

Electronic Supplementary Information

Control of up-to-down/down-to-up light induced ferroelectric polarization reversal

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Table S1. Summary of LSMO+LNO (which overlap) LNO, SRO and BTO lattice parameters, for BTO/LSMO, BTO/LNO and BTO/SRO samples extracted from RSM data of Figure 1. (*) indicates that bulk values are for LSMO.

Diffraction peak	Parameter	Bulk	BTO/LSMO/LNO	BTO/LNO	BTO/SRO/LSMO
LAO	a (Å)	3.79			
	c (Å)	3.79			
LSMO+LNO	a (Å)	3.88*	3.805		
	c (Å)	3.88*	3.956		
LNO	a (Å)	3.83		3.79	3.79
	c (Å)	3.83		3.939	3.936
SRO	a (Å)	3.93			3.95
	c (Å)	3.93			3.96
BTO	a (Å)	3.994	4.003	4.005	4.016
	c (Å)	4.038	4.110	4.093	4.090

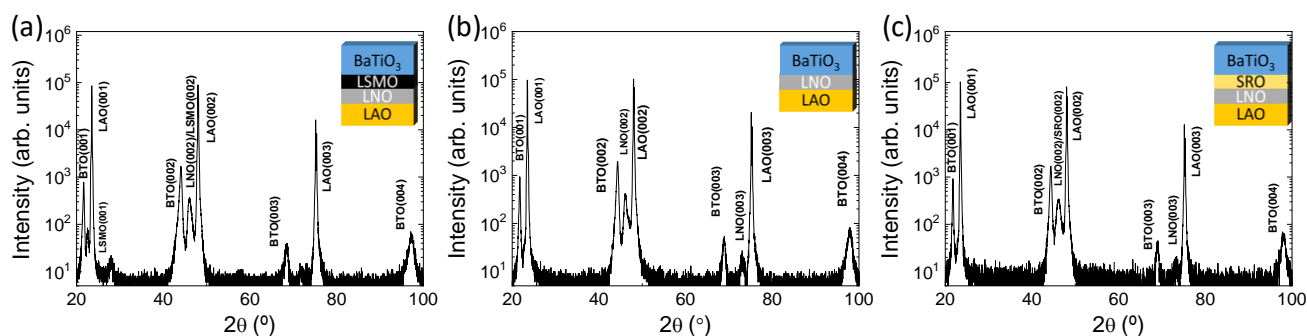


Fig. S1 XRD θ - 2θ specular scan of the (a) BTO/LSMO, (b) BTO/LNO and (c) BTO/SRO samples.

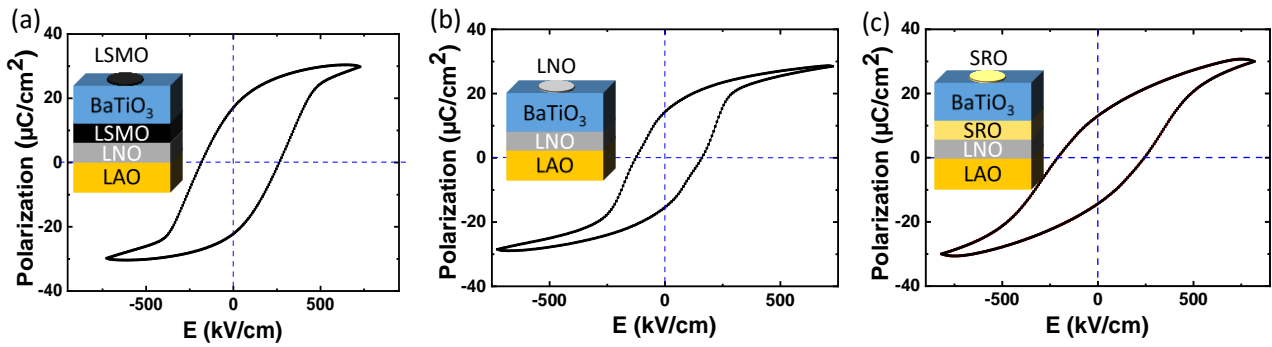


Fig. S2 (a,b,c) Ferroelectric hysteresis P-E loops of the BTO/LSMO, BTO/LNO and BTO/SRO samples, respectively. The loops are collected grounding the bottom electrode and biasing the top one, i.e. LSMO, LNO or SRO depending on the sample. In this configuration, the ferroelectric nature of the three films is clearly revealed by the shown plots. Note that E_{imp} resulting from the ferroelectric/electrode interface cancels out.⁶ Therefore, in this configuration, only the E_{imp} resulting from the bulk of the film remains and it can be observed that it is negligible.

