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Electric modulation of magnetization at the BaTiO₃/La_{0.67}Sr_{0.33}MnO₃ interfaces

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We report large (>10%) magnetization modulation by ferroelectric polarization reversal in the ferroelectric-ferromagnetic BaTiO₃/La_{0.67}Sr_{0.33}MnO₃ (BTO/LSMO) heterostructures. We find that the electrically induced change in magnetization is limited to the BTO/LSMO interface but extends about 3 nm deep into the LSMO layer—far beyond the expected screening length of metallic LSMO. It is suggested that this effect is due to a metal-insulator transition occurring at the BTO/LSMO interface as a result of electrostatic doping. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4726427>]

While the possibility of a magnetoelectric effect was first mentioned by Pierre Curie in 1894 (Ref. 1) and predicted to exist in Cr₂O₃ in 1960,² until recently it was assumed that the external electric fields cannot significantly alter the properties of magnetic materials. The effect was considered simply too small for any practical applications. The experimental observation of electrical modulation of magnetization in III-V magnetic semiconductors³ sparked a surge of activity in search of other materials exhibiting magnetoelectric coupling. Breakthrough possibilities have been offered with the recent advances in fundamental understanding and experimental studies of coexistence of ferroelectric and ferromagnetic ordering in multiferroic materials.^{4,5} As single-phase room-temperature multiferroics are rare in nature (with Cr₂O₃ as a noticeable exception⁶), artificial multiferroic heterostructures made of ferroelectric and ferromagnetic phases have been extensively engineered and studied in search for large magnetoelectric coupling.^{7–10} Among the most attractive materials for practical implementation of multiferroic heterostructures and fundamental studies of magnetoelectric coupling are the doped lanthanum manganites La_{1-x}A_xMnO₃, (A = Ca, Sr, or Ba).¹¹ These materials are characterized by a strong interplay between electron transport, magnetism, and crystal lattice distortions and a rich carrier-density-temperature phase diagram. A possibility of effective electrical modulation of carrier concentration and magnetic properties makes these materials extremely attractive for numerous applications in field-effect devices.¹² Modulation of carrier density in metallic manganite films can be realized by employing polarization ferroelectric materials as the gate electrodes.¹³ The sensitivity of the magnetic state of manganites to electrical charge was predicted for BaTiO₃/La_{1-x}Sr_xMnO₃ (BTO/LSMO) interfaces¹⁴ and experimentally explored using spin-dependent tunneling.¹⁵ A number of theoretical and experimental studies^{16–21} attributed magnetoelectric coupling to the interplay between changes in magnetization and accumulation/depletion states at the ferroelectric-ferromagnetic interfaces, which is fundamen-

tally different from the coupling mechanism in single phase materials.²

In this paper, we investigate modulation of magnetization by ferroelectric polarization reversal in ferroelectric-ferromagnetic BaTiO₃/La_{0.67}Sr_{0.33}MnO₃/SrTiO₃ (BTO/LSMO/STO) heterostructures as a function the ferromagnetic layer thickness. We find that the effect is limited to the BTO/LSMO interface but extends up to about 3 nm into the LSMO layer beyond the expected screening length of metallic LSMO. It is suggested that the latter effect is due to a metal-insulator transition occurring at the BTO/LSMO interface as a result of electrostatic doping.

Pulsed laser deposition was used to grow atomically flat single-crystalline epitaxial BaTiO₃ films on TiO₂-terminated SrTiO₃ substrates with the La_{0.67}Sr_{0.33}MnO₃ films of variable thickness as bottom electrodes. It has been shown previously²² that growing compressively strained BaTiO₃ films on SrTiO₃ substrates enhances the resulting polarization and aligns it normal to the surface. In our studies, thickness of BaTiO₃ was chosen to be 48 unit cells (~19 nm) to ensure stable and switchable polarization, and LSMO thickness varied in the range from 10 to 50 nm. Structural quality and ferroelectric properties of the grown heterostructures are very similar to those reported in our earlier studies.²³ The atomic force microscopy (AFM) topographic image of the surface morphology in Fig. 1(a) shows atomically flat terraces with one unit-cell high (~4 Å) steps indicating epitaxial structure of the BaTiO₃ films. Details on sample preparation can be found elsewhere.²²

A commercial AFM system (MFP-3D, Asylum Research) was used for piezoresponse force microscopy (PFM) testing of polarization state in the BaTiO₃ layers. Pt-coated conductive cantilevers (DPER18/Pt, Mikromasch) were employed for polarization imaging at a frequency of about 300 kHz in the resonance-enhanced PFM mode. Superconductivity quantum interference device (SQUID) magnetometer (MPMS, Quantum Design) and Kerr microscope based on longitudinal magnetooptical Kerr effect (MOKE) have been used to measure magnetization of LSMO films.

