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Electric-poling-induced magnetic anisotropy and electric-field-induced magnetization reorientation in magnetoelectric Ni/(011) $[Pb(Mg_{1/3}Nb_{2/3})O_3]_{(1-x)}$-$[PbTiO_3]_x$ heterostructure

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This study reports the influence of poling a PMN-PT single crystal laminated structure on the magnetic properties of a 35 nm polycrystalline Ni thin film. During the poling process, a large anisotropic remanent strain is developed in the PMN-PT that is transferred to the ferromagnetic film creating a large predefined magnetic anisotropy. Test results show that operating the PMN-PT substrate in the linear regime following poling produces sufficient anisotropic strain to reversibly reorient the magnetization toward an easy axis oriented 90° to the magnetic easy axis induced during poling. The influence of poling prestress on the magnetic anisotropy field, coercive field and magnetic remanence is discussed. © *2011 American Institute of Physics*. [doi:10.1063/1.3563040]

Magnetoelectric (ME) heterostructures have attracted substantial attention due to the strong coupling between ferroelectric and ferromagnetic phases.[1,2] The converse magnetoelectric effect (CME), i.e., magnetization change by an applied electric field, offers substantial promise for a wide range of applications, such as highly sensitive magnetometers,[3] tunable inductors,[4] nanoscale electromagnet,[5] and magnetic random access memory (MRAM).[6,7] Several studies have reported that electric-field-induced strain,[8–12] especially a large anisotropic strain,[13–16] can alter the magnetic properties of ferromagnetic materials.

The single crystal relaxor ferroelectrics $[Pb(Mn_{1/3}Nb_{2/3})O_3]_{(1-x)}$-$[PbTiO_3]_x$ (PMN-PT, $0 < x < 0.35$) and $[Pb(Mn_{1/3}Nb_{2/3})O_3]_{(1-x)}$-$[PbTiO_3]_x$ (PMN-PT, $0 < x < 0.35$) exhibit large electromechanical coupling and piezoelectric coefficients, which are attractive candidates to electrically control magnetic properties in ME heterostructures. Most studies take advantage of the large linear piezoelectric response in single crystal relaxor ferroelectrics to change the magnetization properties. However, for applications that require magnetization easy axis reorientation, a predefined initial magnetic anisotropy is mandatory. Such requirements can be tailored using magnetocrystalline anisotropy or magnetic annealing.[17] Alternatively, a predefined magnetoelastic anisotropy can be developed by creating a residual stress during the fabrication process due to lattice mismatch or difference in thermal expansion coefficient between the ferromagnetic layer and substrate. In addition to residual stress, single crystal ferroelectrics with specific crystal orientation can produce a large remanent strain during the poling process.[18,19] One can take advantage of the large anisotropic remanent strain produced during ferroelectric poling as an novel alternative source of induced magnetic anisotropy in a thin film Ni layer.

In this paper, we report an electric-poling-induced magnetic anisotropy followed by an electric-field-induced magnetization easy axis reorientation in a 35 nm Ni thin film deposited on (011)-oriented PMN-PT ($x \approx 0.32$) single crystal ferroelectric substrate. A large anisotropic remanent strain takes place during the poling process, which produces an initial predefined magnetic anisotropy in the Ni thin film. After poling, the PMN-PT is electrically excited in the linear piezoelectric strain regime, producing sufficient anisotropic strains to reversibly reorient the magnetization easy axis by 90°. A discussion of the influence of poling and electric-field-induced strains on the magnetic remanence, anisotropy field and the coercivity is provided.

The sample was prepared on a polished $10 \times 10 \times 0.5$ mm^3 (011)-oriented PMN-PT substrate (ATOM OPTICS CO., LTD., Shanghai, China). 10 nm Ti and 50 nm Pt layers are deposited on both sides as electrodes. 5 nm Ti and 35 nm Ni thin films are deposited by e-beam evaporation on the top surface electrode. The PMN-PT substrate is poled after Ti/Ni deposition to take advantage of the poling prestress and induce a permanent magnetic anisotropy.

The in-plane piezoelectric strain response of the PMN-PT is characterized by attaching a biaxial strain gauge on the bottom surface electrode. A unipolar triangular waveform electric field up to 1 MV/m is applied through the PMN-PT thickness at a frequency of 0.01 Hz. The magnetic properties of the Ni thin film are characterized by measuring normalized Kerr rotation *M-H* curves using a longitudinal mode magneto-optical Kerr effect (MOKE) magnetometry, in which the incident laser plane is parallel to the applied magnetic field.

