Modulation of Magnetic Anisotropy in Flexible Multiferroic FeGa/PVDF Heterostructures Under Various Strains

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Control of the magnetic anisotropy under different strain states is important for the manipulation of magnetization in flexible spintronic devices. Here, the electric field control of magnetic anisotropy was investigated in flexible $Fe_{81}Ga_{19}$(FeGa)/polyvinylidene fluoride multiferroic heterostructures under different compressive strains. The initial uniaxial magnetic anisotropy is enhanced with increasing the compressive strain. When the strain is larger than 0.06%, the electric field can only change the strength of the magnetic anisotropy but cannot change its direction. When the strain is smaller than 0.06%, the electric field of 267 kV/cm can reorientate the easy axis by 90°. The present results may be helpful for designing flexible multiferroic devices with a suitable initial strain in order to realize the 90° reorientation of the magnetization.

***Index Terms*—FeGa, flexible, magnetic anisotropy, multiferroic heterostructures, polyvinylidene fluoride (PVDF).**

I. INTRODUCTION

IN TRADITIONAL electronics, charges and spins were separately controlled by electric and magnetic fields, respectively [1]. By controlling the magnetization direction in magnetic multilayers, giant magnetoresistance (GMR) was discovered, which paves the way for spintronics [2]. Due to the large magnetoresistance, the read heads based on the GMR and the following tunneling magnetoresistance (TMR) have greatly contributed to the fast rise in the storage density of the hard disks [3]. Recently, GMR or TMR devices fabricated on flexible substrates, so-called flexible spintronics, have attracted a lot of interests due to their mechanical flexibility, light weight, and low cost [4], [5]. In spintronic devices, the magnetic anisotropy, which determines the magnetization direction and thus the spin transport, is the key issue and need to be well controlled [6], [7].

Multiferroic materials show the coupled ferroelectricity and ferromagnetism, in which the magnetism can be controlled by an electric field [8], [9]. However, most of the single-phase multiferroic materials exhibit a low Curie temperature or a weak intrinsic magnetoelectric (ME) coupling especially above room temperature [10]. An attractive alternative way is to use ferromagnetic/ferroelectric (FM/FE) multiferroic heterostructures. Consequently, the electric control of magnetism can be realized through strain-mediated ME coupling across the interface [11], [12]. In strain-mediated multiferroic heterostructures, an effective uniaxial strain produced through the converse piezoelectric effect when an electric field applied on FE layer is transferred to FM layer, due to the inverse magnetostrictive effect, resulting in the change of magnetic anisotropy [13]. However, for FM/FE thin-film heterostructures deposited on rigid substrates, the strain transfer across the interface is remarkably clamped by the rigid substrates, which limits the tunability of magnetic properties by an electric field [11]. In contrast, for flexible multiferroic heterostructures that can be used in arbitrary surface [14], the mechanical strain due to the deformation is inevitably produced and gives rise to an additional magnetic anisotropy in the flexible multiferroic heterostructures. Therefore, the modulation of magnetic anisotropy under different strain states is important in flexible multiferroic heterostructures and need to be known prior to the application in flexible spintronic devices. Here, we report the electric field control of magnetic anisotropy in flexible $Fe_{81}Ga_{19}$ (FeGa)/polyvinylidene fluoride (PVDF) multiferroic heterostructures under different compressive strains. It is found that the uniaxial magnetic anisotropy can be changed 90° by an electric field of 267 kV/cm when the external strain is smaller than 0.06%.

II. EXPERIMENT

PVDF is organic FE materials exhibiting excellent mechanical flexibility, relatively good piezoelectric properties ($d_{31} = 21.4$ pC N^{-1}, $d_{32} = 2.3$ pC N^{-1}), and low production cost, which are good candidates for developing flexible multiferroic heterostructures [15], [16]. The $Fe_{81}Ga_{19}$ (FeGa) alloy is a typical magnetostrictive material exhibiting a moderate magnetostriction of 350 ppm under a very low magnetic field and excellent mechanical properties [17]. The commercial 30 μm thick PVDF membranes were coated on both sides by the 50 nm thick Al layers. The $Fe_{81}Ga_{19}$ films and Au protective layers were prepared by magnetron sputtering at room temperature. The base pressure of the vacuum chamber was in the range of 10^{-5} Pa. During sputtering, the argon flow was kept at 50 sccm and the pressure was set at 0.5 Pa. The growth rate for growing FeGa film is about 10.0 nm/min. The thicknesses of FeGa and Au layers are about

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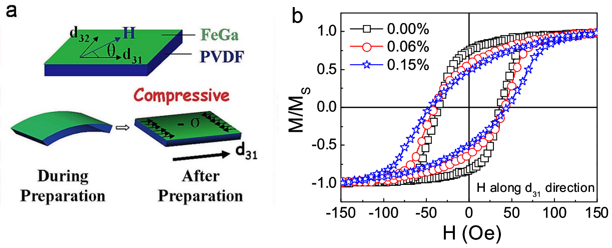

Fig. 1. (a) Schematic of FeGa/PVDF heterostructures during and after preparation. (b) Magnetic hysteresis loops of the FeGa/PVDF films under different compressive strains with an in-plane magnetic field along the d_{31} direction.

