

Supporting Information for

**Optical Switching of Robust Ferroelectric Polarization on Epitaxial Hf<sub>0.5</sub>Zr<sub>0.5</sub>O<sub>2</sub> Integrated with BaTiO<sub>3</sub>**

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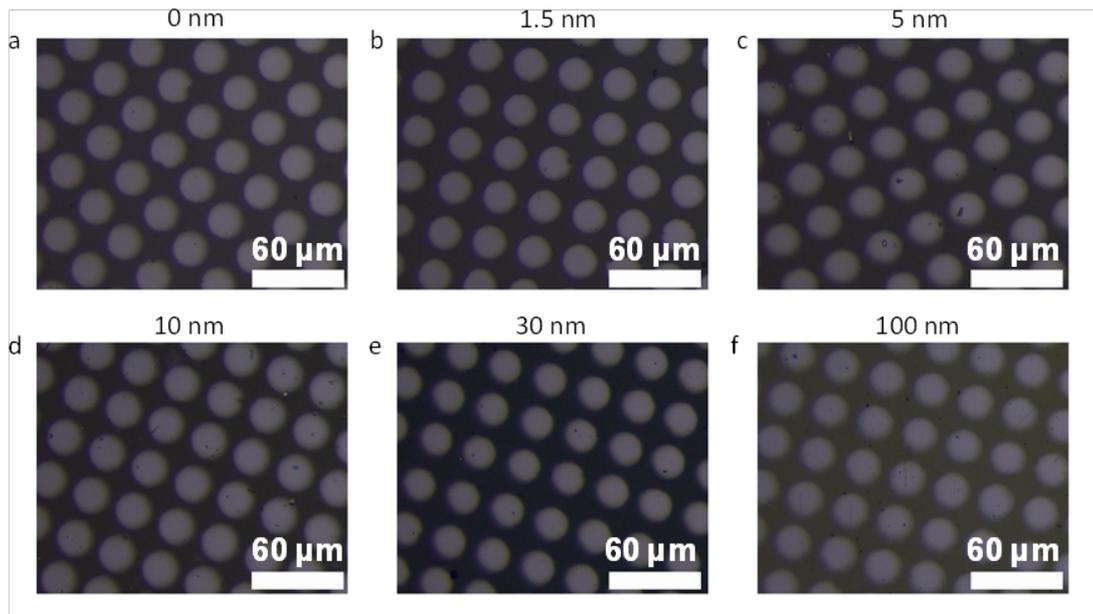
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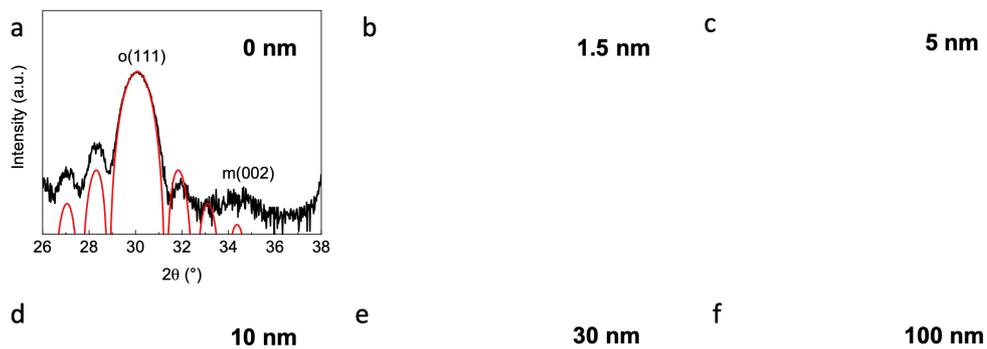
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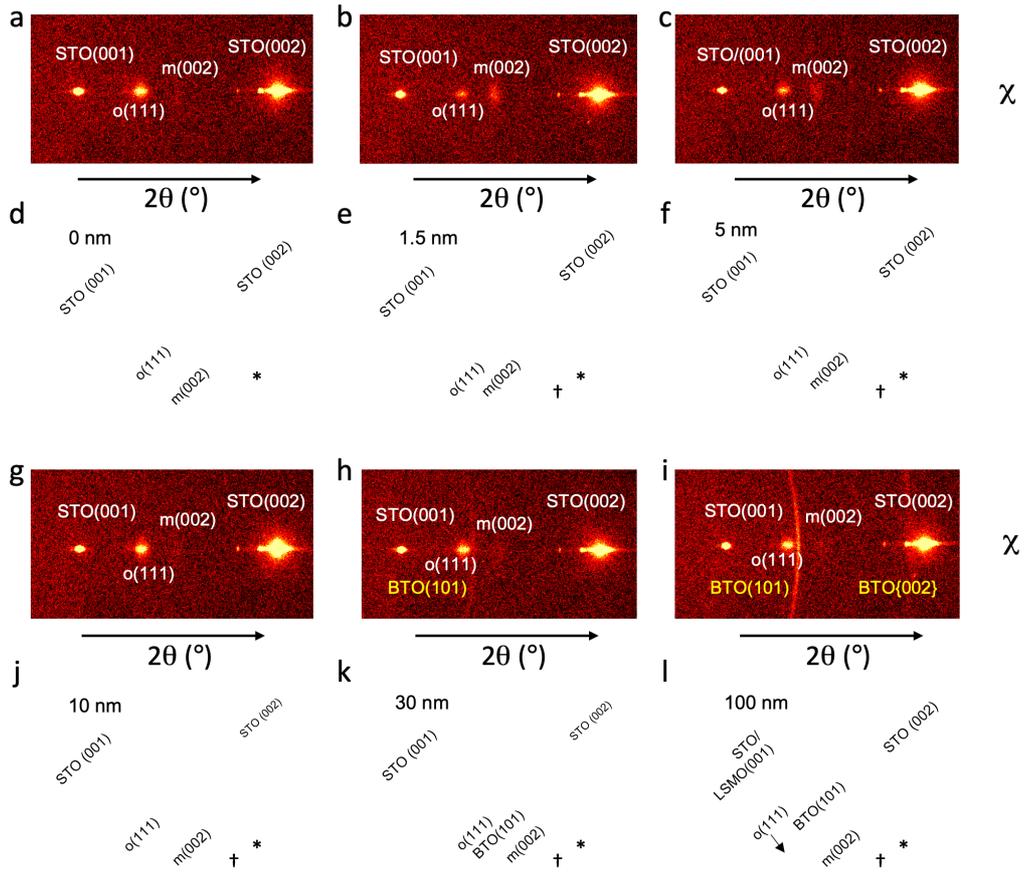
**Supplementary Figures and Tables**