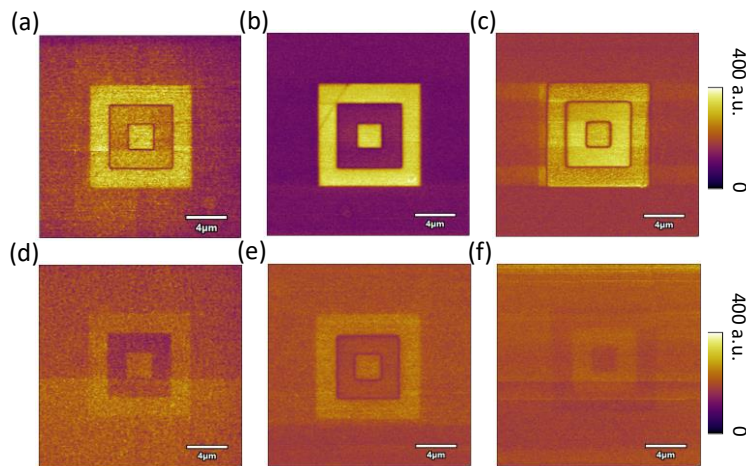


Fig. S3 PFM amplitude images collected (a,b,c) before and (d,e,f) after illumination for the BTO/LSMO, BTO/LNO and BTO/SRO samples, respectively. Note that in all panels there is some contrast related to charging specially for the sample for the BTO/LNO sample in panel b. Charging does not preclude the observation of near zero signal in panels (a,b,c,e) at the domain walls as expected for intrinsic ferroelectric response.

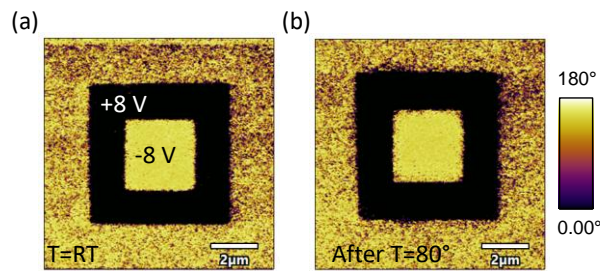


Fig. S4 PFM phase images collected (a) before and (b) after heating up to 80°C during 10 min for the BTO/LSMO sample.

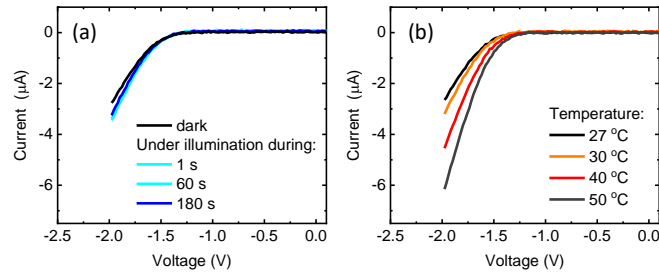


Fig. S5 (a) I-V characteristics in dark and under illumination during indicated time. (b) I-V characteristics at indicated temperatures.