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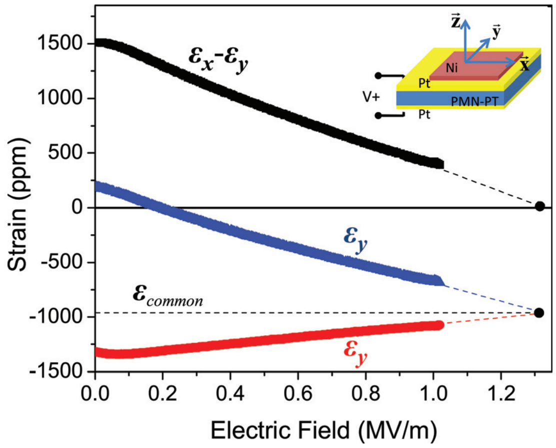

FIG. 1. (Color online) In-plane strains of the PMN-PT substrate along x and y direction and the strain difference $\varepsilon_x-\varepsilon_y$ as a function of applied electric fields; the inset shows the schematic of fabricated Ni/PMN-PT heterostructure.

Figure 1 plots the strains ε_x and ε_y as well as their difference ($\varepsilon_x-\varepsilon_y$) measured at the surface of the PMN-PT substrate as a function of the applied electric field, whereas the inset shows the sample configuration and corresponding coordinate system. The strain of a unpoled state was set to zero strain reference. After applying electric fields larger than the coercive field E_c 0.2 MV/m, large anisotropic remanent strains of 220 and -1320 ppm are present in both x and y directions, respectively, corresponding to 1540 ppm compressive strain difference ($\varepsilon_x-\varepsilon_y$). Afterwards, the poled PMN-PT exhibits a linear piezoelectric response with large anisotropic piezoelectric coefficients $d_{31} = -870$ pC/N and $d_{32} = +285$ pC/N, where subscripts 1 and 2 refer, respectively, to x and y. We note that this specific (011)-oriented PMN-PT produces moderate tensile remanent strain along x and large compressive remanent strain along y, during the poling process. However, when the PMN-PT is excited electrically after poling, the piezoelectric trains are compressive along x and tensile along y for positive electric fields. Thus the ε_x and ε_y curves intersect at 1.32 MV/m with a common strain ε_{common} of approximately -1000 ppm.

Figure 2 shows the normalized magnetic Kerr hysteresis (M-H curves) measured along both x [Fig. 2(a)] and y [Fig. 2(b)] for different applied electric fields. The measurements are performed after PMN-PT poling. As it can be seen in the initial state (E=0 MV/m), a significant magnetic anisotropy exists along the y axis. This easy axis is produced by the anisotropic strains induced during poling. The large M_r/M_s along y and low M_r/M_s along x indicate that the Ni thin film has a predefined magnetization easy axis along the y after poling. This predefined magnetization easy axis is explained in terms of magnetic anisotropy.[16] The magnetoelastic anisotropy K_L defined along x can be expressed as, whereis the saturation magnetostriction of Ni thin film, Y is the Young's modulus of Ni. Since the saturation magnetostriction λ of Ni is negative, positive strain difference ($\varepsilon_x - \varepsilon_y > 0$) due to poling produces a negative magnetoelastic anisotropy K_L, which creates a hard axis along x and an easy axis along y.

Figure 3(a) shows the normalized remanence M_r/M_s and Fig. 3(b) shows the anisotropy field H_a as a function of the electric field for both x and y directions. In both cases, the

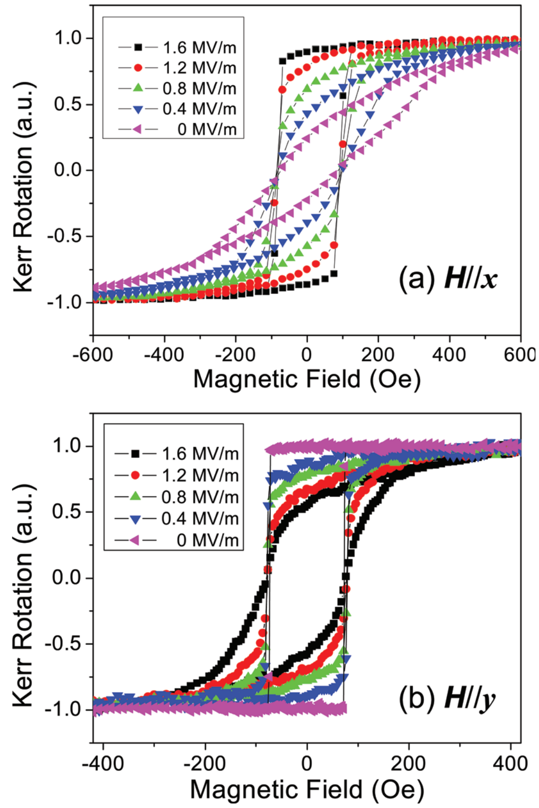

FIG. 2. (Color online) Normalized magnetic Kerr rotation M-H curves measured along (a) x and (b) y directions for different electric fields on poled PMN-PT substrate.