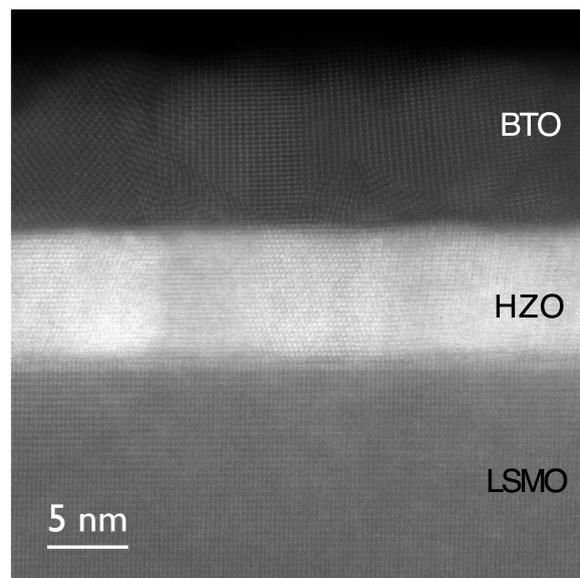
**Fig. S1 (a-e)** Optical image (top view) of the deposited Pt electrodes of the samples with BTO thickness of 0, 1.5, 5, 10, 30 and 100 nm, respectively