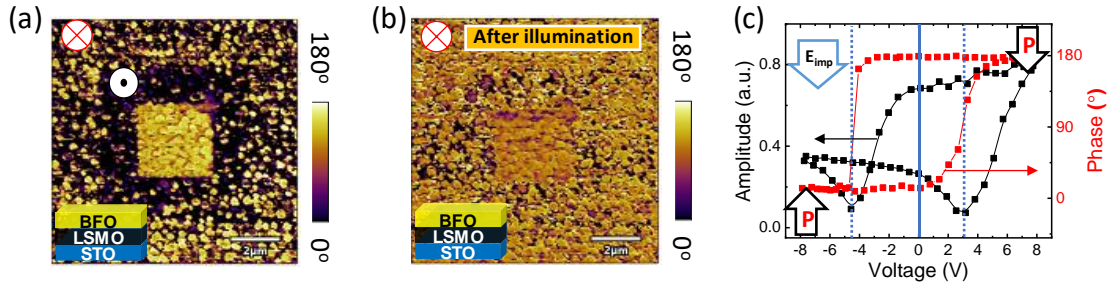


Fig. S6 (a, b) PFM phase images collected before and after illumination, as indicated, for a BFO/LSMO//SrTiO₃(001) sample grown by chemical vapor deposition. (c) PFM amplitude and phase loops.

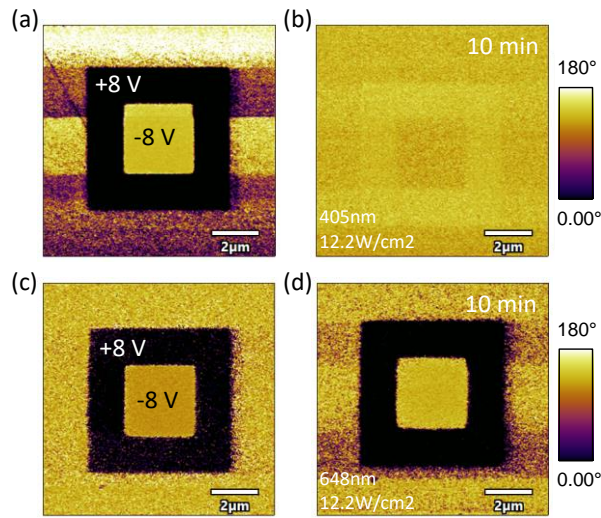


Fig. S7 PFM phase images collected (a) before and (b) after illumination of 405 nm and 12.2 W/cm² for the BTO/LSMO sample. PFM phase images collected (c) before and (d) after illumination of 648 nm and 12.2 W/cm² for the BTO/LSMO sample.

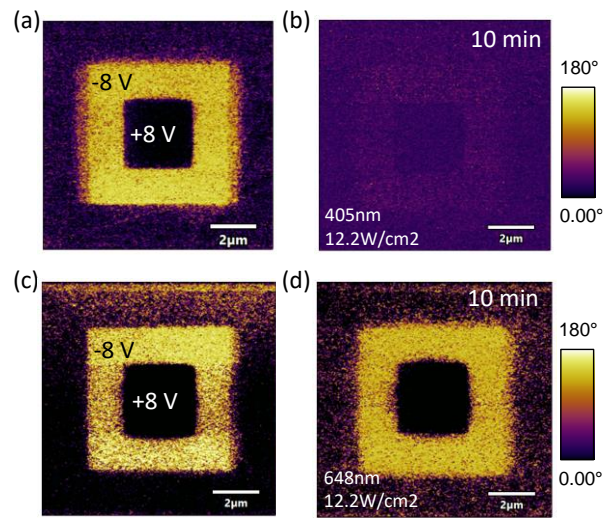


Fig. S8 PFM phase images collected (a) before and (b) after illumination of 405 nm and 12.2 W/cm² for the BTO/SRO sample. PFM phase images collected (c) before and (d) after illumination of 648 nm and 12.2 W/cm² for the BTO/SRO sample.

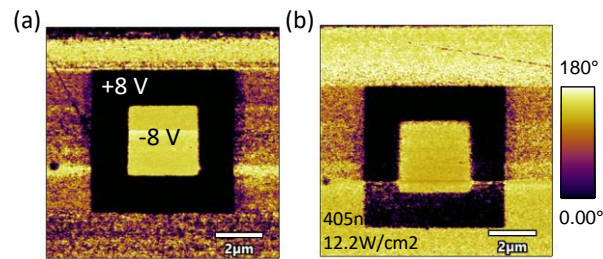


Fig. S9 PFM phase images collected (a) before and (b) after illumination of 405 nm and 12.2 W/cm² for the BTO/LSMO sample 1 mm far from the illuminated area.