data highlights a change in the magnetic anisotropy when the electric field is applied. An initial magnetization easy axis is observed along y at 0 MV/m, showing large M_r/M_s along y and low M_r/M_s along x. When the electric field is increased, the magnetic anisotropy is reduced, as illustrated by the decrease of H_a along x. As the electric field is further increased to a critical value ($\approx$1.1 MV/m), the magnetization of the Ni thin film returns to an isotropic magnetization state corresponding to an easy plane of magnetization.[16] At this critical electric field, the electric-field-induced strains in PMN-PT counterbalance the strains induced during poling. Further increasing the electric field beyond this critical point, a reorientation of the magnetization easy axis from y to x is observed. The relative strain difference ($\varepsilon_x-\varepsilon_y$) is reversed and a positive magnetoelastic anisotropy along x takes place. Therefore, M_r/M_s along x approaches 100% and a 90° magnetization reorientation is demonstrated. We note that there is a discrepancy between the transition point predicted by the strain data (1.32 MV/m in Fig. 1) and the M_r/M_s data [$\sim$1.1 MV/m in Fig. 3(a)], which is due to the fact that the magnetic and ferroelectric characterization has been performed on two distinct PMN-PT substrates. Single crystal ferroelectric materials are highly history-dependent,[20,21] and therefore different polarization states may produce different remanent strains during poling process.

As can be seen in Figs. 3(a) and 3(b), both remanence M_r/M_s and anisotropy fields H_a evolve gradually with the electric field, demonstrating an electrical control of the magnetic anisotropy as well as reorientation of the magnetic easy axis. Surprisingly, both M-H curves in Fig. 2 indicate that

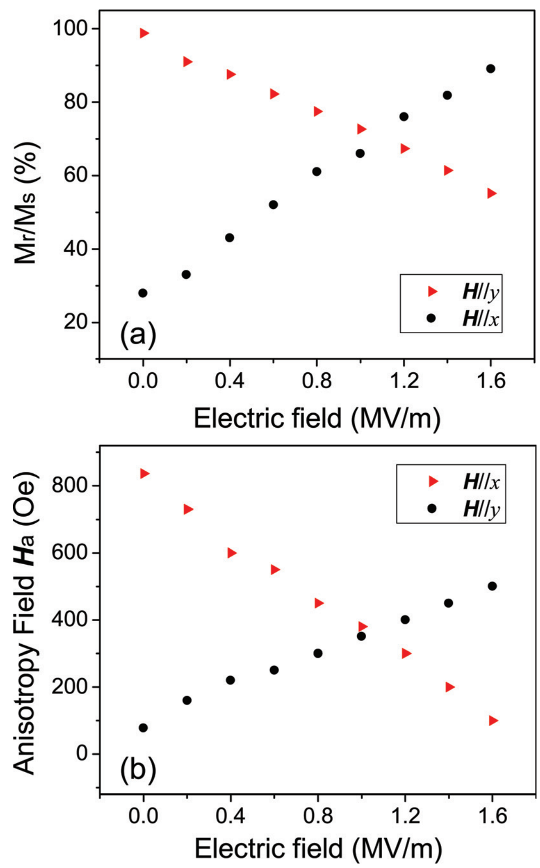

FIG. 3. (Color online) (a) The normalized remanence M_r/M_s, and (b) magnetic anisotropy field H_a as a function of electric fields for both x and y measurement directions.

there is no significant change in the coercivity H_c as a function of electric field, which may contradict some other reports in the literature.[12,15,16,20]

However, compared to our previous results[16] in which a relative small coercivity of ~35 Oe is present for the as-deposited Ni film, the electric-poling-induced the remanent strain/stress change the coercivity of Ni film to ~80 Oe. The pinning of coercivity while operating at the linear piezoelectric regime may be attributed to large biaxial compressive poling prestress. By comparing the absolute value of the strains ε_x and ε_y rather than the relative strain difference, we can hypothetically assume that after poling, a large biaxial common compressive strain component ε_{common} on the order of 1000 ppm is superimposed with the relative electric-field-induced anisotropic strain, as indicated in Fig. 1. This large biaxial compressive strain ε_{common} may significantly modify the magnetic property of the Ni thin film, in which the coercivity cannot change under such a large prestress condition.

Regarding to the magnetic anisotropy change due to electric-field-induced anisotropic strains, both anisotropy field H_a and coercivity H_c change are reported. However, H_a and H_c are related to distinct magnetization reversal mechanisms.[17] The former is related to magnetization reversal along a hard axis with coherent reversal and the latter related to magnetization reversal along a magnetization easy axis with incoherent reversal. Further first principle calculations or micromagnetic simulation may be required to analyze the change in H_a and H_c, respectively. Nonetheless, it appears that for the large electric-poling-induced strain condition, H_c is not altered in the electric field ranges studied.

We report a novel approach to induce a predefined magnetic anisotropy in ME heterostructure based on remanent anisotropic strain induced during the ferroelectric poling of a (011) single crystal PMN-PT substrate. The poling prestress produces a large permanent magnetization easy axis in the ferromagnetic film. After poling, the large electric-field-induced anisotropic strain can overcome the poling-induced remanent strain, reorienting the magnetization easy axis by 90°. The electric-poling-induced magnetic anisotropy and electric-field-induced magnetization reorientation provides a new approach for magnetic memory and spintronics applications.

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