Direct reversible magnetoelectric coupling in ferroelectric/ferromagnetic structure controlled by series resistance engineering

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ABSTRACT GRAPHIC

ABSTRACT Achieving large magnetoelectric coupling is of interest for memory and communication applications. In multiferroic hybrid structures (combining ferroelectric and magnetic materials) in presence of a magnetoelectric coupling, the ferroelectric properties can be modulated by a magnetic field. This is called direct magnetoelectric effect. Measuring the ferroelectric properties in multiferroic materials most commonly requires using metallic electrodes that sandwich the ferroelectric material. In the present work, we use the series resistance introduced by the metallic electrode ($\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$) to manipulate one relevant ferroelectric parameter, i.e. the coercive voltage, of an adjacent ferroelectric layer (BaTiO_3) by a magnetic field. We will show that the variations are fully reversible and more apparent at high frequencies. Being the high frequency regime the frequency range where most of the envisaged multiferroic devices would work, our results are particularly interesting for applications, in addition of being of relevance from the methodological point of view.

1. Introduction

The quest of magnetoelectric coupling at room temperature has motivated a huge amount of research in multiferroic systems.¹⁻⁴ Single-phase multiferroic materials⁵⁻⁶ or hybrid (combination of ferroelectric and ferromagnetic materials) multiferroic structures have been investigated.⁷⁻¹¹ Recent reports have demonstrated that magnetoelectric coupling enables a large increase in energy efficiency of memory devices.¹² Control of magnetic properties by electric field (named converse magnetoelectric effect) has been reported in several hybrid systems at, or near room temperature.¹³⁻¹⁸ For applications, the direct magnetoelectric effect, that is the control of ferroelectric properties [dielectric permittivity, remanent polarization (P_r) and coercive voltage (V_c)] by magnetic field is also of interest. For instance, similarly to the well-known operation principle of heat-assisted magnetic memories where the coercive magnetic field is temporally reduced by a local temperature increase, one can envisage a ferroelectric memory where the coercive electric field is reduced (or increased) by magnetic field (as sketched in Figure 1). Thus, application of external magnetic field can be used to enable (or disable) ferroelectric switching, i.e. data writing, which can be used in memory devices with improved security, where data writing is only allowed by appropriate application of external magnetic field.

In the direct magnetoelectric approaches described above, research has been focused on the modulation of the intrinsic properties of the ferroelectric element, mainly P_r , by an external parameter (the magnetic field). This approach requires a strong intrinsic magnetoelectric coupling between atomic spins and polarization, that so far has been limited to few oxides and at low temperature.⁵⁻⁶ Extrinsic magnetoelectric response can also emerge due to magnetoresistance of the material³³ or induced by other effect associated with the measuring set-up.³⁴ However, the magnetoelectric response of a device under test is not exclusively dictated by the magnetoelectric

properties of the material, but also by the physical surrounding and electric connects, as described in the present work. Besides, different contributions can also modify the measured ferroelectric properties in an extrinsic manner.¹⁹⁻²⁰ Probably, the most well-known extrinsic contributions that can modify the measured ferroelectric properties are the leakage currents,^{19, 21-23} the interfaces between the metal and the ferroelectric,²⁴⁻²⁸ and parasitic capacitance of the measuring circuit (of relevance when characterizing nanosized capacitors by using proximity probes).²⁹⁻³²

Figure 1. Sketch of the P - V loop shape modification in a magnetoelectric system without and with the application of an external magnetic field. For convenience, the case in which magnetic field results in a decrease of the P_r and V_c is shown.

When measuring current-voltage (I - V) curves in ferroelectric capacitors, the series resistance (R_s) associated to electrodes, is typically neglected because most commonly it has a minute contribution to the measured current. However, when measuring ferroelectric thin films grown on ultrathin metallic oxide bottom electrodes, the resistance of the metal layer can be substantial and may impact the measured I - V with consequences on the ferroelectric parameters extracted.

For instance, the extracted coercive voltages V_c^* may differ from the intrinsic one (V_c) in a non-trivial manner. As V_c^* is the voltage where polarization reverses in a practical structure, understanding and controlling its value is of major importance for device engineering.

