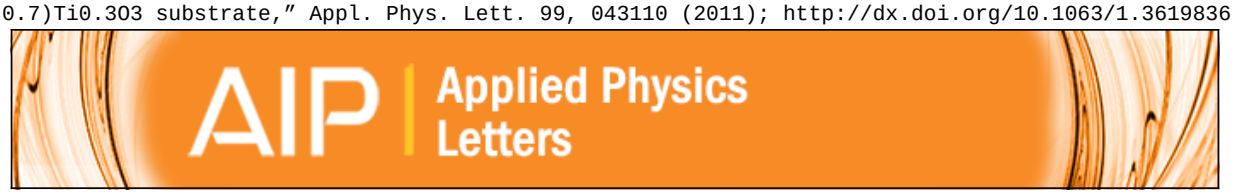

Giant electric field controlled magnetic anisotropy in epitaxial BiFeO3-CoFe2O4 thin film heterostructures on single crystal Pb(Mg1/3Nb2/3)0.7Ti0.3O3 substrate

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Giant electric field controlled magnetic anisotropy in epitaxial $BiFeO_3$-$CoFe_2O_4$ thin film heterostructures on single crystal $Pb(Mg_{1/3}Nb_{2/3})_{0.7}Ti_{0.3}O_3$ substrate

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We have deposited self-assembled $BiFeO_3$ (BFO)-$CoFe_2O_4$ (CFO) thin films on $Pb(Mg_{1/3}Nb_{2/3})_{0.7}Ti_{0.3}O_3$ (PMN-PT) substrates and studied the change in magnetic anisotropy under different strain conditions induced by an applied electric field. After electric field poling, we observed (i) giant magnetization change: magnetization of original CFO phase is three times larger than that of strained one and (ii) magnetic force microscopy line profiles that exhibited significant change in the CFO magnetic domain response in accordance to magnetization-field (M-H) loops. Together, these results demonstrate good control of the magnetic properties of CFO via an electric field induced strain. © *2011 American Institute of Physics*. [doi:10.1063/1.3619836]

Perpendicular recording technology has recently been introduced in hard disk drives, as the traditional longitudinal recording technology is approaching its storage limit due to superparamagnetic effects.[1] Large magnetic anisotropy is thus necessary to guarantee bit stability, while preserving high density storage capability. Controllable magnetic anisotropy is desirable to enable magnetic recording with a weak field. The hard magnetic material $CoFe_2O_4$ (CFO) has been studied in detail due to its high coercivity (~5400 Oe) and large magnetic anisotropy, as well as high magnetostriction.[2] Another important property of CFO is that it can be grown epitaxially together with perovskite structured phases due to fairly close lattice matching, forming well-defined self-assembled structures with CFO nano-pillars embedded in a perovskite matrix.[3–5] These self-assembled nano-structures allow much larger interfacial areas between ferromagnetic and ferroelectric phases, thereby promising much larger magnetoelectric (ME) coupling effects.[6–9] Zavaliche *et al.*[1] then reported the synthesis of $BiFeO_3$ (BFO)-CFO self-assembled thin films on $SrTiO_3$ (STO) substrates and measured two different electrically switchable perpendicular magnetic states at ambient conditions. Zhao *et al.*[10] also reported magnetic switching of CFO phase in BFO matrix after application of an electric field by photoemission electron microscopy and x-ray magnetic circular dichroism. However, the difficulty in poling the entire sample for thin films on STO substrate makes it very complicated to measure the total magnetization change induced by strain. Adoption of piezoelectric single crystal substrates then becomes a solution.

Here, we report the deposition of BFO-CFO self-assembled thin film on $Pb(Mg_{1/3}Nb_{2/3})_{0.7}Ti_{0.3}O_3$ (PMN-PT) single crystal substrate. The combination of the strain-sensitive magnetic anisotropy of CFO nano-pillars and the giant d_{33} values of PMN-PT crystal substrates[11–13] results in a giant magnetic anisotropy change in the heterostructured films.

