Electronic Supplementary Information: Direct magnetoelectric effect in ferroelectric - ferromagnetic epitaxial heterostructures

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Supplementary Information 1

In Figures S1 we show cross section Scanning Transmission Electron Microscopy (STEM) images of BTObot and BTOtop heterostructures. The morphology and thickness of the distinct layers in each structure can be clearly appreciated.

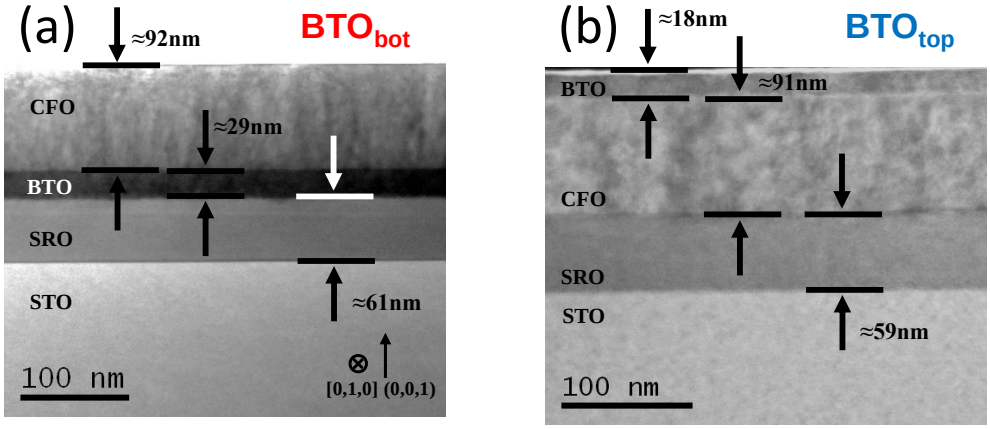

Figure S1. Bright field Scanning Transmission Electron Microscopy images of cross sections of BTO_{bot} (a) and BTO_{top} (b). Both BTO_{bot} (mechanical polishing preparation) and BTO_{top} (FIB preparation) samples have been observed in [010] zone axis orientation.

Supplementary Information 2

Room-temperature magnetization M(H) loops (Figures S2) recorded using a Superconducting Quantum Interference Device (SQUID) magnetometer. Measurements have been taken with the magnetic field applied both in-plane and out-of-plane. Data in Figures S2(a,b) show both samples have very similar magnetic response. Loops are also similar to those reported for single CoFe2O4 thin films on SrTiO3 (001) substrates [S2.1].

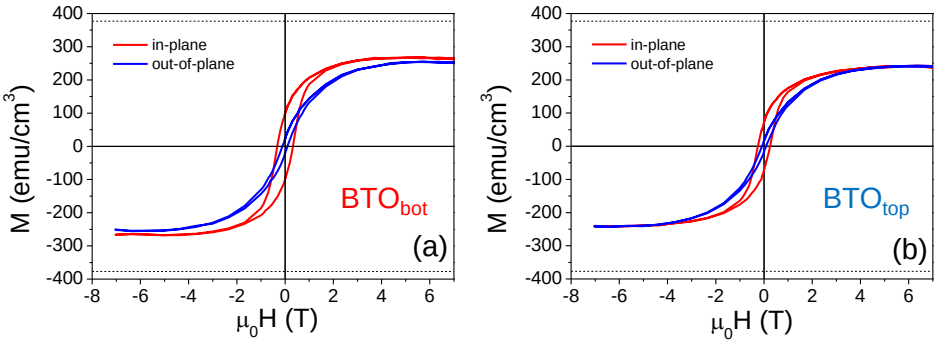

Figure S2. M(H) loops recorded at room temperature of (a) BTO_{bot} and (b) BTO_{top} samples. The magnetization of bulk CFO is indicated (dashed lines).

[S2.1] F. Rigato, J. Geshev, V. Skumryev, and J. Fontcuberta, J. Appl. Phys. **2009**, 106, 113924.

Supplementary Information 3

We have performed temperature-dependent X-ray diffraction experiments focusing on the (002) BTO and (004) CFO reflections of BTObot and BTOtop bilayers. Experiments were conducted on BTObot and BTOtop heterostructures having similar thicknesses as those described in the manuscript (CFO 100 nm and BTO 25 nm) but having La2/3Sr1/3MnO3 (LSMO) as bottom electrode (STO//LSMO/BTO/CFO and STO//LSMO/CFO/BTO, respectively). The observed linear temperature evolution of the STO (002) reflection assessed the quality of the experimental protocol [S3.1]. In Figures S3(a) and S3(b) we show the temperature evolution of the BTO (left axes) and CFO (right axes) out-of-plane parameters in the 90 K – 470 K temperature range, for BTOtop and BTObot respectively. In Figures S3 dashed vertical lines indicate the temperatures (T1, T2 and T3) where BTO bulk has its structural transitions and solid vertical lines indicate the temperatures (T1', T2' and T3') where changes of the slope of out-of-plane lattice parameter (of both BTO and CFO) vs temperature dependence are appreciated. It is clear from these data that in BTObot the temperatures T1', T2' and T3' are all shifted below its corresponding bulk values by about approx. 30 K. In contrast, in BTOtop the temperatures T1', T2' and T3' coincide with bulk T1, T2 and T3 transitions. This is exactly the same behavior as observed in samples of Figures 5. The most remarkable feature is the coincidence of the temperatures at which the slope of out-of-plane lattice occur for BTO and CFO, confirming the existence of structural coupling among the BTO and CFO layers, thus providing a solid ground for the understanding of the dielectric anomalies shown in Figures 5(a,b).