Here, we report on systematic measurements of an archetypical multiferroic hybrid structure,^{7, 35} consisting on a ferroelectric layer [BaTiO₃ (BTO)] grown on metallic ferromagnetic layer [La_{2/3}Sr_{1/3}MnO₃ (LSMO)]. First, we demonstrate the impact of modifying the series resistance on the I - V measurement and on the corresponding P - V loops. Second, we demonstrate that the R_s contribution to P - V increases with the frequency of the applied voltage V - t pulses. It will be argued that this sensitivity to frequency could be of interest for application, where commutation rates are expected to be high. Finally, we will make use of the R_s contribution to P - V to demonstrate a significant control the effective coercive field V_c^* of the ferroelectric capacitor by using magnetic fields, as sketched in Figure 1. We exploit this response to produce a reversible direct magnetoelectric effect, consisting on the 1 % modulation of V_c^* by applying a modest external magnetic field. In contrast to earlier approaches, the magnetoelectric response we observe is dictated by the electrodes magnetoresistance rather than by the dielectric magnetoresistance itself, which is what rules the well-known extrinsic Maxwell-Wagner magnetoelectric response.³³ We end by illustrating the contribution of R_s in the characterization of non-ferroelectric capacitors and we show that, in some cases, P - V loops get bananas due to R_s -contribution.

2. Results and discussion

5 mm x 5 mm BTO(100 nm)/LSMO(5 nm) and BTO(100 nm)/LSMO(20 nm) bilayers were grown in-situ by pulsed laser deposition (PLD) on (001)(La, Sr)(Al, Ta)O₃ (LSAT) and

(001)SrTiO₃ (STO) substrates, using condition described elsewhere.³⁶ Structural characterization indicated epitaxial growth of both layers, being LSMO fully strained on both substrates but BTO on LSAT more relaxed than on STO.³⁷ Pt electrodes (square-shaped and 50 μm lateral size) were deposited on top of BTO through a shadow mask by DC Sputtering. For electrical testing, two top Pt electrodes are connected by using tungsten needles fixed to a micro-positioning arms. One of the needles is grounded and the driving voltage V - t is applied to the other one, as shown in Figure 2(a). Therefore, the measured circuit consist into two BTO capacitors connected in series via the metallic LSMO bottom electrode, which short-circuits them. The measured structure is symmetric: Pt/BTO/LSMO/BTO/Pt. The electric field (current) paths though BTO (LSMO) are sketched in Figure 2(e). The parallel in-plane contribution from BTO (with resistivity approx. $10^7 \Omega \cdot \text{cm}$)³⁸ has been neglected due to the BTO in-plane resistance is expected to be approx. $2 \cdot 10^{11}$ larger than the LSMO one (with resistivity approx. $10^{-3} \Omega \cdot \text{cm}$),³⁹ as described in Supp. Inform. S1.

To analyze the effect of the series resistance associated to the LSMO electrode, on the measured ferroelectric loops, ferroelectric characterization is performed contacting two Pt electrodes separated by different distances, that is different lengths of the common LSMO bottom electrode. As shown in Figure 2(a-d), one needle is kept fixed on top of a given electrode and the other tip is systematically re-positioned at a different Pt electrode placed at different distance. In Figure 2(e), we sketch the electric field path for a couple of different top electrodes to illustrate that the arrangement is similar for any selected pair of contacts. In Figure 2(e), the electric field lines are represented by solid lines for closest contacted electrodes and by dashed lines for far contacted electrodes. We note that measurements on different electrodes must show similar ferroelectric properties as the BaTiO₃ film is continuous, although minor differences

could emerge because the probed BTO capacitors may not be identical in size and dielectric properties. However, importantly here, the current path along the bottom metallic electrode (LSMO) varies depending on the distance between the used top electrodes. If the contact Pt pads are faraway, the LSMO length (l_{LSMO})⁴⁰ is larger [dashed red line in Figure 2(e)], than if the contacted Pt electrodes are nearby [solid red line in Figure 2(e)], and consequently the series resistance of the bottom electrode is larger in the former case.

In Figure 2(f), we show the I - V loops recorded at $\nu = 50$ kHz for different distances between contacts ($l_{LSMO} = 85, 425$ and 850 μm). The I - V loop collected at $l_{LSMO} = 85$ μm shows clear ferroelectric switching peaks for negative and positive voltages. The wideness of the switching peak is inherent to the unavoidable double switching process in the top-top measurement configuration used here, as described in detail by Liu et al.³⁷ It can also be appreciated in Figure 2(f) that when increasing the length of the current path along the bottom electrode ($l_{LSMO} = 425$ and 850 μm), the switching gets gradually blurred and only a small bump in the I - V curve remains visible. The integration of the I - V data allows obtaining the polarization P - V loops shown in Figure 2(g). It can be clearly appreciated in Figure 2(g) that P - V loops display different zero-crossing voltages depending on l_{LSMO} . Precisely, V_c^* values increase when increasing l_{LSMO} (see Supp. Inform. S2). Moreover, one can notice that the P - V loops become gradually more round-shaped, when increasing l_{LSMO} . It will be shown that both effects are due to the important R_s contribution at the used measuring frequency. Indeed, the P - V loops collected at lower frequency for $l_{LSMO} = 85$ μm show well-saturated shape as seen in Supp. Inform. S3.