FIG. 1. (a) AFM image of BaTiO₃ surface topography showing terraces and periodic steps with a unit-cell height. (b) Schematic illustration of non-contact macroscopic poling method: a large needle held at a dc bias of ± 800 V and at ~ 100 μm above sample surface scans the sample surface ($\sim 5 \times 5 \text{ mm}^2$) in high vacuum conditions. The step increment during scanning was 50 μm . (c) PFM phase image of the polydomain structure of the as-grown BTO film. (d) PFM phase image of the uniformly polarized BTO film after noncontact poling. (e), (f) PFM amplitude (e) and phase (f) images showing comparison between the macroscopic non-contact poling and poling by PFM. Central $2 \times 2 \mu\text{m}^2$ and $0.5 \times 0.5 \mu\text{m}^2$ squares have been produced by scanning with an AFM tip under $+4 \text{ V}/-4 \text{ V}$ dc bias in contact mode, while an outer region shows the result of macroscopic non-contact poling. Saturated polarization has been obtained for both poling methods. Scan size is the same for all AFM/PFM images.

MOKE measurements have been performed by focusing the green ($\lambda = 532 \text{ nm}$) laser beam to the spot of less than 100 μm indiameter on the BaTiO₃ surface.

Investigation of magnetic response by means of SQUID requires large-scale sample area poling (about several mm^2), which is difficult to accomplish if the ferroelectric film does not have a top electrode (for comparison, an AFM with a conductive probe can only pole an area of up to $150 \times 150 \mu\text{m}^2$ which might also take up to an hour of scanning). To overcome this problem, we have employed macroscopic non-contact probe poling technique. A Pt needle with the 20- μm -apex-curvature-radius held at a distance of about 100 μm above the BTO film surface and electrically biased at ± 800 V scanned the whole sample area in high vacuum conditions (4×10^{-9} Torr) as is schematically shown in Fig. 1(b). PFM analysis of several regions over the sample surface has been used to verify the result of non-contact poling.

Figures 1(c) and 1(d) show $3 \times 3 \mu\text{m}^2$ PFM phase images of the BTO film before and after noncontact poling demonstrating that the polarization had been effectively switched from as-grown polydomain state to a single domain state. Figures 1(e) and 1(f) show a comparison of the poling results by the macroscopic non-contact method and by PFM (in PFM, the BTO film has been poled by scanning the surface in contact with the PFM probe under a $\pm 4 \text{ V}$ dc bias). It can be seen that both poling methods result in fully switched single-domain polarization states.

SQUID measurements of the BTO/LSMO/STO heterostructures have been performed for both BTO polarization directions after verification of the results of large-scale non-contact poling by PFM. Cycles of the BTO films poling followed by the SQUID measurement have been carried out several times to ensure that magnetic response of the LSMO layer is reproducible and not affected by possible artifacts,

such as sample misalignment. Typical in-plane magnetization curves of LSMO acquired at room temperature for two antiparallel polarization states of the BTO film are shown in Fig. 2(a). It is seen that polarization reversal in BTO leads to a sizable change in the magnetic moment of the adjacent LSMO layer. For all studied samples, the magnetic moment of LSMO decreases as the BTO film is electrically switched from the upward (away from the substrate) to downward (toward the substrate) polarization state.

To substantiate the results obtained by SQUID, additional measurements by local MOKE measurements have been performed using the same samples. Ferroelectric poling of two regions of approximately $100 \times 100 \mu\text{m}^2$ of the BTO film have been carried out using a conductive PFM probe in contact with the surface by applying a dc bias of $\pm 5 \text{ V}$, followed by MOKE measurements of in-plane magnetization (using surface topographic markers the laser spot was focused on exactly the same regions where polarization reversal had been performed). The results in Fig. 2(b) indicate the same trend as those in Fig. 2(a): LSMO magnetization decreases when the polarization in the BTO layer is switched from the upward to downward direction.

