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Magnetoelectric coupling in micropatterned BaTiO₃/CoFe₂O₄ epitaxial thin film structures: augmentation and site-dependency

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Abstract

The prominent magnetoelectric (ME) effect in ME composites usually originates from interfacial strain-mediated coupling between ferroelectric and ferromagnetic components. While ME composite bilayer thin films have been proposed for microdevice applications, the ME coupling is usually diminished because of the substrate clamping effect. Here, we apply simple CMOS/MEMS compatible top-down fabrication techniques to ferroelectric BaTiO₃ (BTO) and ferromagnetic CoFe₂O₄ (CFO) bilayer epitaxial thin films to control the substrate clamping. We found augmented ME coupling in micro-patterned bilayer thin films compared to the as-deposited films due to reduced substrate clamping. In addition, site-dependent ME coupling within the microstructure was observed. Larger ME coupling was obtained near the edge of the microstructure, and site-dependent ferroelectric imprints were observed on the micropattern. This can be attributed to the non-uniform substrate clamping across the film and the strain gradient developed in the BTO layer due to the magnetostriction of CFO. Our findings provide additional insights to the design of micro- and nanoscale devices based on 2-2 ME composite thin films.

Keywords: microfabrication, magnetoelectric effect, clamping effect, thin film, strain gradient

Multiferroic and magnetoelectric (ME) materials have attracted a great deal of attention for their ability to couple magnetic and electric properties ¹⁻³. Owing to the coupled magnetic and electric order parameters, ME materials provide potential to control the electric polarization by an external magnetic field (direct magnetoelectric effect) or to control the magnetic properties by an electric field (converse magnetoelectric effect). For this reason, several micro- and nanodevices for electronic ^{4, 5}, spintronic ⁶, magneto-electrochemical ^{7, 8}, and biomedical ⁹ applications have been suggested using magnetoelectric materials. Particularly, ME composite thin films where ferroelectric and ferromagnetic phases coexist, have been widely utilized because of the rare existence of single phase magnetoelectric materials ¹⁰. For such ME composite films, ME coupling is usually an outcome of strain-mediated interfacial interaction between ferroelectric and ferromagnetic materials ^{11, 12}. However, in 2-2 type ME bilayer thin films where layer interfaces are parallel to the substrate, the ME coupling is usually diminished because of the substrate clamping effect ¹³.

To enhance the ME coupling in thin films, several strategies have been proposed such as strain engineering ¹⁴, thickness/stacking order variation ^{15, 16}, superlattices ¹⁷, and tuning the orientation of the film ¹⁸. Nonetheless, those methods cannot lift the underlying problem of the weak ME coupling, i.e. substrate clamping. Zheng *et al.* proposed 1-3 type thin film nanostructures where ferromagnetic nanostructures are embedded in ferroelectric matrix ¹⁹. In this type of 1-3 structure, interfaces between the ferroelectric matrix and the ferromagnetic nanostructures are vertical to the substrate, so the interfacial strain transfer is free from the substrate clamping ^{13, 20}. Several different thin film nano-architectures including 1-3, 0-3 type epitaxial thin films ²¹⁻²³ and nanodots ^{24, 25} have been also proposed to overcome substrate clamping and to enhance ME coupling. In spite of enormous efforts on thin film nano-architectures, electrical leakage through the conductive ferroelectric-ferromagnetic interface and uncontrollable nucleation and deposition process hinder these nano-architectures to be used for practical device applications ^{26, 27}.

Meanwhile, several studies have reported that the substrate clamping can be highly reduced by downsizing the lateral length of the thin films using focused ion beam (FIB) milling. Interestingly, extraordinary ferroic properties, such as increase of piezoelectric response ²⁸, dynamic ferroelastic domain movement ²⁹ and tetragonal-rhombohedral phase transition ³⁰, which are absent in as-deposited thin films, have been observed when substrate clamping is released. In this regard, CMOS and MEMS compatible top-down fabrication techniques can be introduced to control the substrate clamping as films must be patterned into microstructures

for device applications in most cases. With sequential processes of standard lithography and dry/wet etching, specific shape and size of the patterns can be designed and unusual functionalities could be expected in these patterns.