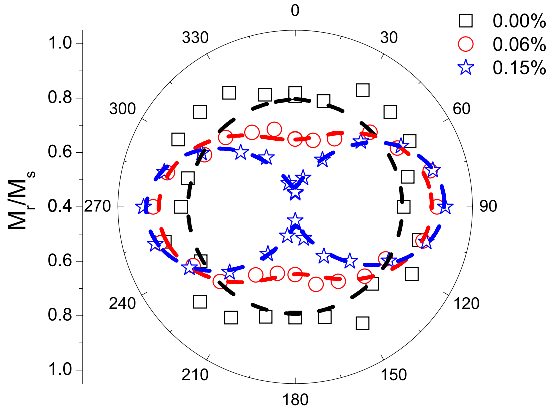

Fig. 2. Angular dependence of normalized remanent magnetization for FeGa/PVDF films with different compressive strains along the d_{31} direction.

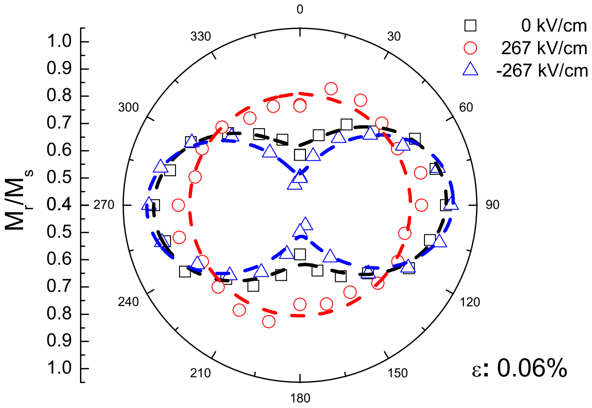

Fig. 3. Angular dependence of normalized remanent magnetization for the FeGa/PVDF film with a compressive strain of 0.06% along the d_{31} direction under different electric fields.

60 and 5 nm, respectively. In order to set the direction of the initial magnetic anisotropy, during the deposition of FeGa layers, apart from the flatten substrates, the PVDF membranes were bowed along the d_{31} direction by directly fixing it to a convex surface with the radii of 25 and 10 mm. After preparation, the bowed substrates were flattened to a plane and a compressive strain along the d_{31} direction was generated in the flexible heterostructures, as shown in Fig. 1(a). The strain is evaluated using the equation $\varepsilon = t/2\rho$, where t is the thickness of the substrate including the film thickness and ρ is the curvature radius of the convex surface. The compressive strains along the d_{31} direction are estimated to be 0.06% and 0.15% for the radii of 25 and 10 mm, respectively.

The thicknesses of FeGa and Au layers were calibrated by X-ray reflectivity. A standardized FE test system (Precision Premier II, Radiant Technologies) was used to measure the electric hysteresis loops of PVDF. The magnetic hysteresis loops of FeGa layer were measured at different polarization states of PVDF by magneto-optical Kerr effect (MOKE) magnetometer. The magnetic field H was applied in-plane with an angle of θ with respect to the d_{31} direction. The Al layers on both sides of PVDF were connected to a voltage source (Keithley 237 high-voltage source-measure unit) with thin Pt wires. During the MOKE measurements, the voltage source provides an electric field to polarize the FE PVDF substrate thought the thickness.