We deposited $65BiFeO_3$–$35CoFe_2O_4$ composite epitaxial thin films using pulsed laser deposition on (001) oriented PMN-PT single crystal substrates which were obtained from the Shanghai Institute of Ceramics Chinese Academy Sciences. The magnetic hysteresis loops were measured with a Lakeshore 7300 series vibrating sample magnetometer (VSM) system. Magnetic force microscopy (MFM) in the tapping-lift mode was used to image the magnetic domain and to detect the change of magnetic anisotropy in the out-of-plane direction for CFO phase, using a Veeco SPI 3100.

Fig. 1(a) shows XRD line scans for the BFO-CFO/PMN-PT heterostructures both before and after poling of the PMN-PT substrate. The PMN-PT substrate had a tetragonal (T) structure, and hence both (002) and (200) zones were evident. The thicknesses of the PMN-PT substrates were 0.5 mm, and a voltage of 250 V (E = 5 kV/cm) was used to pole the whole samples after deposit Au electrodes on both sides at room temperature. An obvious left-shift of the CFO and BFO peaks can be seen after the poling process, which resulted from an elongation of the *c*-axis of both the CFO and BFO due to electric field induced domain redistribution within the PMN-PT. Fig. 1(b) shows scanning electron microscopy (SEM) result of a typical self-assembled BFO-CFO thin film on PMN-PT substrate which was very similar to previous reported BFO-CFO on STO substrates.[3]

The (002) and (200) peaks for PMN-PT in Fig. 1 correspond to *c* and *a* ferroelectric domains, respectively. Please note after poling that the intensity of the (002) peak was significantly increased while that of the (200) was decreased.

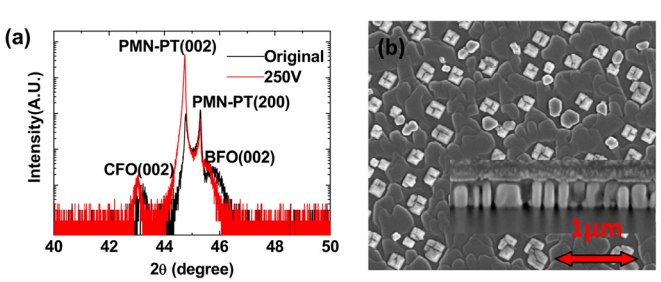

FIG. 1. (Color online) (a) XRD line scan of BFO-CFO on PMN-PT before and after polling and (b) SEM image of the BFO-CFO layer, where the inset shows a cross-sectional view.

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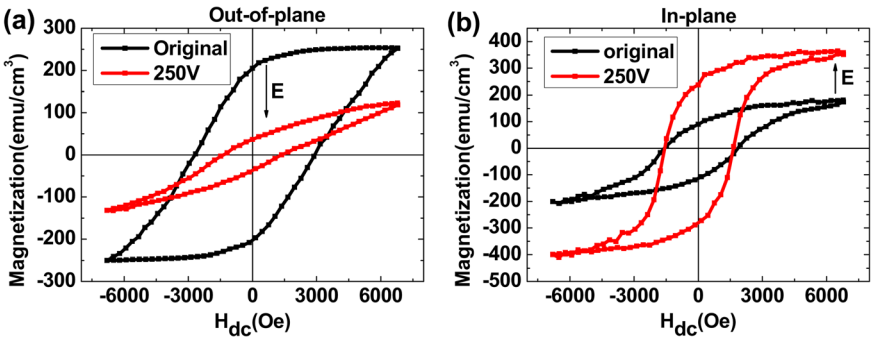

FIG. 2. (Color online) Magnetic hysteresis loop comparison before and after polling in the out-of-plane (a) and in-plane (b) direction.