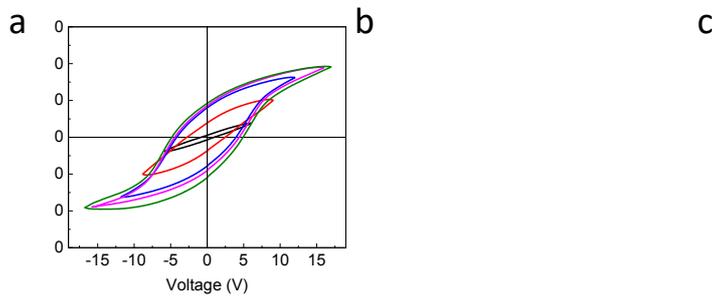
**Fig. S2 (a-e)** XRD  $\theta-2\theta$  scans (measured with a point detector ) of the samples with BTO thickness of 0, 1.5, 5, 10, 30 and 100 nm, respectively



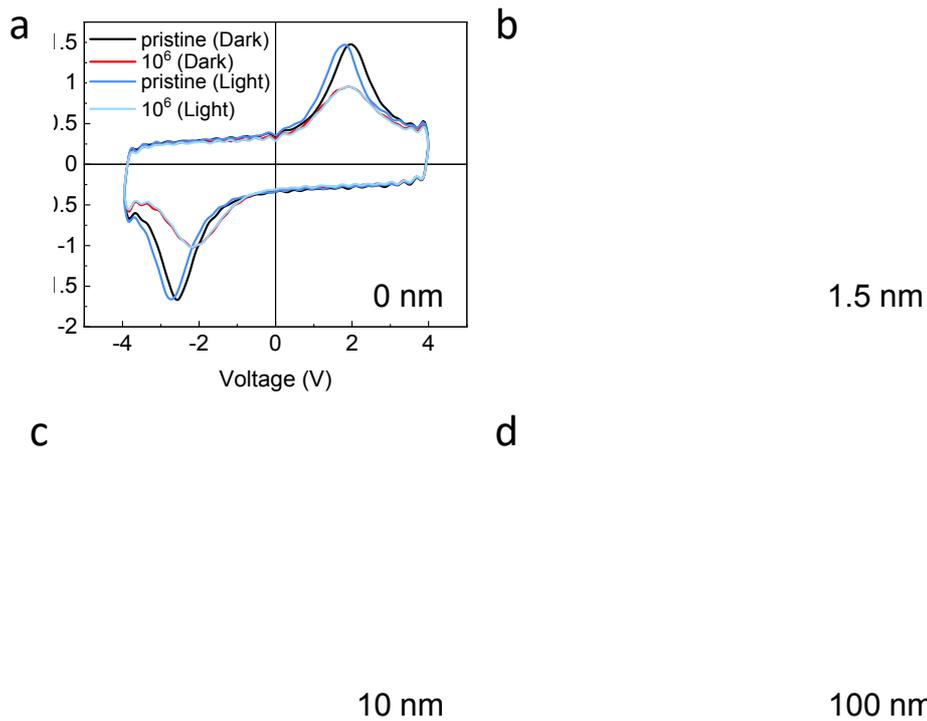
**Fig. S3 (a-l)** XRD  $2\theta - \chi$  images and integrated, in the  $\chi = \pm 10^\circ$  range around  $\chi = 0^\circ$ ,  $\theta - 2\theta$  scans of samples with BTO thickness of 0, 1.5, 5, 10, 30 and 100 nm as indicated. The † and \* symbols mark the positions of the STO(002) reflections due to spurious Cu-K<sub>b</sub> and W-L lines, respectively