Further, we studied the relative change in saturated magnetization, $\Delta M = (M(P_{\text{up}}) - M(P_{\text{down}}))/M(P_{\text{down}})$, as a function of LSMO thickness (Fig. 2(c)). It should be mentioned that both upward and downward polarization states in the BTO film were found to be stable for the 25- and 50-nm-thick LSMO layers, while for the 10-nm-thick LSMO layer, only upward polarization was found to be stable. Attempts to switch the BTO film downward in this case resulted in formation of the polydomain structure with the $P_{\text{down}}/P_{\text{up}}$ ratio of approximately 1/2 (most likely due to spontaneous back-switching). This effectively means that only 1/3 of the total downward polarization state had contributed to the

FIG. 2. (a) In-plane M-H curves of BTO/LSMO(25 nm) heterostructure measured by SQUID at room temperature for upward and downward polarization directions in BTO. (b) In-plane M-H curve of BTO/LSMO (10 nm) heterostructure measured by MOKE at room temperature for upward and downward polarization directions in BTO showing the same trend as in (a). (c) Dependence of the relative change of magnetization ΔM on the LSMO layer thickness t . The open circle indicates a normalized data point. The solid curve is a fit to the $1/t$ function. (d) Temperature dependence of saturation magnetization of BTO/LSMO (25 nm) heterostructure for upward and downward BTO polarizations measured at 2500 Oe. The inset shows temperature dependence of the relative magnetization change ΔM .

magnetization change. By taking into account this correction, which reflects non-complete downward poling, the resulting relative magnetization change ΔM was estimated to be about 27% (without this correction the measured ΔM value was $\sim 9\%$).

Figure 2(d) shows a temperature dependence of saturated magnetization in the BTO/LSMO(25 nm)/STO heterostructure measured at a constant magnetic field 2500 Oe. No noticeable change in the Curie temperature with polarization reversal has been observed. The temperature dependence of ΔM (see the inset in Fig. 2(d)) shows that the polarization-induced change in magnetization peaks in the vicinity of the Curie temperature and decreases as temperature decreases.

Data in Fig. 2(c) show that the relative change in magnetization ΔM becomes more pronounced as the LSMO thickness decreases. The thickness dependence of ΔM can be fitted well to the t_m/t function, where t is the LSMO layer thickness and t_m is the fitting parameter. This suggests an interface mechanism behind the observed magnetization modulation by polarization reversal, i.e., that the magnetization change in LSMO is limited to a thin layer at the BTO/LSMO interface. The fit allows us to estimate the effective layer thickness at the BTO/LSMO interface where the magnetization change occurs. Assuming that within this layer the reversal of ferroelectric polarization completely destroys magnetism, we can interpret t_m as the thickness of this layer. From the fit we find that t_m is approximately 2.9 nm.

The observed effect is in qualitative agreement with our first-principles calculations performed using the plane-wave pseudopotential method.²⁴ Reversal of the ferroelectric polarization leads to a change in the accumulation/depletion charge built up on the LSMO side of the interface. We find

that all of the electronic screening is limited to an accumulation/depletion of majority spin electrons, which leads to a significant change in magnetic moment near the interface. In bulk, $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ has a magnetization of $3.67 \mu_B/\text{Mn}$, whereas the average Mn moment varies significantly in the first 2–3 unit-cells near the interface with BTO, depending on the polarization orientation. For the BTO polarization pointing towards the BTO/LSMO the interface, for example, the magnetic moments on the Mn sites are essentially the same as they are in the bulk,¹⁴ leading to a near complete absence of electronic screening. On the other hand, for the BTO polarization pointing away from this interface, the negative surface polarization charge from the BTO leads to a significant depletion of majority spin electrons in the first 2–3 unit-cells of LSMO: the first Mn layer at the interface is reduced by $\sim 0.45 \mu_B$, the second is reduced by $\sim 0.2 \mu_B$, and the third is reduced by $\sim 0.05 \mu_B$. This corresponds to an effective penetration depth of the magnetoelectric response on the order of 0.8–1.2 nm in our zero temperature calculations, which is in qualitative agreement with our experimental results.

