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Electrically controlled switching of the magnetization state in multiferroic BaTiO3\CoFe sub-micron structures

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Abstract:

The development of reliable and highly energy efficient multiferroic nanosystems, which can function at room temperature, is key for the design of ultra-low power magnetoelectric devices. Here we report the electrically controlled magnetic domain wall motion and magnetization switching in $BaTiO₃/CO₅₀Fe₅₀$ microstructures, at room temperature. The perfect one-to-one connection between the ferroelectric domain pattern of the $BaTiO₃$ crystal and the ferromagnetic state of the CoFe microstructures, which relies on a strain-induced magnetic anisotropy modification, is the cause of the observed magnetoelectric effect. As a result, the observed electrically driven magnetization switching is highly reliable, independently of the shape and size of the microstructures. This is a key factor that makes the studied multiferroic system very promising for the integration in real-world magnetoelectric devices.

New generation magnetic memory and logic devices are expected to merge high thermal stability and operational reliability with a low power consumption [1]. At the moment, magnetic memories are based on the manipulation of magnetization by electric currents [2], resulting in high energy consumption. Accordingly, a new paradigm based on the control of magnetization by an electric field is highly desirable [3,4]. Multiferroic systems [5,6], where both the ferromagnetic and the ferroelectric order parameters are present and coupled to each other, have been extensively investigated for this scope. In particular, hetero-structures consisting of a ferromagnetic thin film deposited on top of a BaTiO₃ ferroelectric/ferroelastic substrate have been reported to be a particularly interesting magnetoelectric system [7–15], where a strong magnetoelectric coupling at the interface between the two materials dominates the magnetic state in the ferromagnet. The very stable one-to-one correspondence between the ferroelectric (FE) and ferromagnetic (FM) domain patterns observed in some of those systems [7–9,16], joined with the naturally occurring micrometer-size of those domains, opens up to a fully deterministic control of magnetism by electric fields at the micro- and nano-scale. This approach is potentially much more reliable if compared with what obtained with other multiferroic heterostructures [17–20], where the strain-based magnetization manipulation can be strongly influenced by uncontrolled nonuniformities in the local strain at the microscale. However, even if already shown in extended films, the feasibility of such approach in micro- and nano-structures remains an open question. Indeed, some attempt to study the magnetoelectric behavior of BaTiO3\ferromagnet micro-devices has been carried on in the past, like in the work by Baldrati *et al.* [21]. However, the lateral size of those devices is in the order of 100 μ m, which is not small enough for studying the electrical control of magnetization at the micrometer and submicrometer scale, where ferromagnetic single domain structures start to emerge.

Here, we report the electric field-controlled magnetic domain wall (DW) motion and magnetization switching in $Co₅₀Fe₅₀$ micro-\sub-microstructures on top of a BaTiO₃ single crystal substrate. By X-ray microscopy we image both the FE state at the top surface of the $BaTiO₃$ substrate and the FM state of the magnetic structures patterned on top of the single crystal. In addition, the electrically induced magneto-elastic anisotropy is characterized by magneto-optic Kerr effect magnetometry. The results here reported are of great interest for the development of energy efficient magnetoelectric devices.

The studied multiferroic system consists of 20 nm-thick $Co₅₀Fe₅₀$ microstructures on top of a 500 µm-thick ferroelectric BaTiO₃ (BTO) single crystal (*SurfaceNet GmbH*; 48432 Rheine, Germany) as in Fig. 1a and 1b. The ferroelectric substrate is in its tetragonal phase, with the polar axis lying into the plane of the crystal and initially in a ferroelectric $a_1 - a_2$ multidomain state [7,22] (stripe-like domains in Fig. 1b). 90°-type FE domain walls (DWs) separate the a_1 from the *a2* domains (Fig. 1c). The BTO crystal has a continuous *Au* bottom-electrode and a *Pt* top-electrode surrounding the patterned magnetic structures. The $Co₅₀Fe₅₀$ (CoFe) structures array includes squares and disks, with lateral sizes of 5 μ m, 2 μ m, 1 μ m and 500 nm. The magnetic structures patterning involves electron-beam lithography, magnetron-sputtering deposition, and lift-off techniques. Finally, a 1.5 nm-thick Pt layer is deposited over the top surface of the sample. The FE state of the BTO and the FM state of the CoFe structures are imaged respectively by exploiting the X-ray Linear Dichroism (XLD) at the Ti L_3 -edge [23] and the X-ray Magnetic Circular Dichroism (XMCD) at the Co L_3 -edge [24], with a Photoemission Electron Microscope (PEEM).