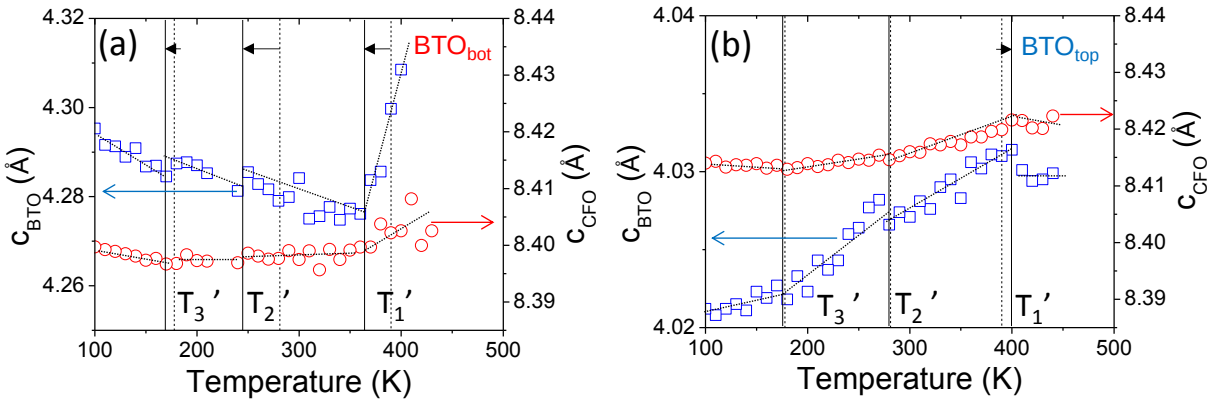

Figure S3. Temperature dependence of the out-of-plane (c-axis) cell parameters of BTO (left axes) and CFO (right axes) of (a) BTO_{bot} and (b) BTO_{top} heterostructures grown on STO(001) buffered with a thin LSMO (15 nm) layer. Vertical dashed lines indicate BTO-bulk transitions and vertical lines indicate changes of the slope of $c_{BTO}(T)$. Lines are eye-guides.

[S3.1] P.-E. Janolin et al., Appl. Phys. Lett. **2007**, 90, 212904.

Supplementary Information 4

Magnetization measurements were performed by using a Superconducting Quantum Interference Device (SQUID) magnetometer from QD.

Magnetic moment vs. temperature curves m(T), have been collected with the magnetic field (10 Oe) applied in-plane. At 300 K and prior to m(T) measurements, the samples were demagnetized (remanence < 1 x10-5 emu). In Figures S4(a) and (b) we show the magnetic moment and its temperature derivative respectively, of the BTObot measured when heating the sample in the 300 K – 400 K temperature range. It can be observed that both m(T) and its derivative dm(T)/dT have a noticeable anomaly at ≈ 360 K. It is worth noticing that the anomaly of m(T) observed in BTObot matches perfectly with T1' (solid vertical line) determined by magnetodielectric measurements of Figure 5(c). Data in Figures S4 indicate that the converse ME response can also be observed in this 2-2 system.

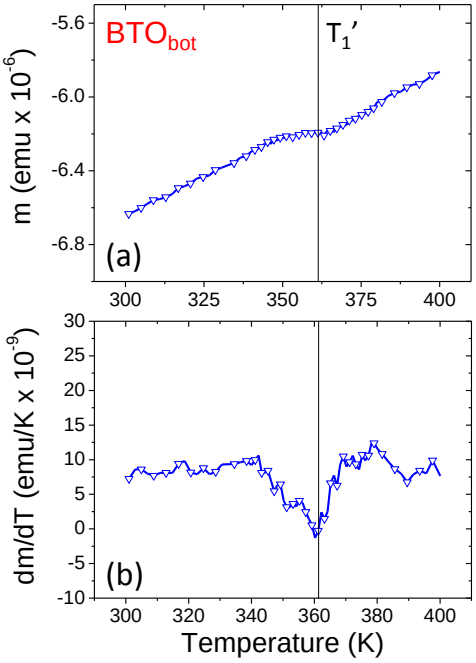

Figure S4. Temperature dependence of the magnetic moment (a) and its temperature derivative dm(T)/dT (b) of BTO_{bot} sample.