using an analytical approach and Monte Carlo and NMAG simulations.

In this study, we used the parameters of the fabricated material and Monte Carlo simulation for the electrical-induced change in magnetization. In the MERAM structure, the ME effect occurred when an electric field was applied to the PZT substrate. This electric field created mechanical stress/strain on the piezoelectric substrate via ME coupling. Then, the stress was transported to the magnetostrictive phase, creating a strain in film planes x and y (corresponding to stress σ_x and σ_y). The total magnetoelastic energy is as stated in Equation 1:

$$F_{me} = -\frac{3}{2}\lambda\sigma_x \sin^2\theta \cos^2\varphi - \frac{3}{2}\lambda\sigma_y \sin^2\theta \sin^2\varphi \qquad [1]$$

where θ is the angle between the magnetization and electrical field directions, φ is the angle between the easy magnetization axis and electrical field direction, and λ is the effective magnetostriction constant in the film plane. This electrical field creates the effective field of $\overline{H_{eff}} = -\nabla_M \cdot F_{me}$. The total free energy is represented by Equation 2:

$$F_{free} = F_{me} + F_{zeeman} = \frac{-3}{2}\lambda\sigma_x \sin^2\theta \cos^2\varphi \frac{-3}{2}\lambda\sigma_y \sin^2\theta \cos^2\varphi - \overline{H} \cdot \overline{M_s} + 2\pi M_s \cos^2\theta$$
[2]

The effective anisotropy field in the x, y, and z axes was computed using Equation 3:

$$H_{eff,x} = \frac{3\lambda(\sigma_x - \sigma_y)}{M_s}, \ H_{eff,y} = \frac{-3\lambda(\sigma_x - \sigma_y)}{M_s}, \ H_{eff,z} = \frac{-3\lambda(\sigma_x + \sigma_y)}{M_s}$$
[3]

where $\sigma_{x_x} \sigma_y$ is the compressive stress in [100] axis and the tensile stress in [011] axis, respectively. The effective anisotropy field is calculated by the following Equations 4 to 6:

$$H_{eff,x} = \frac{3\lambda Y}{M_s(1+\upsilon)} (d_{31} - d_{32})E$$
[4]

$$H_{eff,y} = \frac{-3\lambda Y}{M_s(1+\upsilon)} (d_{31} - d_{32})E$$
[5]

$$H_{eff,z} = \frac{-3\lambda Y}{M_{s}(1+\upsilon)} (d_{31} + d_{32})E$$
[6]

where λ is the magnetostriction constant in the film plane, Y is Young's modulus, d₃₁, and d₃₂ are the piezoelectric coefficients, E is the electric field, and υ is Poisson's constant. The total magnetoelastic energy is given by the following Equation 7:

$$F_{ME,x} = -H_{eff,x} \cdot \overrightarrow{M_s} \quad F_{ME,y} = -H_{eff,y} \cdot \overrightarrow{M_s} \quad F_{ME,z} = -H_{eff,z} \cdot \overrightarrow{M_s}$$
[7]

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In the ME material, the rotation process can only occur when the condition $\Delta F_{total} < 0$ is satisfied. The spin switched from the film plane to out of the plane, corresponding to the switching between the "0" and "1" bits in MERAM. For the calculation for $F_{ME,x}$ was determined by using Equation 7, and with the material parameters, we obtained remarkable results that will be discussed in the below section.

RESULTS AND DISCUSSION

PZT/NiFe/CoFe Nanostructure Composites

Magnetic Properties. Figure 2 shows the room temperature M(H) curves of sample N1 under magnetic fields up to 10 kOe at various angles between the film plane and magnetic-field directions of $\alpha = 0^{\circ}$, 45°, and 90°. The results indicate that the M(H) curves

were easier magnetized along the in-plane direction (i.e., $\alpha = 0^{\circ}$) than in 45° and out-of-plane (i.e., $\alpha = 90^{\circ}$) directions. The coercivity (H_c) values are 136, 147, 163 Oe; the saturation magnetization (M_s) values are 1074, 1057, 1041 µemu; and the remanence magnetization (M_R) values are 179, 136, 59 μ emu, corresponding to the angles of 0°, 45° , and 90° , respectively. The smallest H_c, highest M_S, and highest M_R values belong to the in-plane M(H) curve. A similar tendency was observed for the samples N2, N3, and N4, whose H_c , M_s , and M_R values are listed in Table 1. These results reveal the in-plane magnetic anisotropy and the typical soft magnetic property of the NiFe/CoFe layers that originated from the contribution of the horizontally polarized PZT (100) substrate.