Herein, we employed top-down fabrication technique on 2-2 type ME composite thin film to investigate the device size effect (substrate clamping effect) on ME coupling. We chose BaTiO₃ (BTO) and CoFe₂O₄ (CFO) as ferroelectric and ferrimagnetic layers, respectively. Owing to the large magnetostriction of CFO and Curie temperatures well above room temperature for both materials, BTO/CFO composite have a potential to give large strain-mediated ME response¹⁹, which is desired for room temperature ME devices. Using pulsed laser deposition and top-down fabrication technique, we produced different sizes of micro-scale BTO/CFO epitaxial films on (001) MgO substrate (Fig. 1(a)). By reducing the lateral size of the film, we observed enhanced ME coupling due to released clamping effect, as expected. Furthermore, we surprisingly found site-dependent ME coupling inside the structures when an external magnetic field is applied (Fig. 1(b)). Opposite ferroelectric imprints were obtained depending on measurement sites and this may be attributed to magnetostrictively induced strain gradients in the microstructures. As most of the electronic and magnetic devices are desired in micro- and nanoscale, our results imply that the non-uniform internal field generation needs to be considered when designing small scale ME devices.

BTO(13nm)/CFO(20nm) bilayer thin films were grown on (001) oriented MgO single crystal substrates using pulsed laser deposition (PLD). X-ray diffraction (XRD) theta-2theta scan reveals that the films are epitaxially deposited on the substrate. Because of the large lattice mismatch between CFO and BTO layers (~5.1 %), BTO is under large tensile strain and exhibits broad diffraction peak owing to (002) and (200) mixed domains^{31, 32} (Fig. 2(a) and 2(b)). For the atomic scale analysis of the film, high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM), also referred to as Z-contrast imaging, was performed. A representative cross-sectional HAADF-STEM image of the BTO film on top of the CFO layer is shown in Fig. 2(c). The image was acquired along the BTO perovskite pseudocubic direction and exhibits atomic columns with two distinct intensities, with the Ba atomic columns appearing brighter than the Ti atomic columns. While the BTO/CFO interface is smooth, misfit dislocations form due to the large lattice mismatch, and are identified as butterfly-like features in the in-plane strain map, which was obtained by geometric phase analysis (GPA) (Fig. 2(d)). The in-plane strain map shows that the BTO layer is under tensile strain near the interface and relaxes gradually towards the surface. The measured in-plane strain difference between the bottom and the top of the BTO film is about 1.5-2.0 % depending on

the region (Fig. S1(b)). The out-of-plane strain map exhibits the opposite trend (Fig. S1(c)). Thus, the out-of-plane BTO lattice parameter is found to be the smallest at the interface and largest at the film surface. In some areas, dislocations were also observed in the middle of the BTO layer, resulting in an abrupt relaxation of the strain (Fig. S2). All these observations point to a mixed domain configuration in the BTO layer, with *a*-domains near the BTO/CFO interface and *c*-domains near the film surface. This is further corroborated by ferroelectric polarization mapping^{33,34}. A polarization map, superimposed on a HAADF-STEM image of the BTO film, calculated from the displacement of the Ti cation in the image plane from the center of its four Ba neighbors, is shown in Fig 2(e)-2(g). Owing to the *a*-domain configuration, the Ti-cations are displaced along the in-plane direction near the interface (note the large deviation along the *x* direction, Fig. 2(f)). On the other hand, the Ti-cations near the film surface are shifted along the out-of-plane direction, which is typical for a *c*-domain configuration (large deviation along the *y* direction, Fig. 2(g)).