Figure 2. (a,b,c,d) Top view of the needles contacting to two top electrodes separated by $l_{LSMO} = 85, 240, 375$ and $500 \mu\text{m}$, respectively. (e) Sketch of the measurement configuration, the electric field lines in BTO (black) and the current lines in LSMO (red) for two top electrodes separated by different length (solid and dashed line). (f) I - V and (g) P - V loops measured at 50 kHz for different $l_{LSMO} = 85, 425$ and $850 \mu\text{m}$.

Taking into account that the l_{LSMO} increase produces an increase of the R_s , one can explain the apparent increase of V_c^* while increasing l_{LSMO} by two mechanisms. The first is that the increase

of R_s results in a larger voltage drop at LSMO, thus the voltage drop at BTO is smaller, i.e. the system can be modeled as a voltage divider. This produces an apparent increase of V_c^* , because the applied voltage partially drops at the LSMO layer. The second mechanism is that when increasing R_s , the time constant (τ_{RC}) of the circuit ($\tau_{RC} = R_s C$, where C is its capacitance) also increases. Therefore, the applied voltage is not the one falling across the BTO layer, because the cycling time ($\tau_{loop} = 1/\nu$) may be comparable or shorter than τ_{RC} . In this scenario, the actual voltage acting on the BTO capacitors is smaller and delayed than the applied voltage. In order to discriminate between these two effects, we perform I - V and P - V loops measurements at different frequencies. The voltage drop at the metallic LSMO should be frequency independent and thus different l_{LSMO} values should lead to the same I - V curves even if measured at different ν . In contrast, if the circuit delay time has some influence on the measurement, the I - V curves should be frequency dependent. We collected I - V data at lower and higher frequencies than that used in Figure 2 (f,g) ($\nu = 50$ kHz). In Figure 3(a) we show the I - V loops recorded at lower frequency ($\nu = 10$ kHz), for different l_{LSMO} . In Figure 3(a), it is shown that the ferroelectric switching peaks appear at lower voltage than in Figure 2(f) and the I - V loops show very similar shape irrespective to the selected l_{LSMO} ; only minor differences can be appreciated in the positive voltage switching current peak. The contribution of the leakage current close to the maximum applied voltage is also visible. The excellent agreement among I - V data collected at different places in the film [Figure 3(a)] confirms its quality and homogeneity and thus the different loops measured at higher frequency [Figure 2(f,g)] are not due spurious differences among electrodes. In addition, the similitude between the different I - V data collected at different l_{LSMO} at low frequency confirms that the BTO in-plane electric field can be neglected as discussed in Supp. Inform. S1. The corresponding P - V loops collected at different l_{LSMO} are shown in Figure 3(c); consistently,

they are also very similar. In Figure 3(b), we show the I - V data collected at higher frequency ($\nu = 100$ kHz). It can be observed that for $l_{LSMO} = 85$ μm , the ferroelectric switching peaks appear at larger voltage compared to the loops collected at 10 kHz [Figure 2(f,g)] and 50 kHz [Figure 3(a,c)]. Moreover, at 100 kHz, in the loops collected at larger l_{LSMO} , the ferroelectric switching peaks have completely vanished. The corresponding P - V loops, shown in Figure 3(d) become more rugby-ball-shape for increasing l_{LSMO} , and V_c^* increases with l_{LSMO} .

Overall, we note that the differences between loops collected at different l_{LSMO} become more apparent when the measurement frequency increases. This effect can be well understood by recalling that when the measuring frequency increases, τ_{loop} decreases and approaches the time constant τ_{RC} of the circuit. Thus, the effect of the time constant of the circuit is more prominent at high frequency. Therefore, the change of τ_{RC} associated to the increase of R_s when increasing l_{LSMO} has a major impact on the I - V data and P - V loops. We notice in Figures 3(b,d) that the influence of R_s is more remarkable when comparing data of $l_{LSMO} = 85$ μm and 425 μm than when comparing $l_{LSMO} = 425$ and 850 μm . This is due to the invers exponential dependence of the charging current and actual capacitor charge (Q) with the delay time [$Q(t) = Q_s(1 - e^{-t/\tau_{RC}})$], where Q_s is the saturated charge at given voltage. The calculated charging characteristics of the Pt/BTO/LSO/BTO/Pt capacitors are shown in Supp. Inform. S4.

On the other hand, it is well known that the coercive voltage of ferroelectrics depends on the measuring frequency.⁴¹ However, we have shown here that in the present case the l_{LSMO} contribution governs the observed frequency dependence of V_c^* , thus disregarding that the observed $V_c^*(\nu)$ variation results from an intrinsic frequency dependence of the V_c .