III. RESULTS AND DISCUSSION

Fig. 1(b) shows the magnetic hysteresis loops of FeGa/PVDF film under different compressive strains with H along the d_{31} direction. Due to the positive magnetostriction of FeGa, the compressive strain leads to the behaviors of hard axis for FeGa/PVDF films along the d_{31} direction [18]–[20]. With the increase in compressive strain, the hysteresis loops exhibit a more slanted shape. For the flat-grown film, i.e., $\varepsilon = 0$, the normalized remanent magnetization M_r/M_s measured along the d_{31} direction is about 0.75. When the strain is increased to 0.15%, M_r/M_s is decreased to 0.45, which indicates the enhancement of uniaxial magnetic anisotropy. In order to directly view the change of magnetic anisotropy under different strains, the hysteresis loops of FeGa/PVDF films are measured at various field orientations. Thus, the angular dependence of M_r/M_s is shown in Fig. 2. The sample with $\varepsilon = 0$ exhibits a nature of magnetic isotropy, since the M_r/M_s ratio of 0.75 nearly keeps unchanged at an arbitrary field direction. When for the FeGa/PVDF film with $\varepsilon = 0.06\%$ along the d_{31} direction, M_r/M_s obtained along the d_{31} direction ($\theta = 0°$) is reduced to 0.58, while M_r/M_s measured along the d_{32} direction ($\theta = 90°$) is enhanced to 0.93. The corresponding angular dependence of M_r/M_s displays a shape of ∞, indicating a uniaxial magnetic anisotropy with an easy axis along the d_{32} direction. When the compressive strain is further increased to 0.15%, the enhancement of the uniaxial magnetic anisotropy results in the decrease of M_r/M_s along the d_{31} direction to 0.44 and the increase of M_r/M_s along the d_{32} direction to 0.95.

Fig. 3 shows the angular dependence of M_r/M_s ratios for the FeGa/PVDF film with $\varepsilon = 0.06\%$ measured at different electric fields applied through the thickness of PVDF. When no electric field is applied, the ∞ shaped curve reveals a uniaxial magnetic anisotropy along the d_{32} direction. When the electric field is increased to 267 kV/cm, the M_r/M_s ratios measured along the d_{31} and d_{32} directions are increased and decreased, respectively. The curve for the angular dependence of M_r/M_s is changed from ∞ to roughly circular, which indicates that the film become the magnetic isotropy under the positive electric field. When the electric field is varied from 0 to -267 kV/cm, the ∞ shape is elongated

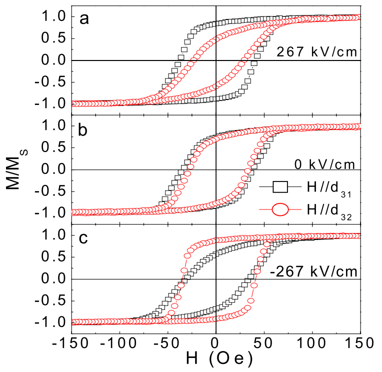

Fig. 4. Magnetic hysteresis loops of the unstrained FeGa/PVDF sample with magnetic field applied along the d_{31} and d_{32} directions at (a) 267, (b) 0, and (c) −267 kV/cm.

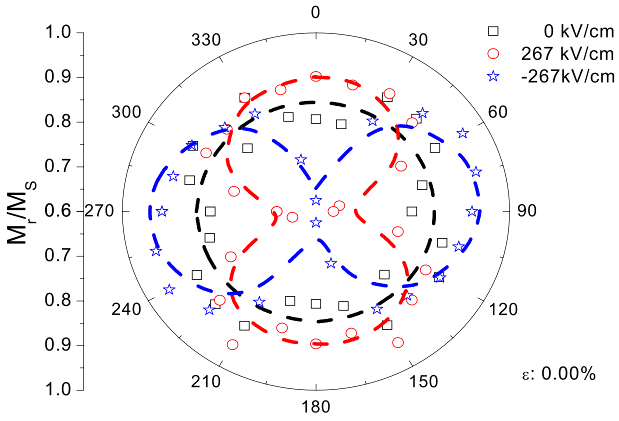

Fig. 5. Angular dependence of normalized remanent magnetization for the unstrained FeGa/PVDF sample at different electric fields.