Figure 2. M(H) loops of 240 nm NiFe/CoFe magnetic films on PZT substrate (sample N1) were measured for in-plane, 45°, and out-of-plane configurations. The inset shows zoom in M(H) loops near the coercive field region

394

246

Comple	H _C (Oe)			M _s (µemu)			M _r (µemu)		
Sample -	//	45°	\perp	//	45°	T	//	45°	\perp
N1	136	147	163	1074	1057	1041	179	136	59
N2	97	102	108	1495	1466	1460	90	69	35
N3	91	97	104	1612	1459	1375	225	135	82

Table 1

N4

58

63

75

Coercivity (H_c), saturation magnetization (M_s), and remanence magnetization (M_R) of samples N1, N2, N3, and N4

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1805

1718

2004

129

Electrical Field-Induced Magnetization Switching in PZT/NiFe/CoFe Nanostructured Composites. Figure 3 depicts the dependence of the magnetization M on the angle α of sample N1 with zero applied bias voltage under various applied magnetic fields of H_{bias} = 0, ± 50, ± 100, ± 200, ± 500, ± 1000, and ±2000 Oe. The magnetization's changing slope, and phase shift in the sinusoidal form can be observed.

Under zero applied bias voltage, the $M(\alpha)$ curve reaches its maximum absolute magnitude while the magnetic field was applied parallelly to the sample plane at $\alpha = 0^{\circ}$, 180°, and 360°. The changing period of $M(\alpha)$ should be $T = 180^{\circ}$. The magnetization was the smallest while the magnetic field was applied perpendicularly to the sample plane (α = 90°, 270°). Furthermore, the sinusoidal form of $M(\alpha)$ curves is in the same direction for $H_{\text{bias}} = -100$ to 2000 Oe. The change in the direction of curves appeared when the magnetic field value of $H_{\text{bias}} = -200$, -1000, and -2000 Oe, which exceeded the value of the out-of-plane coercivity field H_{c} of 163 Oe. This behavior is supposed to be the elastic stress-induced transition from the piezoelectric PZT substrate to the ferromagnetic NiFe/ CoFe layers resulting in the electrical field-induced magnetization switching. Particularly, the stress caused by the film-to-substrate mismatch may substantially impact the observed anisotropy. Two couplings are supposed to be responsible for the rotation of magnetization orientation when an electric field is applied: (1) the exchange interaction between NiFe and CoFe layers and (2) the electromagnetic coupling between NiFe/CoFe and PZT phases.

In order to investigate the dependence of the magnetic reversal of sample N1 in bias voltage applications through the PZT substrate, the $M(\alpha)$ curve at $H_{bias} = 50$ Oe under applied bias voltages of U = 0 V, 100 V, and -200 V were plotted (Figure 4). Under an applied bias voltage of U = 100 V, the maximum absolute magnitude of the $M(\alpha)$ curve increased



Figure 3. M(α) curve of sample N1 measured at various applied magnetic field H_{bias}

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higher than that under zero bias voltage at $\alpha = 30^{\circ}$, 150° , and 270° . The changing period of T = 120° is lower than that under zero bias. In particular, a revered sinusoidal M(α) curve was also observed with an applied bias voltage of U = -200 V, similar to the above case of applied magnetic field H_{bias} > H_C. This result proposed that the change in the magnetic reversal mechanism under a sufficiently large applied bias voltage is due to the competition between magnetic and electrical energy. Moreover, the magnetization change depends not only on the electric field applying against the PZT



Figure 4. $M(\alpha)$ curves of sample N1 measured at H_{bias} = 50 Oe under bias voltage U = 0 V (black line), U = 100 V (red line), and U = -200 V (blue line)

substrate but also on the direction of the electric field applying against the magnetic field. When an electric field is present, the competition between the electric and magnetic fields will determine the magnitude and anisotropy of the magnetic layers.

Table 2 shows the variation of magnetization $\Delta M = M_{max} - M_{min}$ measured at $\alpha = 0^{\circ}$ under various electric voltages and magnetic fields. At $H_{bias} = 50$ Oe, the ΔM value is 120 µemu under zero bias voltage and increases to 215 µemu when the bias voltage of U = 100 V, then reaches 230 µemu when the bias voltage up to U = -200 V. Since the NiFe/CoFe films were sputtered directly onto the piezoelectric PZT substrate, and the piezoelectric force will be transferred directly from the substrate to the thin films that make changes in the composite magnetization. Furthermore, the $\Delta M(\alpha)$ showed different magnitudes depending on the magnetic field H_{bias} demonstrating the competition of electric and magnetic fields in the process to varying degrees.