We conclude that the dependence of the shape of the I - V and P - V loops on l_{LSMO} for 10 kHz, 50 kHz and 100 kHz results from the variation of the circuit time constant with l_{LSMO} and the corresponding R_S variation. Quantitative evaluation of the relation between τ_{RC} and R_S with l_{LSMO} is summarized in Supp. Inform. S5.

Figure 3. (a,b) I - V and (c,d) P - V loops measured at 10 and 100 kHz, respectively, for different $l_{LSMO} = 85, 425$ and $850 \mu\text{m}$.

As a final crosscheck of the role of R_S , BTO/LSMO heterostructures were grown under nominally identical conditions but using thicker LSMO layer (20 nm). It is expected that the resistance of the bottom LSMO electrode will be lower due to its higher thickness. Indeed, four-probes measurements showed that it is about 10 times lower.⁴² We recorded again I - V and P - V loops at 50 kHz, using different l_{LSMO} distances between contacts. As shown in Figure 4(a,b), the measured I - V and P - V loops are identical for different l_{LSMO} . This observation further confirms that the effects of R_S on the I - V data vanishes if R_S decreases. Only a small increase in the V_c^* of

the positive ferroelectric switching peak in the I - V loop of Figure 4(a) can be observed.

Comparing the P - V loops of the thin [Figure 2(g)] and thick [Figure 4(b)] LSMO layer samples measured using $l_{LSMO} = 85\mu\text{m}$, we observe that for the thin LSMO layer, $V_c^* = 5.8$ V and this values decreases to $V_c^* = 3.4$ V for the thick LSMO layer. Again, the larger V_c^* value observed for the thinner LSMO film, is due to the larger R_s . Comparing the P_r values of the thin [$\approx 30 \mu\text{C}/\text{cm}^2$] LSMO and thick [$\approx 15 \mu\text{C}/\text{cm}^2$] measured for $l_{LSMO} = 85 \mu\text{m}$, an increase of the P_r value while decreasing the LSMO thickness can be also observed. In Supp. Inform. S6, P - V loops are simulated to better illustrate the effects of R_s .

Figure 4. (a) I - V and (b) P - V loops measured at 50 kHz for the BTO/LSMO (20nm) sample.

Figure 5(a,b) summarizes the V_c^* dependence on frequency and l_{LSMO} maps for thin and thick LSMO bottom electrodes. For the thin LSMO electrode [Figure 5(a)] the V_c^* strongly increases (red region) with frequency and l_{LSMO} . Instead, for the thick LSMO sample the V_c^* is near independent of these both parameters, and only a small increase with frequency can be observed, which most probably has an intrinsic origin.⁴¹ A fully equivalent set of results were obtained in BTO/LSMO/STO heterostructures, shown in Supp. Inform. S7. Therefore, the main message is that the dielectric measurements and ferroelectric characterization may be largely affected if the

bottom electrodes contributes with a large series resistance to the time constant of the measuring circuit. We also stress that as $\tau_{RC} = R_s C$, the role of R_s and the junction capacitance are similar.

Figure 5. V_c^* dependence on l_{LSMO} and frequency maps for the (a) BTO/LSMO (5 nm) and (b) BTO/LSMO(20nm) samples. V_c^* is extracted from the P - V loops collected at different l_{LSMO} and frequencies.

Overall, data show that an increase of R_s results in an extrinsic increase of V_c^* , particularly visible at high frequency. Having clarified the role of the R_s on the measured V_c^* , we focus on the control of V_c^* by magnetic field. The used bottom electrode LSMO is magnetic and it is known to display a large negative magnetoresistance, which depends on film thickness and temperature. Therefore, if its resistance decreases with the applied magnetic field one can expect, considering the aforementioned results, that the effective V_c^* also decrease. In Figure 6(a,b), we show the I - V and P - V loops of the BTO/LSMO(5 nm)/LSAT sample measured using largely spaced [$l_{LSMO} (= 850\mu\text{m})$] top electrodes. Data have been collected under no magnetic field ($\mu_0 H = 0$) and with an in-plane magnetic field of $\mu_0 H = 700\text{mT}$ applied along the [100] direction of the substrate. The current switching peak in the I - V characteristics shifts towards smaller voltage when magnetic field is applied (red line) compared with the measurement without

