Electric-field controlled magnetic reorientation in exchange coupled CoFeB/Ni bilayer microstructures

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Abstract. A novel strain-mediated composite multiferroic system is investigated in this work. The system is composed of magnetostrictive microstructures made of a ferromagnetic bilayer of negative magnetostrictive Ni and positive magnetostrictive CoFeB fabricated on a ferroelectric Pb(Mg¹/₃Nb²/₃)₀.₆₉Ti₀.₃₁O₃ (PMN-PT) single crystal substrate. When an electric field is applied across the PMN-PT substrate, magnetization reorientation occurs in the microstructures that are predominantly initialized into flux-closure states. An x-ray magnetic circular dichroism-photoemission electron microscope is used to experimentally visualize the electrically induced changes of the magnetization state of the individual layers as well as the exchange coupling between the two layers. Such heterostructures with tunable magnetoelectric properties have potential for new magnetic memory and sensor applications.

Keywords: Multiferroics, magnetostriction, straintronics, energy-efficient magnetoelectrics.

1. Introduction

Multiferroic systems hold great promise for energy-efficient control of magnetization in applications such as memory and logic devices [1], actuators and transducers [2] and other high-frequency magneto-electric devices [3]. In the past decade, much progress has been made in controlling magnetization states of ferromagnetic microstructures grown on ferroelectric substrates through applied electric fields at room temperature.
Particularly interesting are materials systems with a thin ferromagnetic layer deposited onto a ferroelectric PMN-PT substrate. So far, much has been reported about the electrically induced strain-driven magnetization reorientation in negative magnetostrictive Ni microstructures on PMN-PT [4-6]. Furthermore, positive magnetostrictive CoFeB has also drawn substantial interest for its significant tunneling magnetoresistance (TMR) in magnetic tunnel junctions of strain-mediated multiferroic random access memory (SME-RAM) [7, 8]. By combining the ease of electrically reorienting magnetization in Ni microstructures [4] and CoFeB’s potential for memory applications [9], this work examines the magnetization reorientation in Ni/CoFeB microstructures when subjected to an applied electric field. We investigate the interplay between the two layers when deposited on the PMN-PT substrate, with the goal of developing more sophisticated magnetostrictive composite structures with tunable magneto-electric coupling.

2. Experimental Design

Several Ni/CoFeB samples of various thickness and lateral dimensions combinations were prepared in a cleanroom facility using conventional nanofabrication processes [4, 5, 10]. After the samples are wire-bonded to leadless chip carriers (LCC), they are magnetized in-plane to initialize the magnetization states. To visualize and characterize the layer-dependent magnetic domain states in the microstructures before and after applying an electric field, the samples are imaged using an x-ray magnetic circular dichroism-photoelectron emission microscope (XMCD-PEEM) at beamline 11.0.1.1 of Advanced Light Source (ALS), Lawrence Berkeley National Laboratory (LBNL). Element specific magnetic contrast images at different magnitudes of the applied electric field were collected for individual microstructures for comparison of strain-induced magnetization reorientation in each ferromagnetic layer.

2.1. Sample Preparation

After dicing the [011] cut polished [Pb(Mg1/3Nb2/3)O3]1−x[PtbTiO3]x (PMN-PT, x ≈ 0.31) single crystal into 1 cm × 1 cm × 500 μm thick pieces, we use a Matrix Plasma Asher to remove organic residues from the substrates with oxygen plasma. 5 nm Ti and 50 nm Pt thin films are deposited by e-beam evaporation via a CHA Solution electron beam evaporator on both surfaces of the PMN-PT substrate. Each film is deposited at a rate of 0.3 Å/s. The Pt films serve as top and bottom electrodes so that the substrate can be actuated in a parallel plate capacitor geometry. Microstructures ranging from 1 μm to 4 μm in length are defined and written by electron beam lithography in the PMMA A2 resist coating the top Ti/Pt surface. Before CoFeB/Ni sputtering deposition, the PMN-PT is electrically prepoled with an electric field of 0.4 MV/m in the [011] direction of the PMN-PT to minimize the residual strain. Next, 5 nm Ta film and bilayers of CoFeB and Ni, each with a thickness of either 2 nm or 15 nm, are grown by magnetron sputtering with a base pressure of 3×10⁻⁸ Torr, capped with a 2 nm Pt layer to prevent oxidation. The roughness of the sample is measured with an atomic force microscope, showing an Rₜ of 0.6 nm. Then the magnetic patterns are lifted off using heated NMP solution at 50°C. The four corners of the samples are cut off to fit in the LCC while having the [100] and [01-1] crystal directions of the PMN-PT substrates pointing along the diagonal directions of the LCC. The reason for such alignment is to have the electrically-induced magnetoelastic easy axes of Ni and CoFeB along the diagonals of the PEEM micrographs for the ease of analysis. The devices are then wire-bonded such that the top surface is grounded and the bottom is connected to a bipolar voltage power supply. They are then magnetized by an initialization field Hᵢ of 0.4 T applied in-plane along the direction bisecting [100] and [01-1] crystallographic directions of the PMN-PT, as illustrated in Figure 1(a).

2.2. Magnetic contrast study by XMCD-PEEM
The magnetic properties of arrays of microscale magnetic square of CoFeB and Ni layers of different thickness combinations are investigated by XMCD-PEEM. XMCD-PEEM is an element-specific magnetic contrast imaging technique. It probes the secondary electrons emitted from the sample surface following the absorption of the incoming circularly polarized x-ray photons, known as the XMCD effect. Element specificity of the PEEM imaging makes it a powerful tool to investigate individual layers in a magnetic multilayer system. For this experiment, the magnetic domain visualization in each layer of the bilayer is achieved by separately probing at the photon energy of the Ni $L_3$ edge for the Ni layer or the Fe $L_3$ edge for the CoFeB layer. In this way the magnetic domains, as indicated by the magnetic contrast, in the two different material layers can be analyzed separately.