This demonstrates that most of the *a*-domains were transformed into *c*-domains, resulting in a near single c-domain state oriented in the out-of-plane direction. From the position of the peaks, the crystal lattice parameters of PMN-PT were determined to be $(a_t, c_t) = (4.032, 4.083)$. The strain induced by the domain reorientation was thus $\varepsilon = (a - c)/a = 0.0126$. In addition, it can be seen that the CFO (002) peak was shifted from 43.242° to 43.058°, which corresponds to a strain of $\varepsilon_{001} = 0.426\%$ in the CFO nano-pillars along the c-axis. This huge change in the CFO crystal lattice parameters will influence the magnetization of the CFO phase in the BFO-CFO self-assembled thin films, via the high magnetostriction of CFO. We then annealed the heterostructures at 200 °C in air for 0.5 h to confirm that their original condition could be recovered. Not only did the CFO and BFO peaks shift back to their original positions but also the relative peak intensities of the PMN-PT (002) and (200) zones were restored to their original values. These results unambiguously demonstrate a correlation of the field induced lattice parameter changes in the PMN-PT substrate with corresponding ones in the two phase epitaxial layers of magnetostrictive CFO nano-pillars and BFO matrix.

Next, we need to ask, how were the magnetic properties affect by poling and the field induced structural changes? Fig. 2 shows the M-H loops of a BFO-CFO two phase epitaxial composite thin film both before and after poling. The data show both out-of-plane and in-plane directions. Fig. 2(a) shows the magnetization as a function of applied magnetic field along the out-of-plane direction in both conditions. We can notice that the remnant magnetization was reduced by ~80% after poling. This demonstrates that only one fifth of the magnetic domains remains aligned in the out-of-plane direction. Such dramatic changes are notably larger than that previously reported for (La,Sr) MnO_3 (LSMO) epitaxial layers grown on $BaTiO_3$ (BTO)[14] and BTO-CFO and BFO-CFO on STO substrates.[15–17] The magnetic coercive field was also significantly decreased by poling: H_c was only about 50% of its value before poling. Correspondingly, Fig. 2(b) shows the changes in the M-H curve along the in-plane direction due to electric field poling. The remnant magnetization was significantly increased by poling: ~3x the value before poling. Together, these results show that the strain induced by electric field poling influences the magnetization along both in-plane and out-of-plane direction in opposite ways.

Fig. 3(a) shows a schematic of our self assembled BFO-CFO thin film heterostructures on PMN-PT. Illustrated by arrows are the stresses on the CFO nano-pillars along the in-plane and out-of-plane directions. These stresses mainly come from the reorientation of the ferroelectric domains of the PMN-PT substrate, not like BFO-CFO on STO,[1,10] where only the BFO matrix would contribute to the strain on CFO. After poling, most of the *c*-domains are aligned out-of-plane. Thus, we can expect an elongation of the CFO nano-pillars as a result of a tensile strain applies along the out-of-plane direction. Correspondingly, CFO should have a compressive strain in the in-plane direction as a result of the compressive stress from the PMN-PT substrate: but unlike the out-of-plane direction where the strain is uniaxial, the strain in this case is biaxial. Thus, one might anticipate larger strain effects along the out-of-plane direction. The CFO nano-pillars have a large negative magnetostriction $(\lambda_{001} = -350 \times 10^{-6})$.[1] Consequently, after poling, one can expect a large decrease in the magnetization of the CFO nano-pillars, as a result of a tensile strain. The magnetic domains are more likely to align in the in-plane direction, giving rise to a lower coercive field in the out-of-plane. These possibilities were confirmed by M-H measurements.