magnetic field (black line) as observed in the zoom inset shown in the inset of Figure 6(a). Consistently, the P - V loops [Figure 6(b)] also reveal that V_c^* shifts to smaller voltages as observed in Figure 6(b)(zoom inset). Therefore, application of a magnetic field allows increase/decrease the measured coercive voltage V_c^* . In Figure 6(c), we summarize the V_c^* data recorded collected sequentially while applying (blue background) or not (white background) a modest magnetic field. It can be observed that the V_c decreases of about 1% under magnetic field and that the V_c^* variation is fully reversible. In Supp. Inform. S8, the same experiments shown in Figure 6 for short l_{LSMO} ($=85\mu\text{m}$), does not reveal any visible change of V_c^* . In summary, the results in Figure 6, indicate that by applying magnetic field we can reduce the effective V_c^* voltage due to the negative magnetoresistance of LSMO that modifies R_S , allowing a sizeable control of *measured* ferroelectric properties by magnetic field. Note that P - V loop measurement under zero magnetic and under magnetic field has been already used to demonstrate intrinsic magnetoelectric coupling in cycloidal magnets in single-crystal⁶ or thin film form.⁴³

Figure 6. (a) I - V and (b) P - V loops measured at 50 kHz with and without in-plane applied magnetic field. (c) V_c^* values obtained from sequentially recorded P - V loops with (blue background) and without applied magnetic field for $l_{LSMO} = 850\mu\text{m}$.

We end by illustrating that the series resistance contribution R_S may have severe implications even when measuring I - V or P - V loops of non-ferroelectric materials. We consider first the effect

of R_S when measuring a linear dielectric. In Figure 7(a), we show the I - V curves measured on a commercial linear capacitor of about ≈ 50 pF. In Figure 7(a), one can observe that the I - V loop at 10 kHz shows a near square-like shape. The deviation from the square shape accounts for the R_S effect and corresponds to the typical shape of a delayed charged capacitor. The corresponding charge-voltage loops (Q - V)⁴⁴, where Q is the capacitor charge, recorded at several frequencies (100 Hz, 1 kHz and 10 kHz) are shown in Figure 7(b). It can be appreciated that for $\nu = 100$ Hz and 1 kHz, Q - V is non-hysteretic, as expected. However, at the highest frequency $\nu = 10$ kHz, the Q - V loop opens and non-zero *remnant-like* charge Q_r and *coercivity-like* V_c can be observed. As in the case of BTO film, the loop broadening occurs only at high frequency. Note that the deviation of the ideal linear curve occurs by the opening of the loop near the maximum applied voltage (red circle). As the used capacitor is a linear dielectric, it is obvious that the observed *remnant-like* charge Q_r and *coercivity-like* are spurious effects associated to R_S and the measuring conditions.

Lastly, we consider now the case of measuring ($\nu = 10$ kHz) a non-linear dielectric. In Figure 7(c), we show the calculated I - V expected when charging a capacitor having a non-linear dielectric represented by the polarization curve shown in Figure 7(d)(black line). We use a non-linear dielectric represented by a P - V loop of a “ferroelectric” material having $V_c = 0$ and $P_r = 30 \mu\text{C}/\text{cm}^2$ (black line); therefore although the assumed polarization is large under the applied voltage, the sample does not show any remnant polarization. This case would also be somehow equivalent to a ferroelectric sample having a very large depolarization field and no remanence. The I - V curve in Figure 7(c) has been calculated assuming a negligible series resistance $R_S = 0$ (black line). The simulation has been repeated [Figure 7(c) (red line)] including a series resistance ($R_S = 15 \text{ k}\Omega$). The most obvious difference is that now the maximum of I - V

does not occurs at $V = 0$ but it is shifted positively or negatively depending on the sense of the voltage excursion. This fake switching peaks give rise to hysteresis in the corresponding polarization data. Indeed, integration of this $I(V-t)$ data gives the *polarization-like* data shown in Figure 7(d) (red curve). It can be observed that now, a *remnant-like* polarization and a *coercive-like* voltage appear ($15 \mu\text{C}/\text{cm}^2$ and around 2.5 V , respectively). In brief, discriminating between the ferroelectric switching or the non-linear dielectric response of a capacitors requires a careful analysis of the frequency dependence of the $I-V$ data.

Figure 7. (a) $I-V$ loop measured at 100 Hz, 1 kHz, and 10 kHz for a 50pF capacitor. Lines through data points corresponds to data fitting according to expressions described in Supp. Inform. S6. (b) Corresponding, $Q-V$ loop of a 50pF capacitor measured at 100 Hz, 1 kHz, and 10 kHz . Simulated (c) $I-V$ and (d) $P-V$ loops of a non-linear dielectric (or a ferroelectric material with $V_c = 0$) with and without R_S .