In order to see the device size effect on ME coupling, arrays of square disks with different lateral lengths of 3, 4, 5, 10, and 20 μm were fabricated using UV photolithography and Ar^+ ion dry etching (Fig. 1(a)). The scanning electron microscopy (SEM) image shows arrays of 3- μm structures after the final step of the fabrication and the atomic force microscopy (AFM) image exhibits smooth surface before and after the fabrication (Fig. S3), revealing that the top surface of the film is uninfluenced by the fabrication process. We then performed piezoresponse force microscopy (PFM) switching characteristic measurements on fabricated structures with and without external in-plane DC magnetic fields. Fig. 3(a)-3(d) show amplitude butterfly loops and phase hysteresis loops of the as-deposited film and the 3- μm structures under the existence of the external magnetic field. When an external magnetic field of 500 Oe along [100] direction is applied, neither a notable shift of the coercive fields nor enhancement/reduction of the amplitude has been observed in the as-deposited film (Fig. 3(a) and 3(b)). In contrast, for the 3- μm structures, notable coercive voltages shift and amplitude increase have been observed (Fig. 3(c) and 3(d)). The positive and negative coercive fields shifted from 3.37 V to 4.24 V and from -2.61 V to -3.02 V, respectively. The asymmetric shift of the PFM loops under the magnetic field indicates the emergence of an internal electric field in the BTO layer, most probably generated by the strain induced by the magnetostriction of the CFO layer³⁵⁻³⁷. We have measured PFM loops on several different 3- μm structures and all measurements showed similar behaviors. Interestingly, this ME coupling characteristic in PFM loops was reversible; when the magnetic field was turned off, the amplitude and phase loops turned back to their original state (black). This demonstrates enhanced ME coupling in 3- μm

structures, which is absent in the as-deposited film. For a quantitative comparison, we calculated the local ME coefficient of each size of the structures (Fig. S4)³⁵. As the lateral length of the structure gets smaller, ME coupling coefficient increased gradually.

Just like enhanced ferroelectric and piezoelectric properties in declamped films^{29, 38, 39}, the augmentation of the ME coupling in the microstructures here can be attributed to the reduced substrate clamping. From continuum mechanics, the degree of substrate clamping and the in-plane effective stress distribution can be expressed as a function of the film lateral size^{40, 41}. In Fig. 3(e), the degree of the substrate clamping on BTO layer has been calculated as previously reported^{29, 30}. For a 3- μm structure, substrate clamping is decreased down to 59% from that of as-deposited film. Meanwhile, magnetostrictive-induced stress distribution along the horizontal direction in BTO-CFO interface can be determined as follows:

$$\sigma_{11,B} = \frac{1}{h_{BTO}} \left(\frac{E_{CFO}}{1-\nu_{CFO}} h_{CFO} \lambda_{(100)} H_{app} \right) e^{-k(l-x)} \quad (1)$$

$$k = \frac{3E_{MgO}}{2(1+\nu_{MgO})h_{MgO}} \left(\frac{1-\nu_{CFO}}{E_{CFO}h_{CFO}} + \frac{1-\nu_{BTO}}{E_{BTO}h_{BTO}} \right) \quad (2)$$

where h is thickness of the film, E is the Young's modulus, ν is the Poisson's ratio, λ is the magnetostrictive coefficient, H is the applied magnetic field, l is the half size of the film's lateral length, and x is the horizontal position of the film. Parameters for BTO, CFO, and MgO can be found in the literature⁴²⁻⁴⁴. In addition, out-of-plane magnetoelectrically induced electric polarization can be expressed as a function of the total stress of piezoelectric (σ^p) and magnetostrictive (σ^m) materials^{13, 20},

$$P_{ME} \propto (\sigma_{33}^p - \sigma_{33}^m) \quad (3)$$

$$\sigma_{33}^{p(m)} = 2E_{13}^{p(m)} \varepsilon_{11}^{p(m)} + E_{33}^{p(m)} \varepsilon_{33}^{p(m)} \quad (4)$$