along the d_{32} direction, which reflects the enhancement of the uniaxial magnetic anisotropy along the d_{32} direction. Obviously, when the FeGa/PVDF sample is suffered a compressive strain along the d_{31} direction larger than 0.06%, the electric field of 267 kV/cm can only tune the strength of the magnetic anisotropy but cannot reorient the direction of uniaxial magnetic anisotropy. In order to realize the reorientation of the uniaxial magnetic anisotropy by electric field, the magnetic properties of the unstrained FeGa/PVDF sample were investigated under various electric fields. Fig. 4 shows the hysteresis loops of the unstrained FeGa/PVDF sample with magnetic field applied along the d_{31} and d_{32} directions under electric fields of 267, 0, and −267 kV/cm. For the zero electric field, the hysteresis loops measured along the d_{31} and d_{32} direction are nearly identical, which shows that the sample is magnetic isotropy. When the electric field of 267 kV/cm is applied, the hysteresis loop measured along the d_{31} direction is more square than the one measured along the d_{32} direction, which indicates that the application of electric field generates a uniaxial magnetic anisotropy with an easy axis along the d_{31} direction. When an electric field of −267 kV/cm is applied, the hysteresis loop along the d_{31} direction is more slanted than that along the d_{32} direction, since a uniaxial magnetic anisotropy is induced along the d_{32} direction. As shown in Fig. 5, the angular dependence of M_r/M_s ratio for the unstrained FeGa/PVDF film displays the distinct changes under different electric fields. When no electric field is applied, M_r/M_s is 0.84 at an arbitrary field orientation. When an electric field of 267 kV/cm is applied, M_r/M_s along the d_{31} direction ($\theta = 0°$) is increased to 0.9, and M_r/M_s along the d_{32} direction ($\theta = 90°$) is decreased to 0.68. The curve shape becomes 8, corresponding to the uniaxial magnetic anisotropy with an easy axis along the d_{31} direction. When an electric field of −267 kV/cm is applied, the values of M_r/M_s measured along the d_{31} and d_{32} directions are decreased and increased to 0.68 and 0.93, respectively. The corresponding curve shape of ∞ confirms the uniaxial magnetic anisotropy with an easy axis along the d_{32} direction. Clearly, our experimental observations demonstrate that the electric field can induce the reorientation of uniaxial magnetic anisotropy in the unstrained FeGa/PVDF sample.

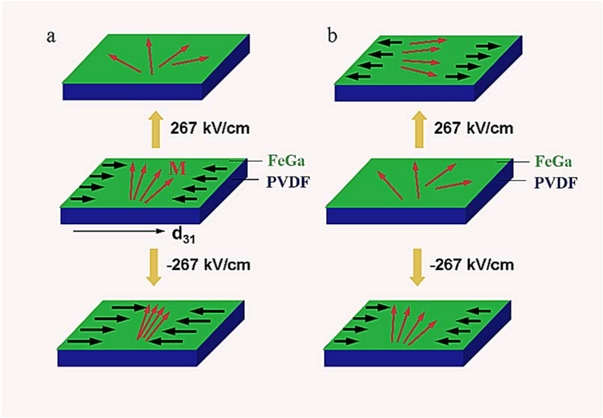

Fig. 6. Schematic of electric field manipulation of the FeGa domain orientations for (a) sample under the compressive strain of 0.06% along the d_{31} direction and (b) unstrained sample.

Fig. 6 schematically shows the orientations of FeGa domains for the FeGa/PVDF sample with the compressive strains under different electric fields. As observed in Fig. 6(a), for the sample with $\varepsilon = 0.06\%$ along the d_{31} direction, the FeGa moments are inclined to align along the d_{32} direction, i.e., the easy axis. The strain ε_E produced by the electric field E via the converse piezoelectric effect is evaluated to be $(d_{31}–d_{32})E$ [21]. When an electric field of 267 kV/cm is applied, the tensile strain along the d_{31} direction is estimated to be 0.051%, which could cancel out the effect caused by the initial compressive strain along the d_{31} direction. Consequently, the sample becomes magnetically isotropic, the domains can orientate randomly in this scenario. When an electric field of −267 kV/cm is applied, the compressive strain along the d_{31} direction is 0.051%, which unstrengthens the initial compressive strain along the d_{31} direction. Therefore, the domain orientations are narrowly concentrated along the d_{32} direction. As observed in Fig. 6(b), the unstrained sample is magnetically isotropic at the as-grown state and the domain orientations are random. When an electric field of 267 kV/cm is applied, the tensile strain along the d_{31} direction is generated, which results in an easy axis along the d_{31} direction. The domain orientations prefer to be aligned along the d_{31} direction. When an electric field of −267 kV/cm is applied, the compressive strain along the d_{31} direction is induced and the easy axis is along the d_{32} direction. Therefore, the domain orientations are squeezed into a narrow distribution along the d_{32} direction.

IV. CONCLUSION

In conclusion, the flexible multiferroic FeGa/PVDF heterostructures have been successfully fabricated with different compressive strains along the d_{31} direction. The uniaxial magnetic anisotropy with an easy axis along the d_{32} direction is enhanced with increasing the compressive strain. When the strain is smaller than 0.06%, an electric field of 267 kV/cm can reorientate the uniaxial magnetic anisotropy. Our experimental results suggest that the reorientation of uniaxial magnetic anisotropy by electric field can be realized in flexible multiferroic heterostructures when the strain generated by the electric field is strong enough to cancel out the initial strain.

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