3. Conclusions

We have shown that when measuring ferroelectric thin films grown on ultrathin metallic oxide electrodes, the electric resistance (R_S) of the bottom electrode, and in general that of the measuring circuit, may have a very severe impact on the current-voltage characteristics and thus on the extracted polarization loops. We have shown that beyond the obvious role of voltage divider associated to the series resistance, a subtler effect appears related to the time constant of the circuit (τ_{RC}) and its relation with the period of the measuring voltage $\tau (= 1/\nu)$. Our experimental results show that the series R_S contribution is more apparent at high frequencies, and it produces a fake increase of V_c^* in ferroelectric films. We have used the sensitivity of V_c^* to R_S to create a magnetoelectric device where the *coercive-like* V_c^* voltage can be modulate reversibly by a magnetic field. As the effect is increasingly sensitive when increasing frequency, the approach shown here can be useful to modulate polarization at high frequency. This strategy can be potentially useful in high frequency magnetoelectric switches and related devices.

ASSOCIATED CONTENT

Supporting Information

The following files are available free of charge.

Supporting_Information.pdf (file type: PDF): complementary data for the manuscript is included.

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Supporting Information of:

Direct reversible magnetoelectric coupling in ferroelectric/ferromagnetic structure controlled by series resistance engineering

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Supporting Information S1

To evaluate the current / electric field paths sketched in Figure 2(e) of the main manuscript, i) comparison between the in-plane parallel resistance across the BTO and LSMO layers and ii) comparison between the device resistance and the in-plane BTO resistance must be performed. Here, we perform both comparisons to demonstrate that the in-plane BTO contribution to the electrical measurements can be neglected.

i) Comparison between the in-plane parallel resistance across the BTO and LSMO layers:

To evaluate the parallel contribution from BTO to the electrical measurements, disregarded in the manuscript, comparison between the in-plane parallel resistance across the BTO and LSMO layers is quantitatively calculated. In-plane contributions of BTO and LSMO are evaluated by simplifying each one to different current paths (equivalent to parallel wires) represented in Figure S1(a). If it is assumed that the in-plane section of BTO layer (A_{BTO}) is the electrode edge ($l_{electrode}$) times its thickness (t_{BTO}) [Figure S1(b)] and the LSMO section (A_{LSMO}) is the electrode edge ($l_{electrode}$) times its thickness (t_{LSMO}) [Figure S1(c)], and that in both cases the length of the in-plane path is l_{LSMO} , the resistance of BTO (R_{BTO}) and LSMO (R_{LSMO}) can be written as:

$$R_{BTO} = \frac{\rho_{BTO} \cdot l_{LSMO}}{A_{BTO}} = \frac{\rho_{BTO} \cdot l_{LSMO}}{t_{BTO} \cdot l_{electrode}} \quad (1)$$

$$R_{LSMO} = \frac{\rho_{LSMO} \cdot l_{LSMO}}{A_{LSMO}} = \frac{\rho_{LSMO} \cdot l_{LSMO}}{t_{LSMO} \cdot l_{electrode}} \quad (2)$$

where ρ_{BTO} is the BTO resistivity and ρ_{LSMO} is the LSMO resistivity. With $t_{BTO} = 100\text{nm}$ and $t_{LSMO} = 5\text{nm}$, $\rho_{BTO} \approx 10^7 \Omega \cdot \text{cm}$,¹ $\rho_{LSMO} \approx 10^{-3} \Omega \cdot \text{cm}$,² the R_{BTO}/R_{LSMO} coefficient, corresponding to the in-plane parallel paths of R_{BTO} and R_{LSMO} sketched in Figure S1(d), is:

$$R_{BTO}/R_{LSMO} = \frac{\rho_{BTO} \cdot t_{LSMO}}{\rho_{LSMO} \cdot t_{BTO}} \approx 5 \cdot 10^8 \quad (3)$$

For the BTO sample with thick LSMO this value is 4 times larger. Thus, the difference between the BTO and LSMO resistance is big enough to neglect the parallel contribution from BTO.

ii) Comparison between the device resistance and the in-plane BTO resistance

To ensure that the sketched current paths of the main manuscript [Figure 2(e)] is correct the R_{BTO} resistance in front of the device resistance (R_{device}) should be also evaluated. With $l_{electrode} = 50 \mu\text{m}$, $l_{LSMO} = 85 \mu\text{m}$ (which corresponds to the minimum evaluated l_{LSMO} and thus the minimum resistance), $t_{BTO} = 100 \text{ nm}$ and $\rho_{BTO} = 10^7 \Omega \cdot \text{cm}$,¹ and, using equation (1), $R_{BTO} \approx 10^{12} \Omega$ is obtained. The device resistance can be written as:

$$R_{device} = \frac{\rho_{BTO} \cdot t_{BTO}}{A_{electrode}} = \frac{\rho_{BTO} \cdot t_{BTO}}{l_{electrode}^2} \quad (4)$$

where $A_{electrode}$ is the Pt electrode area. Thus, $R_{device} \approx 10^6 \Omega$, and $R_{BTO}/R_{device} \approx 10^6$. It can be concluded that the difference between the BTO and LSMO resistance is big enough to neglect the parallel contribution from BTO.