where $\varepsilon^{p(m)}$ is the total strain including residual and magnetostrictive-induced strain. When the substrate clamping effect decreases, the magnetostrictive strain component $\varepsilon_{11}^p (= \varepsilon_{11}^m)$ increases as an exponential function. As a result, σ_{33}^p increases and this contributes largely to electric polarization P_{ME} , giving rise to the enhanced ME coupling. The enhancement of the interfacial strain transfer can be further corroborated by measuring the vibrational mode frequency change in the BTO layer. To this end, magnetic field dependent micro-Raman spectroscopy measurements were performed on the 3- μm structures and the as-deposited film (Fig. 3(f)). For the 3- μm structures, blue shift in BTO soft-mode $A_1(\text{TO})$ were observed while no noticeable shift was detected in the as-deposited film. When an external magnetic field is applied, CFO layer will shrink in the direction parallel to the magnetic field because of its

negative magnetostriction. Therefore, CFO gives in-plane compressive strain to the BTO layer, which increases the frequency of soft-mode $A_1(\text{TO})$ ^{45,46}.

Another interesting point from the analytical calculation above is the non-uniform stress distribution (inset of Fig. 3(e)) in the microstructures; stress from the substrate clamping becomes much smaller near the edge of the structure compared to the center³⁸. This indicates that ME induced interfacial strain would be uneven within the structure because of the non-uniform substrate clamping. In order to scrutinize the non-uniformity of ME coupling, we performed local PFM hysteresis loop measurements on 5×5 grids of 3 μm structure surface (Fig. 4). Although we couldn't quantitatively calculate the ME generated electric field nor ME coupling coefficients with our experimental setup, it is possible to study the degree of the ME coupling by comparing the deviation of coercive voltages. For a semi-quantitative comparison, we defined k_{ME} as following:

$$k_{ME} = \Delta \bar{E} \cdot \frac{A'_N A'_P}{A_N A_P} \quad (5)$$

where $\Delta \bar{E}$ is an average of the deviation of coercive fields, $A'_{N(P)}$ and $A_{N(P)}$ are piezoresponse amplitudes under negative (positive) bias with and without external magnetic fields, respectively. k_{ME} values from each grid are represented as a color map in Fig. 4(a) and representative PFM amplitude hysteresis are provided in Fig. 4(d) and 4(e). Larger k_{ME} values were obtained near the edge compared to the center of the structure, indicating the significant role of the reduced substrate clamping.

Interestingly, depending on the location, opposite ferroelectric imprints and internal electric field directions (positive and negative k_{ME} values) were observed. This could be explained with the strain gradients, i.e. flexoelectric effect, generated by external magnetic field. When mechanical strain is applied to the substrate (e.g. by bending of the film), strain gradient is introduced in the ferroelectric film, and a flexoelectric field ($E_f = f \nabla \varepsilon$) is developed, resulting in ferroelectric imprints⁴⁷⁻⁵⁰. In our thin film microstructures, when an in-plane magnetic field is applied, the CFO layer imposes magnetostrictively induced strain to the BTO layer. Because of the negative magnetostriction coefficient of the CFO, compressive strain is applied to the BTO layer along the direction parallel to the magnetic field (y-axis) while tensile strain is applied to the vertical direction (x-axis) due to the Poisson effect. The interfacial strain will be relaxed towards the surface, resulting in strain gradients and thus flexoelectric fields across the the film. Owing to the non-uniform substrate clamping in the microstructures, different magnitudes of magnetostrictive strain would be applied to the BTO layer depending on the location, leading to the strongest flexoelectric fields close to the edges.

For example, at position (i), the BTO layer experiences compressive strain and an upward inner electric field is generated (Fig. 4(c)); thus, a negative imprint is observed in PFM results (Fig. 4(d)). On the other hand, at position (ii), BTO experiences tensile strain and therefore shows a positive imprint (Fig. 4(e)). We also performed finite element method COMSOL simulation on a 3 μm structure (calculation details in Supplementary information). As shown in Fig. 4(b), positive out-of-plane flexoelectric field is developed along the direction parallel to the magnetic field (y-axis) while negative field is developed in vertical direction (x-axis), which coincides with the local PFM hysteresis measurements in Fig. 4(a).

