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In-plane anisotropic converse magnetoelectric coupling effect in FeGa/polyvinylidene fluoride heterostructure films

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We investigated the converse magnetoelectric (CME) effect in the $Fe_{81}Ga_{19}$/polyvinylidene fluoride (PVDF) heterostructure films. A weak in-plane uniaxial magnetic anisotropy was observed in the as-deposited magnetostrictive FeGa films. When a positive (negative) electric field is applied on the ferroelectric PVDF substrates, both the coercivity and the squareness of magnetic hysteresis loops of FeGa films for the magnetic field parallel to the easy axis become larger (smaller), but for the magnetic field parallel to the hard axis the coercivity and the remanence get smaller (larger), indicating an anisotropic CME effect in FeGa/PVDF heterostructure films. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4793780]

Enormous attention has been paid on the multiferroic materials in view of the possibility of realizing control electric or magnetic properties by magnetic or electric fields via magnetoelectric (ME) or converse magnetoelectric (CME) effect during the last decade.[1–3] Electrostatic control of magnetization or magnetic control of electricity, if realized, could prevail in information storage and sensors, etc.[4–6] Typically, there are two classes of multiferroic materials: single-phase multiferroics and multiferroic composites comprised ferroelectric (FE) and ferromagnetic (FM) substances.[1,7] The ME and CME couplings in the strain-mediated multiferroic composites are much stronger than that in the discovered single-phase multiferroics.[8] Therefore, the multiferroic composites seem much promising toward the application of the electric control of magnetism or magnetic control of electricity.

Recently, numerous efforts focused on the CME effect in a number of multiferroic composites, including Fe_3O_4/$(PbMg_{1/3}Nb_{2/3}O_3)_{1-x}$-$(PbTiO_3)_x$ (PMN-PT),[9] Ni/PMN-PT,[10] FeCoB/Pb(Zr,Ti)O_3 (PZT),[11] etc. Liu *et al.*, found that with the electric field applied on the PMN-PT single crystal, the Fe_3O_4 films display remarkably different magnetic switching processes when it is magnetized along the two in-plane orthogonal directions [01–1] and [100] of the PMN-PT slab.[9] A Ni/PMN-PT heterostructure provides an electric-field-induced switching between two reversible and permanent magnetic easy axes which are perpendicular to each other.[10] With positively poling the PZT layer along the easy axis of the FeCoB films, the uniaxial magnetic anisotropy of FeCoB/PZT films can be reinforced; while the positive polarization along the hard axis, the strength of the uniaxial anisotropy is lowered.[11] These works show an indication of the anisotropic CME response with changing the magnetic field orientations. Therefore, in order to obtain a large CME response, it is very important to study the angular dependent CME effect. In this work, the angular dependent CME effect in $Fe_{81}Ga_{19}$ (FeGa)/polyvinylidene fluoride (PVDF) heterostructure films was studied in detail. The PVDF is used as both the ferroelectric layer and the flexible substrate due to the potential applications in flexible sensors, curved memory, and other non-planar devices.[12–14] The magnetostrictive layer investigated here is based on FeGa alloy arising from the giant positive magnetostriction coefficient ($\sim$400 ppm for the typical bulk), which is also crucial for the enhancement of CME coupling in the ferroelectric/ferromagnetic heterostructure films. In such flexible FeGa/PVDF heterostructures system, we observed an in-plane anisotropic CME coupling effect. When the magnetic field is applied parallel to the easy axis, the coercivity and the squareness of FeGa films are enhanced with increasing the external electric field applied on PVDF while the behaviors are the opposite when the magnetic field applied along the hard axis. The CME effect in FeGa/PVDF heterostructure films is of anisotropy.

FeGa films with 60 nm in thickness were prepared by magnetron sputtering on the 25 μm-thick PVDF substrates, which have been coated with Al layer on both sides. FeGa films were grown at a DC sputtering power of 35 W and an Ar pressure of 1.0 Pa at room temperature. A 5 nm-thick Au capping layer was deposited on top of FeGa films to prevent oxidation. Magneto-optical Kerr effect (MOKE) magnetometry was utilized to probe the magnetic properties of the FeGa films under various polarizations of PVDF substrates by applying different electric voltages between the Au capping layer and the bottom Al layer. The magnetic field was applied in the plane of FeGa films with various orientations θ, as schematically shown in Fig. 1(a).