Figure S1. (a) Sketch of the in-plane contribution from BTO and LSMO to the current path. Note that both contributions are in parallel. (b) Sketch of the BTO current path geometry. (c) Sketch of the LSMO current path geometry. (d) Equivalent circuit based on materials resistance.

In brief, we have shown that the R_{BTO} is very large compared with the R_{LSMO} and R_{device} allowing to disregard the in-plane electric field across the BTO between measured electrodes.

Supporting Information S2

Figure S2. V_c^* dependence on I_{LSMO} for different groups of electrodes in the same sample. It can be observed that V_c^* increases with I_{LSMO} until saturation because V_c^* is approaching the maximum applied voltage.

Supporting Information S3

Figure S3. (a) P - V loops for $I_{LSMO} = 85 \mu m$ collected at 1 kHz and several maximum applied voltages (V_{max}). Dependence of (b) V_c and (c) P_r on V_{max} showing intrinsic V_c of 1.6 V and P_r of 10 $\mu C/cm^2$.

Supporting Information S4

Figure S4. Q - t characteristics of a R_sC circuit. The arbitrary τ_0 of the circuit has been modified for the three displayed curves with values relevant in the measurements shown in Figure 2(g,h). The used relation is $Q(t) = Q_s(1 - e^{-t/\tau})$, where Q and Q_s are the actual and the saturated charge, respectively, and $\tau = R_sC$.

Supporting Information S5

In Figure S5 the time constant and the extracted R_S for several I_{LSMO} is quantitatively evaluated. In Figure S5(a), the current versus time measured while applying the decreasing branch of a triangular voltage pulse of 100 kHz equivalent frequency is plotted for several I_{LSMO} . It can be observed that the decay time is larger for increasing I_{LSMO} . Solid lines correspond to exponential fitting ($J=A \cdot e^{t/\tau_{RC}}$). The extracted τ_{RC} corresponding to the time constant of the circuit is calculated and it has been plotted as a function of I_{LSMO} in Figure S5(b). It can be observed that the scales with the I_{LSMO} . Assuming that the characterized measurement circuit corresponds to the BTO capacitor (C_{BTO}) in series to the LSMO resistance (R_S), as demonstrated in Supp. Inform. SX, the $\tau_{RC} = R_S \cdot C_{BTO}$. C_{BTO} has been estimated from P - V loops collected at low frequency (1 kHz) and show in Figure S5(c). In Figure S5(c), the P - V loops collected at maximum applied voltage (V_{max}) well-below coercive voltage ($V_{max} = 1V$) and well-above coercive field ($V_{max} = 8V$) are plotted. From the slope of the loop collected at $V_{max} = 1V$, $\epsilon = 200$ is obtained, corresponding to the permittivity at small excitation voltage. From the slopes near the positive and negative V_{max} of the loop collected at $V_{max} = 8V$, $\epsilon = 165$ and $\epsilon = 135$ are obtained, corresponding to the permittivity at positive and negative saturation, respectively. Being the permittivity at saturation the one that should correspond to the experiment displayed in Figure S5(c), $\epsilon \approx 150$ has been used for C_{BTO} calculation:

$$C_{BTO} = \frac{\epsilon \cdot A_{BTO}}{t_{BTO}}$$

where $A_{BTO} = 50 \times 50 \mu m^2$ is the electrode area and $t_{BTO} = 100$ nm the BTO capacitor thickness. Thus $C_{BTO} = 30$ .pF. Using this value the R_S is calculated using $\tau_{RC} = R_S \cdot C_{BTO}$. R_S versus the I_{LSMO} is plotted in Figure S5(d). It can be observed that the R_S scales well with I_{LSMO} , reinforcing the assumption that R_S corresponds to the series resistance of the LSMO layer. Indeed, using the data points fitting shown in Figure S5(d) and using the geometry discussed in Supp. Inform. Figure S1 for the LSMO resistance, LSMO resistivity ≈ 1 m $\Omega \cdot cm$ is obtained, which is in good agreement with reported values.²

Figure S5. (a) Current versus time measured for the decreasing voltage branch of a triangular voltage pulse of $V_{max} = 8$ V of equivalent frequency of 100 kHz. Lines through data points correspond to exponential decay fitting using the expression: $J = A \cdot e^{-t/\tau_{RC}}$. (b) τ_{RC} versus l_{LSMO} . τ_{RC} has been obtained from data fitting displayed in panel (a). (c) P - E loops measured and corresponding slope fitting. (d) R_s obtained from τ_{RC} values dependence on l_{LSMO} .