FIG. 1. (a) Sample schematic. (b) Optical microscopy image of the magnetic microstructures. Surrounding the microstructures is the *Pt* top electrode. (c) Aggregate of XLD-PEEM images showing the initial FE state of the sample in the ROI (FE polarization state described in the inset). (d) Aggregate of XMCD-PEEM images showing the initial FM state. Dashed lines indicate the position of the two FE DWs. The low inset describes the found magnetic anisotropy after deposition. The grey scale wheel describes the magnetic contrast. The blue arrow indicates the initializing magnetic field, $\mu_0H_{in} = 250$ mT.

The initial FE state at the top surface of the BTO crystal in the region of interest (ROI) is shown in Fig. 1c. Three different domains are visible, with the central domain (dark grey) having a different polarization state than the others (light grey). Initially, an external magnetic field (μ_0H_{in})

 $= 250$ mT) is applied along the 45 $^{\circ}$ direction and then removed in order to initialize the magnetic structures. The resulting FM state of the CoFe structures is reported in Fig. 1d. Most of the magnets are found to be magnetized along the positive x-direction (white contrast). However, the magnetic structures lying on top of the central FE domain are found to be in a different state. Indeed, as recognizable in the magnetic structures located on top of the FE DWs (dashed orange lines in Fig. 1d), the parts of them lying on the central domain are mostly grey (magnetization // y-axis), while the parts lying in the lateral domains are white (magnetization // x-axis). This is in agreement with what reported in literature [7,16], where a similar one-to-one FE-FM connection was observed for thin films. This is a magneto-elastic effect, where the magnetization inside the FM thin film aligns with the polar axis of the FE domains. Accordingly, we can conclude that the FE state in Fig. 1c is due to the presence of an $a_1-a_2-a_1$ domain pattern (inset in Fig. 1c), which generates a $K_X-K_Y-K_X$ magneto-elastic anisotropy pattern for the CoFe magnets (inset in Fig. 1d).

After characterizing the electrically virgin sample, an electric field is applied *in-situ* between the top and the bottom electrode. When a DC electric field $E = 0.6$ MV/m is applied, a lateral displacement of the central a_2 FE domain is observed. The new state of the crystal is shown in Fig. 2a, where the red arrows indicate the electrically driven displacement of the DWs. As described in Fig. 2a: on the left, the polarization in the BTO crystal rotates from a_2 to a_1 ; while on the right, the rotation is from a_1 to a_2 . A possible way to interpret the observed FE DWs motion is based on the interaction between a new electrically nucleated out-of-plane domain (*c* domain) and the initial in-plane domains (*a* domains). Indeed, it has been reported that during the poling process of a tetragonal ferroelectric, new FE domains are nucleated and the propagation of those new ferroelectric domains can be stopped by ferroelastic domains initially present in the

crystal, with their polarization being normal to the applied electric field [25,26]. Accordingly, the observed displacement of the central domain can be interpreted as the repulsion between such domain and a newly nucleated ferroelectric domain underneath. However, due to the surface sensitive nature of PEEM, we are unable to verify the actual presence of new FE *c* domains nucleated in the bulk of the BTO crystal. Finally, as visible in the magnetic image in Fig. 2b, the 90° rotation of the electrical polarization in the BTO corresponds to a 90° rotation of the magnetization in the CoFe structures affected by the DWs motion, testifying for a very effective electrical modification of the magnetic anisotropy. On the left (right), the magnetic easy-axis is rotates from *y* to *x* (*x* to *y*). The magnetization rotation is in general non-uniform, resulting in the generation of a reoriented magnetic multi-domain state (black-white contrast in Fig. 2b) in most of the affected magnetic structures.

FIG. 2. (a) FE state at the bottom-left corner of the ROI with $E = 0.6$ MV/m. The red arrows indicate the electrically induced FE DWs displacement. Insets describe the reorientation of the polar axis. (b) Corresponding FM state. The dashed orange and solid green lines delimit the areas where a magnetization is rotated. Insets describe the magnetic easy-axis rotation.