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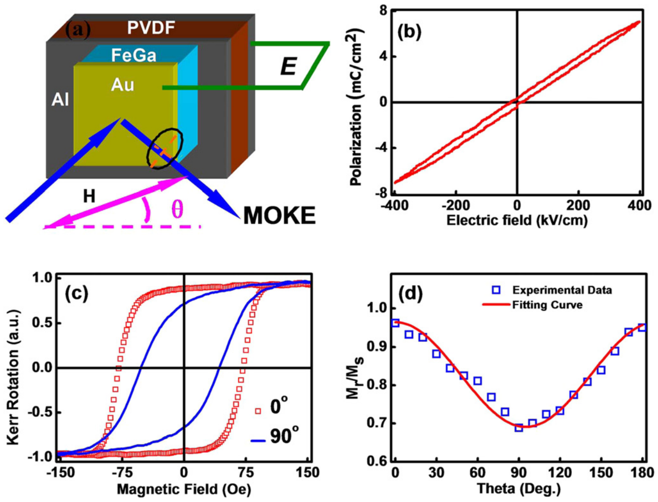

FIG. 1. (a) Schematic diagram of the sample structure and the measurement configuration. (b) Polarization versus electric field hysteresis loops of the PVDF substrate. (c) Typical Kerr hysteresis loops of the FeGa/PVDF films with the magnetic field applied along the easy and hard axes in absence of an applied electric field. (d) Angular dependence of squareness of the hysteresis loops for FeGa/PVDF.

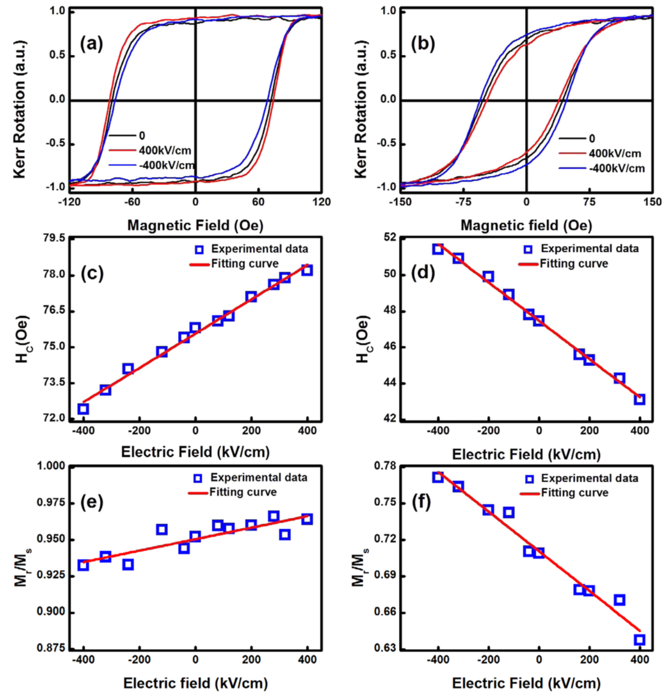

FIG. 2. Typical magnetic hysteresis loops of FeGa films along (a) the easy and (b) the hard axes under various electric fields. (c) The coercivity and (e) the squareness of FeGa films as a function of electric field when the magnetic field applied along the easy axis. (d) The coercivity and (f) the squareness of FeGa films as a function of electric field when the magnetic field applied parallel to the hard axis.

Prior to measure the Kerr hysteresis loops of FeGa films, the ferroelectric hysteresis loop of PVDF substrate is obtained, as shown in Fig. 1(b). Although the electric field reaches 400 kV/cm, the ferroelectric hysteresis loop shows that the polarization of PVDF is non-saturated. The normalized longitudinal Kerr hysteresis loops for the as-grown FeGa films are displayed in Fig. 1(c). The magnetic hysteresis loop for the magnetic field applied parallel to the easy axis ($\theta = 0°$) of FeGa layers is relatively square with the considerable coercive field H_c and the magnetic remanence M_r which is close to the saturation magnetization M_s. The magnetic hysteresis curve for the magnetic field applied along the hard axis ($\theta = 90°$) possesses the low coercive field and low remanent magnetization. The squareness M_r/M_s of the magnetic hysteresis loops as a function of the field orientation is in a typical cosine curve, as presented in Fig. 1(d), indicating a weak in-plane uniaxial magnetic anisotropy in FeGa films. During the deposition process of FeGa films, the in-plane magnetic anisotropy is formed due to the anisotropic thermal expansion of PVDF substrate at the elevated temperature. The thermal expansion coefficients α_1 along the elongated direction during fabrication and α_2 perpendicular to the elongated direction of PVDF are 0.13×10^{-4} K^{-1} and 1.45×10^{-4} K^{-1}, respectively.[15] As a result, the directions parallel to the elongated direction and perpendicular to the elongated direction of the PVDF substrates are the easy and hard axes for the FeGa films, respectively.