Quantitatively, however, the experimentally observed magnetoelectric effect is significantly larger in magnitude than that predicted theoretically. Also, the thickness of the ferroelectrically modulated magnetic layer estimated from the experimental data is much larger than theoretically predicted (~ 3 nm versus ~ 1 nm, respectively), which cannot be explained solely within the model assuming uniform and constant metallicity of LSMO at the BTO/LSMO interface. Below, we propose a qualitative model, which explains our experimental data based on the metal-insulator transition at the LSMO interface induced by the ferroelectric polarization.

FIG. 3. (a) Temperature-composition phase diagram of LSMO showing ferromagnetic metal (FM), paramagnetic insulator (PI), ferromagnetic insulator (FI), and paramagnetic metal (PM) phases. (Reprinted with permission from E. Dagotto, T. Hotta, and A. Moreo, Phys. Rep. **344**, 1 (2001). Copyright 2001, American Institute of Physics.) Also shown are calculations of the shift of the phase boundary between insulating and metallic states for two possible BaTiO₃ polarization values. Polarization screening shifts the effective charge concentration x at the interface: the leftward shift leads to metal-insulator transition while the rightward shift keeps the system in the metallic state. (b) Schematic illustration of the metal-insulator phase transition in LSMO due to accumulation of screening charges at the BTO/LSMO interface. (c) Temperature dependence of the thickness of the magnetically modulated layer in LSMO.

The screening charge at the ferroelectric-manganite interface produces carrier depletion or accumulation in LSMO near the interface, analogous with chemical doping. Since x in the $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ chemical formula reflects the substitution of trivalent La with divalent Sr and is an effective measure of holes concentration, we can consider the effect of electric field as a corresponding shift in the LSMO phase diagram¹¹ (Fig. 3(a)) to the right, if a negative polarization charge is to be screened, or to the left, if positive polarization charge is to be screened, relative to the starting point of $x=0.33$. It can be seen from the diagram that while the rightward shift keeps the LSMO layer in the metallic state, the leftward shift can lead to a metal-insulator transition. The presence of the metal-insulator transition occurring locally, within the screening length at the LSMO interface, would inevitably move the screening region deeper into the bulk LSMO (Fig. 3(b)). For example, assuming that the strained BTO has the polarization of $48 \mu\text{C}/\text{cm}^2$, and all the screening charge is accumulated within 2.5 unit cells of LSMO (as in our theoretical calculations), the total doping (both chemical and electrostatic) within the screening region becomes $x \approx 0.13$, which lies deep in the paramagnetic insulator region at room temperature in the phase diagram of LSMO (Fig. 3(a)). This suggests that the screening region expands beyond that found for the metallic phase of LSMO (Fig. 3(b)). This effect is expected to be more pronounced in the vicinity of T_C , where the difference in free energy between the metal and insulating states for the LSMO film is minimal.

In order to correlate our experimental results with the phase diagram of LSMO, we have calculated the occurrence of the metal-insulator phase transition based on our experimental data for the polarization-induced change in magnetization shown in the inset of Fig. 2(d). Assuming as before that within the layer of thickness t_m ferroelectric polarization reversal completely destroys magnetism, we can calculate t_m as a function of temperature (Fig. 3(c)). Using this temperature dependence and known values of the polarization charge for BTO we can find the phase boundary between the metal

and insulator regions in the phase diagram simply by estimating the amount of doping level in the layer of thickness t_m as a function of temperature. The estimated equivalent changes in the holes concentrations (change in x) that are required to accommodate all the polarization charge are shown in Fig. 3(a) for two BaTiO₃ polarization values: experimentally measured²⁵ value of $35 \mu\text{C}/\text{cm}^2$ and predicted in our first-principles calculations value of $48 \mu\text{C}/\text{cm}^2$. There is a clear correlation between the bulk phase diagram of LSMO and the predicted metal-insulator phase boundaries.

In summary, we have observed large (>10%) room-temperature electrical modulation of magnetization in ferroelectric/ferromagnetic BaTiO₃/La_{0.67}Sr_{0.33}MnO₃/SrTiO₃ heterostructures induced by polarization reversal in BaTiO₃. The relative magnetization change is inversely proportional to the LSMO layer thickness, indicating that the effect is limited to a thin (~ 3 nm) LSMO layer at the BTO/LSMO interface. The thickness of this layer and the magnitude of magnetization change are significantly larger than those predicted by first-principle calculations at zero temperature. The experimental data can be qualitatively explained using a model based on a polarization-induced metal-insulator transition in the LSMO layer near the interface.

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