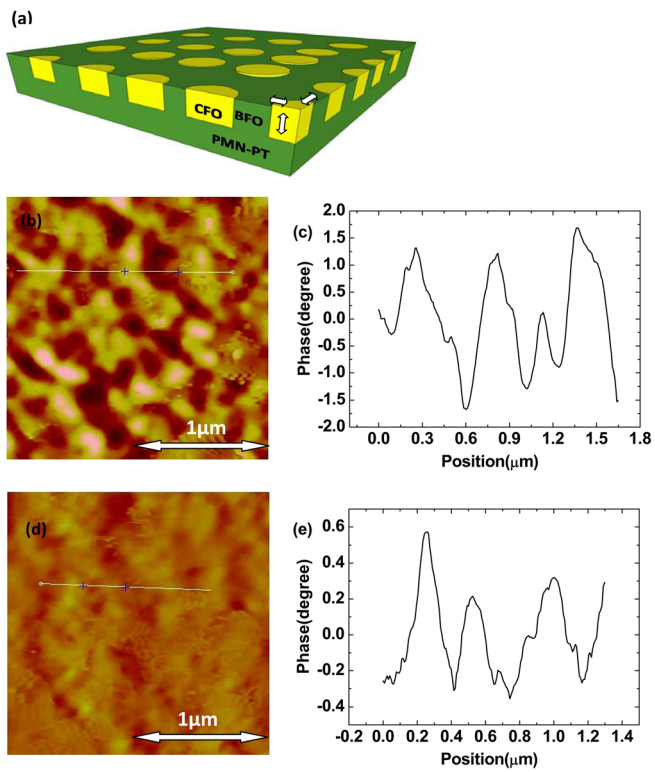

FIG. 3. (Color online) (a) Schematic of the strain condition during the poling process; MFM result of the BFO-CFO nanostructure before (b) and after polling (d); and line scans extracted from the MFM images before (c) and after (d) poling.

Finally, we used MFM to analyze the magnetic domain response in individual CFO nano-pillars. Figs. 3(b) and 3(d) show the MFM images of BFO-CFO thin films before and after poling, respectively. The size of the dark and light contrast areas was about 100 nm, the same size as shown in SEM image (see Fig. 1). In Fig. 3(b), one can clearly see dark and light contrast areas which corresponded to the magnetic CFO domains. However, after poling, the contrast in the MFM images was decreased, and a clear interface between dark and light contrast areas was not evident. Please note that the MFM images were obtained using a tapping mode, without applied DC magnetic (the magnetization of the MFM probe is quite small) or electric fields. One can then conclude that variations in the remnant magnetization of the BFO-CFO thin films in the out-of-plane direction make the dominant contribution to the MFM image contrast. Thus, these MFM results indicate that the remnant magnetization is decreased notably by poling, which was confirmed by the M-H measurements. Next, Figs. 3(c) and 3(e) show line profiles extracted from the MFM images under different poling/strain conditions, where the profile intensity is a measure of the magnetization. In these line scans, small "hills and valleys" can be seen at regular intervals, which are indicative of the intensity and direction of the magnetic lines of force on the surface of the sample. From the line scan of the sample before poling, the intensity can be seen to vary from $-1.7°$ to $+1.5°$; whereas in the poled condition, the intensity varied from $-0.3°$ to $+0.3°$. From these values, the decrease in magnetization by poling can be estimated to be $\sim$20% of the original value before poling, which is in agreement with the calculated ratio from the M-H measurements.

These results demonstrate that we can reversibly use strain to control the magnetic anisotropy of the CFO nano-pillars in the BFO-CFO layer. Accordingly, it is relevant to note that this giant electric field controlled magnetic anisotropy might have importance to perpendicular recording. One could electrically pole a single CFO nano-pillar area in the BFO-CFO two phase layers. Then, the magnetic field required to magnetize the CFO domains along the out-of-plane direction would be notably lowered. The area could then be depoled electrically, resulting in a large increase in the vertical magnetization.

In summary, we have deposited vertically aligned BFO-CFO self-assembled thin films on PMN-PT substrates. Electric fields applied to the PMN-PT have been shown to affect the lattice parameters of CFO nano-pillars. Correspondingly, a significant decrease was found in both the remnant magnetization and coercive field for the CFO nano-pillars in the out-of-plane direction. Similar changes in magnetization of the CFO nano-pillars were found in the MFM images under the same condition. These findings demonstrate a giant electric field controlled magnetic anisotropy, which may have important application in recording devices.

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