Supporting Information S6

From the comparison of data for the thick LSMO (20 nm) and thin LSMO (5 nm) samples shown in Figure S6(a), it can be observed that the thick LSMO (20 nm) sample shows larger V_c and P_r than the thin LSMO (5 nm) sample. Similar V_c and P_r increase is observed in the simulated P - V loops shown in Figure S6(b) comparing the loop with low R_s (38 k Ω) with the one of high R_s (470 k Ω). Thus, it can be concluded that the main contribution that produces the V_c and P_r increase for the thin LSMO sample is the increase of the LSMO resistance and thus the R_s -contribution. It can be also observed that additional V_c and P_r increase for the thin LSMO sample, which can be ascribed to the intrinsically different nature of the BTO layer due to small morphological/structural differences. While comparing the simulated P - V loops of Figure S6(b), it can be also observed that at 50 kHz the R_s -contribution for $R_s = 38$ k Ω is not relevant. However, the increasing effect of R_s for increasing frequency can be inferred from the noticeable difference of the simulated P - V loops of Figure S6(c), and the strong V_c dependence on frequency shown in Figure S6(d). It can be concluded that R_s -contribution results in a V_c and P_r increase (if polarization is fully saturated) and that this effect increases with frequency.

Figure S6. (a) Experimental P - V loops measured at 50 kHz for the thin LSMO (5 nm) and thick LSMO (20 nm) samples at $l_{\text{LSMO}} = 85\text{ }\mu\text{m}$. Loops correspond to the data shown in Figures 1(g) and Figure 4(b). (b) Simulated P - V loops (a) with $R_s = 38\text{ k}\Omega$ and $470\text{ k}\Omega$, corresponding to the resistance values obtained for the thin LSMO (5 nm) and thick LSMO (20 nm) samples using two probes DC measurements, respectively, and without R_s -contribution at 50 kHz. (c) Simulated P - V loops with $R_s = 470\text{ k}\Omega$, corresponding to the resistance values obtained for the thick LSMO (20 nm), at several frequencies. (d) V_c dependence on frequency of loops displayed in panel (c). It shows the increase of V_c with frequency due to R_s -contribution.

Ferroelectric loops simulation has been done using the relations for dielectric, ferroelectric, leakage and series resistance contributions. The overall current is obtained by:

$$I = \frac{dP\chi}{dt} + \frac{dP_{FE}}{dt} + I_{leakage}$$

where $P\chi$ corresponds to the non-switchable polarization intrinsic from any dielectric material, P_{FE} to the switchable one, and $I_{leakage}$ corresponds to the leakage current. These three different contributions are:

$$P\chi = \varepsilon_s E$$

where ε_s is the permittivity at saturation and E is the applied electric field,

$$P_{FE} = P_0 \cdot \tanh\left(\frac{E - E_c}{\delta}\right)$$

where E_c is the coercive field and δ is the broadness of the coercive field, and

$$I_{leakage} = I_0 \cdot E \cdot e^{\sqrt{E/E_0}}$$

Where I_0 and E_0 are parameters that correspond to the magnitude and the onset voltage of the leakage current. This relation corresponds to the Poole-Frenkel conduction mechanism, which we found in good agreement with our experimental data at high electric field regime. In the simulated data, this term contribution is negligible, but we include it here for completeness.

The series resistance effect is calculated for each step (n) of the loop using the following expression that takes into account the delay introduced by the series resistance:

$$P_n = P_{n-1} + (P_{n,RS=0} - P_{n-1}) \cdot (1 - e^{-t/R_s C})$$

where $P_{n,RS=0}$ is the polarization before taking into account the R_s contribution.

The electric field to V conversion for the plotted in the manuscript data is done accordingly to $E = V/t$.

Further details can be found in ref. 3.

Supporting Information S7

Figure S7. (a,b) I - V and (c,d) P - V loops measured at 50 kHz for different $l_{LSMO} = 85, 425$ and $850 \mu\text{m}$ for BTO/LSMO//STO samples with LSMO thicknesses of 5 and 20 nm, respectively. V_c^* dependence on l_{LSMO} and frequency maps for the (e) BTO/LSMO (5 nm) and (f) BTO/LSMO(20 nm) samples. Although the V_c^* variations here are smaller, these fully agree with those found in BTO/LSMO//LSAT samples reported in the main manuscript.

Supporting Information S8

Figure S8. (a) I - V and (b) P - V loops measured with and without in-plane applied magnetic field. (c) V_c^* values obtained from sequentially recorded P - V loops with (blue background) and without applied magnetic field for $l_{\text{LSMO}} = 85\mu\text{m}$. No dependence on magnetic field of V_c^* is here observed.

SUPPORTING INFORMATION REFERENCES

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