Figure 1(a) is a schematic of the PEEM microscope and the position of the sample with respect to the incident x-ray and the initialization magnetic field. Figure 1(c) shows the device (Figure 1(b)) mounted onto a PEEM sample holder that allows in-situ electric-field application.

![Figure 1](image)

**Figure 1.** (a) Schematic of the XMCD-PEEM imaging setup. The orientation of the device with respect to the incident x-ray beam is illustrated. Prior to imaging, an initialization magnetic field ($H_\text{init}$) was applied. The crystallographic orientations of the [011]-cut PMN-PT are highlighted, with [100] being the compressive strain axis, and [01-1] being the tensile strain axis. (b) A completed device on an LCC with crystallographic directions of [011] cut PMN-PT and the orientation of the magnetic field for magnetization state initialization ($H_\text{L}$) marked. The substrate is mounted on a piece of Si wafer of 0.3 mm thick so that the sample surface is at the proper height (0.8 mm from the LCC surface) for XMCD-PEEM imaging. The top surface of the multiferroic heterostructure is grounded (as indicated by “GND”, and the bottom is connected to a hot wire (as indicated by “+V”). (c) A device is mounted onto a PEEM-3 sample holder at beamline 11.0.1.1 of ALS, LBNL.

### 2.3 Magnetization state in the bilayer systems before and after applying electric field

Prior to applying the electric field, the magnetic domain states in the microsquares tend to form a flux-closure state (Landau state [11]) due to the competition between demagnetization (dipolar) energy, also known as shape anisotropy energy, and exchange energy once the initialization magnetic field is removed. Figure 2 shows the representative PEEM micrographs of a microsquare consisting of 2 nm thick Ni and 2 nm thick CoFeB, taken at both the Ni $L_3$ and Co $L_3$ edge. XMCD-PEEM images of both Ni and Fe are individually taken to show the magnetization state in the two layers, respectively. We mainly focus on how these microstructures evolve, from their initial state at zero field, after the electric field is applied up to 0.8 MV/m. This is done following a recent study where individual Ni microsquares on PMN-PT have been shown to form a two-domain state with a magnetic domain wall along the diagonal of the microsquares separating the two domains, as a sufficient strain is induced [4].
After applying an electric field through the PMN-PT thickness, a compressive strain is generated along the [100] crystallographic direction of the PMN-PT substrate, and a tensile strain along the [01-1] direction. The strain is transferred from the ferroelectric substrate to the top electrode and ferromagnetic bilayer and the magnetic domain configuration in the microstructures is modulated by the magnetoelastic anisotropy energy. Due to the inverse magnetostrictive effect in such multiferroics heterostructures [12], it is known that for positive magnetostrictive CoFeB, the magnetic moments tend to realign with the tensile strain direction, whereas the magnetic moments in negative magnetostrictive Ni are expected to align toward the compressive strain direction. However, in this study both materials are present on the PMN-PT substrate, coupled to each other via exchange coupling. Figure 2b shows what is obtained in this study. The magnetization state for the two ferromagnetic layers of the same microsquare in Figure 2a after the field is applied are identical, resulting in a new global magnetic easy axis aligned with the one expected for the CoFeB layer.

Figure 2. Representative layer-resolved PEEM micrographs and schematics of magnetic contrast of a microsquare of 2 μm in length and consisting of 2 nm thick Ni and 2 nm thick CoFeB. (a) Before applying the electric field. (b) After applying an electric field of 0.72 MV/m, the magnetic contrast level is reduced when compared to that in (a), implying the easy axis is in the process of rotating towards the [01-1] direction. The blue line is in the direction of tensile train.

3. Results and Discussions
The correspondence between the contrast in the PEEM images of both Ni and CoFeB layers before and after applying the electric field highlights the strong exchange coupling behaviour between the two materials at initialization. Table 1 summarizes the net in-plane magnetic anisotropy induced by the anisotropic piezo-strain in each of the samples. For positive magnetostrictive CoFeB, the magnetic moments realign to the tensile strain direction, whereas the negative magnetostrictive Ni aligns toward the compressive strain direction, in agreement with what reported in literature so far [4, 6, 12]. However, when both CoFeB and Ni are present, the magnetic anisotropy of the composite tends to favour magnetization reorientation in the tensile strain direction due to the CoFeB, except when a thick (15 nm) Ni is employed.

Table 1. Magnetic easy axis reorientation direction upon applying electric field up to 0.8 MV/m.

| Sample | Material stack                  | Direction of new easy axis | Dominant Material |
|--------|---------------------------------|----------------------------|-------------------|
| 1      | CoFeB (2nm) / PMN-PT            | Tensile strain [01-1]      | CoFeB             |
| 2      | CoFeB (2nm) / Ni(2nm) / PMN-PT  | Tensile strain [01-1]      | CoFeB             |
| 3      | Ni (2nm) / CoFeB (2nm) / PMN-PT | Tensile strain [01-1]      | CoFeB             |
| 4      | CoFeB (2nm) / Ni (15nm) / PMN-PT| Compressive strain [100]   | Ni                |
| 5      | Ni (2nm) / CoFeB (15 nm) / PMN-PT| Tensile strain [01-1]      | CoFeB             |

4. Conclusions
In summary, we report the strong influence of exchange coupling on the electrical magnetization reorientation in a ferromagnetic bilayer on top of a piezoelectric substrate, highlighting the potential for hybrid ferromagnetics in composite multiferroics for new generation magnetoelectric applications.
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