From a technological point of view, it is very interesting to study the influence of the magnets' shape and size on the electrically controlled magnetic reorientation. Based on what shown in Fig. 2b, the size of the magnetic squares does not affect the ability to switch their magnetization. Indeed, every magnet located on top of the displaced DWs is reoriented, independently of its lateral size $(5 \mu m - 500 \text{ nm})$. Furthermore, the left column in Fig. 3 shows the electrically induced magnetic state of three different areas of the ROI. The magnetization reorientation happens in the squares as well as in the disks, independently of the shape. The right column in Fig. 3 shows more in detail the reorientation process in some of the small magnets, with a lateral size of 1 µm and 500 nm. At these lateral sizes some of the magnets are observed to be in an initial single domain state (white domain, dashed red frames). When the electric field is applied, the magnetization in those same magnets is switched to the y-axis (grey domain, solid red frames). This demonstrates an electrically induced single spin-like 90° magnetization switching, which is a key result in the applicability of such material system in a hypothetical magnetoelectric memory device, where *white* (or *black*) and *grey* would represent the digital *1* and *0* states of a memory bit.

 Based on the magnetization switching effect described above, we can calculate a converse magnetoelectric coupling coefficient (CMC) $\alpha_{\text{CME}} = \mu_0 \Delta M / \Delta E = 2.7 \times 10^{-6}$ s/m; where ΔM is the induced change in magnetization (M_S = 1.3x10⁶ A/m, measured by vibrating sample magnetometry on a reference film on Si) and ΔE is the required electric field to induce such magnetization change. This value is in line with the highest CMCs reported so far [7,27], highlighting the outstanding magnetoelectric properties of the studied multiferroic microstructures.

FIG. 3. The left column shows the magnetic state in three areas of the ROI, with $E = 0.6$ MV/m. The black, yellow and blue solid frames indicate the regions enlarged in the right column, which report the magnetic state of 1 μ m and 500 nm magnets at $E = 0$ MV/m (left) and 0.6 MV/m (right). The dashed and the solid red frames indicate the initial and the final state of the highlighted nanomagnets, respectively.

Finally, we study the magnetic state of the CoFe structures when further electrically poling the BTO crystal along the out-of-plane direction, so to directly quantify the electrically induced magnetic anisotropy in the CoFe structures. A single FE domain is observed for $E \ge 0.64$ MV/m (polarization pointing *up*, see sample schematic in Fig. 1a) everywhere in the ROI, which is retained after removing the electric field. The corresponding magnetization state of the CoFe structures is shown in Fig. 4a. All the magnetic parts lying on the original position of the central *a2* domain are now magnetized along *x* (light grey), while the rest of the structures are magnetized along *y* (dark grey). This is in agreement with what reported in literature [7], where the compressive strain generated along *x* (*y*) during the $a_1 > c$ ($a_2 > c$) transition in the FE state

induces a magneto-elastic anisotropy pattern inside the ferromagnetic layer that is one-to-one connected to the initial $a_1 - a_2$ multidomain state.

Magneto-optic Kerr effect (MOKE) magnetometry (using a Kerr microscope) is employed in order to quantify the final magnetic anisotropy inside the CoFe structures. Longitudinal-MOKE hysteresis loops are acquired, with a magnetic field applied along the *x* and *y*, for the two 5 µm magnetic disks at the top-right corner of Fig. 4a. As shown in Fig. 4b, the top-right dot is characterized by a magnetic easy-axis along x (M_x v_s H_x , solid line), with a coercive field $\mu_0H_c = 66$ mT; while the magnetic hard-axis is along *y* (M_y *vs* H_y, dashed line). The opposite situation is found for the top-center disk (see Fig. 4c): magnetic easy-axis along *y* (M_v *vs* H_v, dashed line), with a coercive field μ_0 H_c = 62 mT; magnetic hard-axis along *x* (M_x *vs* H_x , solid line). Based on this, we can assert that to a single *c* domain in the BTO crystal correspond three different regions in the FM array, with induced magnetic uniaxial anisotropy along y (K₁) and x (K₂) (see Fig. 4d). The hard-axis anisotropy field, H_K , is extracted by fitting the plots in Fig. 4b and 4c with the following function: $sin(atan(H/H_K))$; where *H* is the applied magnetic field along the hard-axis. An anisotropy field $\mu_0 H_K = 72 \pm 2$ mT (74 \pm 4 mT) is extracted for the top-right (top-center) magnetic disk. We can calculate the induced magneto-elastic anisotropy as $K = \mu_0 M_S H_K/2$, so that the values $K_1 = 47 \pm 1$ kJ/m³ and $K_2 = 48 \pm 3$ kJ/m³ are extracted.