In summary, we fabricated micro-scale 2-2 thin film structures and investigated the effect of the pattern size on ME coupling. We found that the degree of substrate clamping dropped when the size of the structure was reduced, which resulted in enhanced ME coupling in the thin film micro patterns. Interestingly, site-dependent ME coupling was observed within the microstructures due to the non-uniform substrate clamping across the film and the strain gradient developed in the BTO layer brought by the magnetostriction of CFO. Our findings are crucial to designing micro- and nanoscale devices based on 2-2 ME composite thin films. We believe that sophisticated control of the distribution of magnetoelectrically generated electric field could be possible by adjusting the pattern shape, external field direction and size.

FIG. 1. Fabrication process and ME coupling in ME microstructures. (a) Schematic illustration of the fabrication process of ME thin film microstructures. (b) Comparison of ME coupling in as-deposited ME thin film (left) and ME thin film microstructures (right). In ME thin film microstructures, site-dependent ME coupling was observed with maximized values near the edge of the structures.

FIG. 2. Structural analysis of BTO/CFO as-deposited thin film. (a) X-ray diffraction theta-2theta scan of as-deposited BTO/CFO film on MgO (001) substrate and (b) magnified scan near MgO (002) peak. Only {001} peaks have been observed, indicating that the films are epitaxial. (c) Cross-sectional HAADF-STEM image of the BTO/CFO bilayer thin film grown on MgO(001). (d) Corresponding in-plane strain map obtained by GPA. Near the interface, the BTO film is under tensile strain (red) while the strain is relaxed towards the film surface (green). The out-of-plane strain map and extracted line profiles are provided in Fig. S1. (e) Polarization map calculated from the displacement of the Ti cations from the center of their four Ba neighbors from the region indicated with a yellow rectangle in panel (d). (f-g) Profiles along the vertical direction of the average deviation of the Ti-cations along the x - (in-plane) and y - (out-of-plane) directions. Near the interface, the Ti-cations are displaced along the in-plane direction, while they are displaced along the out-of-plane direction near the surface. All scale bars denote 2nm.

FIG. 3. Enhanced magnetoelectric coupling from reduced substrate clamping effect. Amplitude and phase PFM loops measured from as-deposited film (a-b) and a $3\mu\text{m}$ structure (c-d) with and without 500Oe external magnetic field. For a $3\mu\text{m}$ structure, measurements were performed at the center of the structure. Clear coercive field shift is observed in a $3\mu\text{m}$ structure while no significant shift is observed in as-deposited film. (e) Degree of clamping as a function of lateral size of the island structure. Inset represents relative stress distribution within the film. (f) Magnetic field dependent Raman spectroscopy measured on as-deposited film and $3\mu\text{m}$ structures. Black, red, green, blue dots represent measurements of as-deposited, as-deposited (55mT), $3\mu\text{m}$ disk, and $3\mu\text{m}$ disk (55mT), respectively.

FIG. 4. Site-dependent magnetoelectrically generated flexoelectric field. (a) $3\mu\text{m}$ disk was divided into 5×5 grids and local PFM loops were measured. On each grid, PFM loops were measured 5 times and averaged. The scale bar indicates calculated k_{ME} values. (b) COMSOL

simulation of the distribution of flexoelectric field ($E_{f,3}$) when 500Oe magnetic field is applied along y-axis in $3\ \mu\text{m}$ disk BTO/CFO bilayer structure. (c) Schematic illustration of magnetoelectrically induced flexoelectric fields in BTO layer along different directions. (d-e) PFM amplitude hysteresis measured on grids of (d) minimum negative and (e) maximum positive k_{ME} values. PFM phases can be found in Fig. S5.

Supplementary Material

See supplementary material for further details.

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The data that support the findings of this study are available from the corresponding author upon reasonable request.

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