In order to study the CME effect of FeGa/PVDF heterostructure films, the Kerr hysteresis loops for the magnetic field along the easy and hard axes of FeGa films are measured under various strengths of the electric fields applied on the PVDF substrates. As displayed in Fig. 2(a), for the magnetic field applied along the easy axis and the electric field increased from −400 kV/cm to 400 kV/cm, the coercivity of the FeGa films is enhanced from 72 Oe to 78 Oe and the squareness is changed from 0.93 to 0.97. In contrast, for the magnetic field applied along the hard axis and the electric field increased from −400 kV/cm to 400 kV/cm, the coercivity of the FeGa films is decreased from 51 Oe to 43 Oe and the squareness is changed from 0.64 to 0.77, as shown in Fig. 2(b). The variations of H_c and M_r/M_s of FeGa films with changing the applied electric field in the range of ±400 kV/cm are summarized in Figs. 2(c)–2(f). For the magnetic field applied along the easy axis, H_c and M_r/M_s of the FeGa films are lineally increased as the electric field is changed from negative to positive while H_c and M_r/M_s are lineally decreased with increasing the electric field when the magnetic field applied along the hard axis. As shown in Fig. 2(e), the M_r/M_s ratio of FeGa films without applying electric field on PVDF substrates reaches 0.95 very close to 1.0 when the magnetic field is applied along the easy axis. The tunability of M_r/M_s resulting from the electric field is very small, when the electric field is changed from −400 kV/cm to 400 kV/cm, M_r/M_s is varied from 0.93 to 0.97. However, M_r/M_s of FeGa films without applying electric field on the PVDF substrates is about 0.77 far from 0 when the magnetic field is applied along the hard axis. The tunability of M_r/M_s is relatively large, M_r/M_s is decreased from 0.77 to 0.64 when the electric field is increased from −400 kV/cm to 400 kV/cm.

It is well known that the ME coupling in multiferroic composites could be mediated by the strain transfer, accumulation of spin-polarized charge, or exchange bias at the FE/FM interface.[16] The charge-mediated ME effect has been found to be significant in multiferroic heterostructures containing ultrathin ferromagnetic films with a thickness less than 10 nm,[17] but in our FeGa/PVDF heterostructures, this effect can be neglected due to the thick FeGa layer. On the other hand, the strain caused by the converse piezoelectric effect of PVDF layer can be transferred to the magnetic layer, producing an effective magnetic anisotropy due to the inverse magnetostrictive effect. The effective magnetic anisotropy field H_{eff} induced by the electric field can be expressed as[9]

$$H_{eff} = \frac{3\lambda Y}{M_s(1+\nu)}(d_{31} - d_{32})E, \quad (1)$$

where Y is the Young's modulus ($2.2 \times 10^9\,\mathrm{N/m^2}$ for PVDF),[18] ν is the Poisson's ratio (0.35 for PVDF),[19] λ is the magnetostriction constant for FeGa films, and d_{31} (24 pC/N for PVDF) and d_{32} (6 pC/N for PVDF) are the linear anisotropic piezoelectric coefficients of PVDF, and E is the applied external electric field. By applying an electric field on the ferroelectric layer, a strain-induced uniaxial magnetic anisotropy is established in the magnetic layer. For the positive magnetostrictive materials, the uniaxial magnetic anisotropy induced by the strain is parallel to the tensile strain. In our experiment, when the positive voltage is applied on the PVDF substrates, the tensile strain is induced by the electric field along both the easy and the hard axes for the reason of converse piezoelectric effect. As mentioned above, d_{31} of PVDF is much larger than d_{32}. As a result, the extrinsic uniaxial magnetic anisotropy induced by the positive electric field is nearly parallel to the intrinsic uniaxial magnetic anisotropy of FeGa films, i.e., the elongated direction of PVDF. Therefore, the magnetic anisotropy of FeGa films is enhanced under positive polarization of PVDF. The corresponding H_c and M_r/M_s for the FeGa films are increased when the magnetic field applied along the easy axis while decreased when the magnetic field applied along the hard axis. When the negative voltage is applied on the PVDF substrates, the extrinsic easy axis of FeGa films caused by the electric field is rotated 90° with respect to the intrinsic easy axis. In this case, the uniaxial magnetic anisotropy for FeGa films is decreased. H_c and M_r/M_s for the FeGa films are decreased when the magnetic field applied along the easy axis while increased when the magnetic field applied along the hard axis.

The difference of H_c for FeGa/PVDF films under the 400 kV/cm and zero electric fields shows a cosine dependence on the magnetic field orientations, as presented in Fig. 3. When the magnetic field is applied along the $(2n+1)\pi/4$ (n = 0, 1, 2, and 3) rad direction, the variations of H_c are nearly zero which indicate that there are minimum changes of the magnetic properties of FeGa films in response to the external electric field applied on the PVDF layers. The variations in H_c strongly suggest that the generated stress of the PVDF substrates in response to an applied electric field is of anisotropy, which means that the amplitude of CME coupling effect depends on the magnetic field direction.

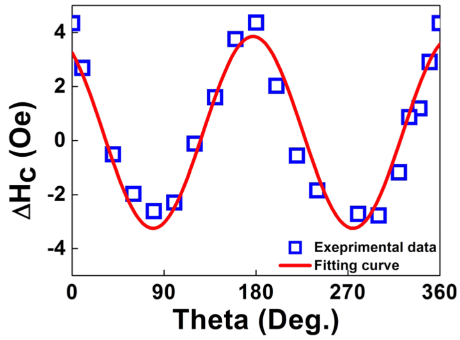

FIG. 3. Differences of coercivity for the FeGa films under the maximum and zero electric fields at various magnetic field orientations.

Therefore, to obtain a larger CME coupling effect, the measuring direction should be taken into account. This work indicates a promising application of the CME effect system as an angle sensor.

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