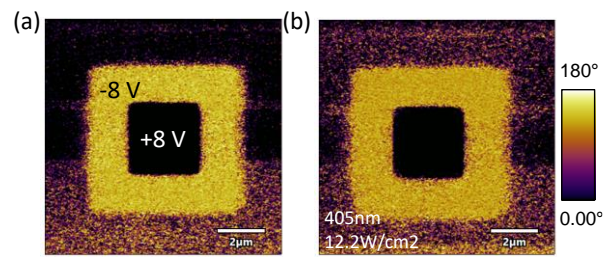


Fig. S10 PFM phase images collected (a) before and (b) after illumination of 405 nm and 12.2 W/cm² for the BTO/SRO sample 1 mm far from the illuminated area.

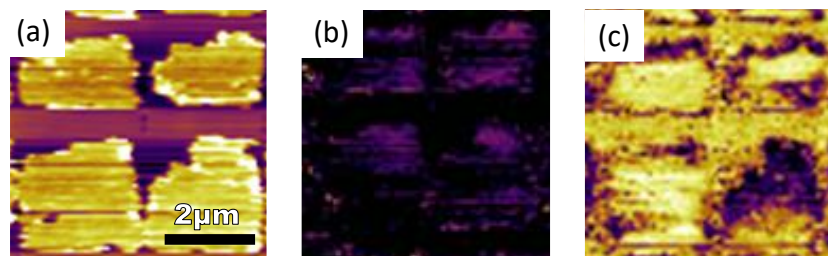


Fig. S11 (a) Topography image of near 3 by 3 μm² contacts. PFM phase images collected (b) before and (c) after illumination of 405 nm and 12.2 W/cm² for the BTO/LSMO sample. In (c), it can be observed that the electrodes where the polarization was prepoled towards down [dark in (b)] have switched to bright. Thus, as mentioned in the description of Figure 2 of the main manuscript polarization can switch from down to up in the BTO/LSMO sample after illumination also in presence of Pt top contact.

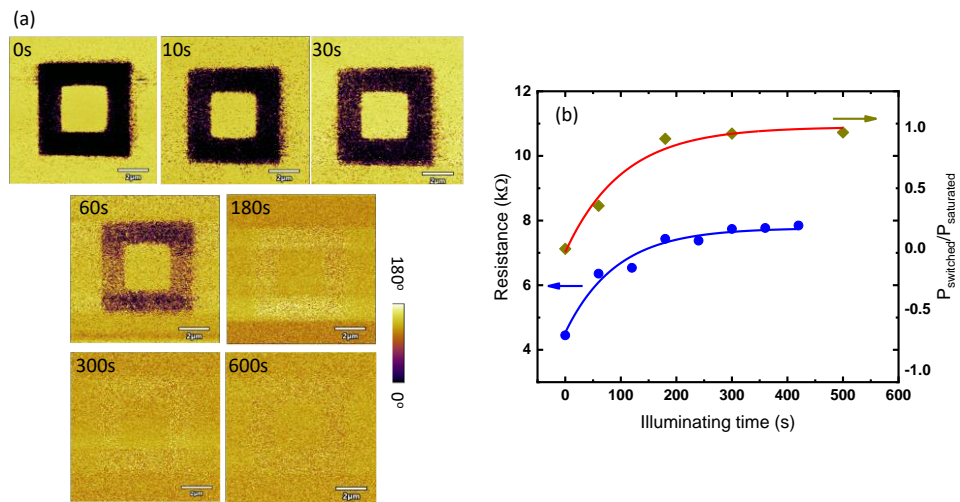


Fig. S12 (a) PFM phase images collected after indicated illumination time for the BTO/LSMO sample. (b) Resistance as a function of illumination time in Pt/BTO/LSMO junctions. Lines through data points are a guide for the eye.