Table 2 The change of magnetization ΔM (µemu) measured at $\alpha = 0^{\circ}$ under various electric voltages and magnetic fields

U (V)	H_{bias} = -2000 Oe	H_{bias} = -50 Oe	$H_{bias} = 50 \text{ Oe}$	$H_{bias} = 200 \text{ Oe}$
0	160	100	120	140
100	240	220	215	245
-200	270	265	230	240

In this work, the distance between two electrodes was 5 mm due to the sample area was fixed at 5×5 mm², resulting in electric fields being E = 0, 0.2, and -0.4 kV/cm for bias voltages of U = 0, 100, and -200 V, respectively. Needless to say, the polarization moments in the PZT substrate are hard to reach the saturation states under these small electric field applications. However, higher bias voltages could not be increased due to the limitation

of our experimental instruments. In order to further investigate the electric field-induced spin-switching behavior in PZT/NiFe/CoFe nanostructured composites, the Monte Carlo and NMAG simulations should be an appropriate approach in this situation.

Analytical and Numerical Approach

The Monte Carlo and NMAG simulations were conducted for the PZT/NiFe/CoFe composite, assuming that the isotropic biaxial stress $d_{31} = d_{32}$ (the deformation on the x and y axes is the same) where the effective field generated by the strain on these axes was zero (Hu et al., 2010). In other words, the distortion in the z-axis mainly generated the effective field value. In this work, the values of $d_{31} = d_{32} = -276$ pC/N were used for the composite.

Figure 5 shows the side-view and top-view simulating images of the magnetic moments under various electric field applications for the PZT/NiFe/CoFe composite. Without an external electric field (Figure 5a), the magnetic moments, described by red arrows, were aligned parallel along the polarization moments of the PZT (100) substrate (i.e., the film plane) due to the interactive energy exchanged being ignored in the initial state. With an external electric field of E = 30 kV/cm, the effective field produced a rearrangement in the magnetic moments that deviated from the film plane with an angle of 6.87° and increased to 45° under a higher electric field of E = 190 kV/cm (Figures 5b & 5c). The

distribution and direction of the magnetic moments in the FM layer are shown in Figure 5d with the critical electric field $E_{\rm cr}$ $\approx 300 \text{ kV/cm}$, at which point the angle θ = 89.9°, and the magnetic moments were nearly perpendicular to the film plane. From this point onward, the system reached the saturation state, and the magnetic moments were oriented parallel to each other and perpendicular to the film plane.

Figures 6a and 6b show the graphs of the changes in the orientations of magnetic moments under various electric fields in $\cos\theta$ and θ , respectively. The change can be explained by the effective field generated when an external electric field is applied, and the magnetic moments are rearranged in the direction of the effective field. The rotation angle of the magnetic moment increased linearly as the electric field



Figure 5. Side-view and top-view images of the magnetic moment orientations under various electric field applications: (a) E = 0 kV/cm; (b) E = 30 kV/ cm; (c) E = 190 kV/cm; (d) E = 540 kV/cm; and (e) 1100 kV/cm, respectively

increased and reached the saturation value of $\theta = 90^{\circ}$ under electric fields of $E \ge E_{cr}$, at which the magnetic moment is perpendicular to the film plane. This finding is proposed to offer an opportunity for fabricating the MERAM devices using the PZT/NiFe/CoFe composite based on the magnetic reversal effect in its isotropic structure mode.



Figure 6. The changes in orientations of the magnetic moment based on the applied electric field through (a) $\cos\theta$ and (b) θ in the isotropic biaxial stress mode

CONCLUSION

The electric field-induced spin switching has been studied in the PZT/NiFe/CoFe nanostructured composites. The rotation of magnetization orientation under an electric field application is demonstrated for the exchange interaction between NiFe and CoFe layers and the electromagnetic coupling between NiFe/CoFe and PZT phases. The NMAG simulation has observed the orientation of magnetic moments under various electric field applications. In addition, using the Monte Carlo simulation, a critical electric field of $E_{cr} \approx 300 \text{ kV/cm}$ has been determined to make a 90° spin switching. The findings proposed an opportunity to achieve new types of memory devices, such as voltage-tunable field random access memories.

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