The theoretically expected maximum magneto-elastic anisotropy due to the $a \rightarrow c$ ferroelectric transformation is given by $K_{me} = -3\lambda_s \epsilon Y/2$; λ_s and Y are respectively the saturation magnetostriction coefficient and the Young's modulus of polycrystalline thin film $Co₅₀Fe₅₀$, while $\varepsilon = -1.1\%$ is the uniaxial compressive strain induced by the full $a \rightarrow c$ transformation in the BTO crystal ($a = 0.3992$ nm, $c = 0.4036$ nm) [7]. Based on the values available in literature [28– 31], we obtain $K_{m.e.} = 218 \text{ kJ/m}^3$ ($\lambda_s = 60 \text{ p.p.m.}$ and $Y = 220 \text{ GPa}$), which is about 4 times bigger than the experimentally extracted values above. This seems to be in agreement with what observed for similar CoFe thin film, where experimental values smaller than 50% of the theoretical maximum value where reported [7]. In part this can be attributed to an inefficient transfer of the strain from substrate to magnetic layer [32]. In addition, the specific crystallographic configuration of the magnetic thin film in our microstructures can also play a role [31,33]. However, an in-depth understanding of this discrepancy is not the scope of this work and we leave this open question to a future study. Nevertheless, we are able to observe a full electrical switching of the magnetization in the magnetic microstructures, which is the main finding and a key result from an application point of view.

Finally, it is worth noting that in the last years the growth and electrical manipulation of $BaTiO₃$ thin films has been extensively studied [21,34–37], making this ferroelectric material compatible for heterogeneous integration on silicon electronics. On the other hand, the electrical control of the ferroelectric/ferromagnetic domain patter at the micron-scale in thin film BaTiO₃\FM systems is yet to be proven, calling for further developments. Accordingly, once the FE DW-driven magnetization switching here reported will be demonstrated in systems with thin film BTO as well, BTO\CoFe multiferroic nanostructures are expected to become a concrete option for the design of real-world magnetoelectric memory devices.

FIG. 4. (a) Differential wide field Kerr microscopy image of the sample with electrically poled substrate. The image is taken at zero magnetic field, after the sample is saturated by a field $\mu_0H_v = 150$ mT along the *y*-axis. (b) Longitudinal-MOKE hysteresis loops for the top-right disk with a magnetic field swept along *x* (solid line) and *y* (dashed line). (c) Same as in (b) for the topcenter disk. (d) Schematic of the final FE state in the ROI and the induced magnetic anisotropy pattern.

In summary, an electric field-controlled magnetic DW motion and magnetization switching is achieved in $Co₅₀Fe₅₀$ micro- and sub-microstructures on top of a BaTiO₃ single crystal. The initial ferromagnetic state of the CoFe structures is defined by the ferroelectric domain patter present in the BTO crystal. The subsequent electrical poling of the substrate induces a motion of the ferroelectric DWs, which drives magnetic DWs to move generating a 90° rotation of the magnetization in the structures. An electrically induced single spin-like magnetization switching is observed in the 1 µm and 500 nm structures which were initially found in a single domain state. Finally, a giant converse magnetoelectric coefficient, $\alpha_{\text{CME}} = 2.7 \times 10^{-6}$ s/m, is extracted, which is among the highest reported so far. All this demonstrates the high reliability and efficiency of the studied BTO\CoFe microstructures, which result to be a promising candidate for the development of new, low power magnetoelectric memory devices. This work is expected to

motivate more experimental and theoretical studied focused on magnetic nanostructures on top of BTO single crystals and epitaxial thin films.

Note added. Concurrently to the original submission of this work, a relevant study by M. Ghidini and colleagues [38] appeared, which focuses on the investigation of the electrical control of magnetism in Ni microdots on a BaTi O_3 single crystal.

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