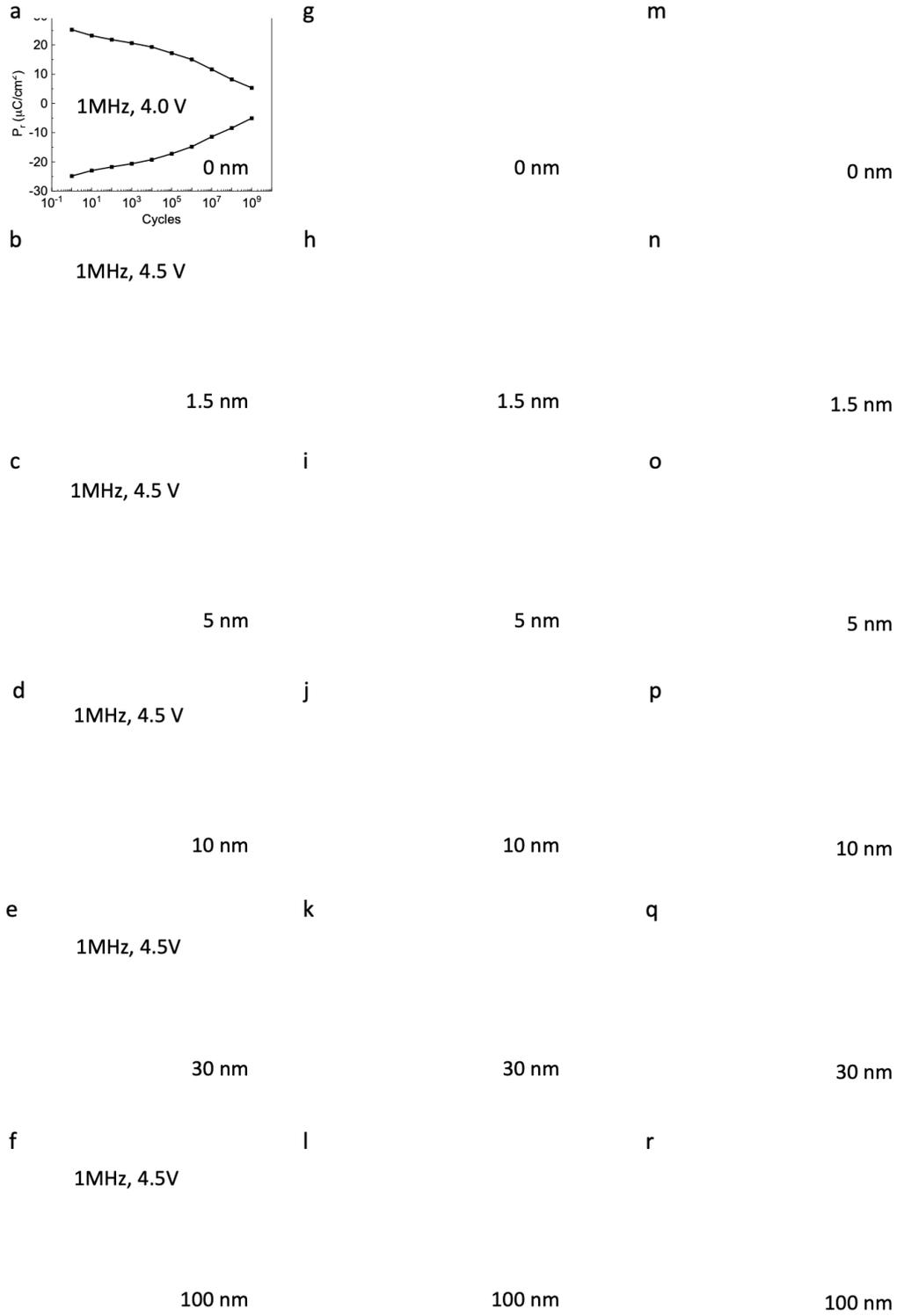
**Fig. S4** Atomic resolution HAADF images of a representative region where the good crystalline quality of the BTO layer can be observed



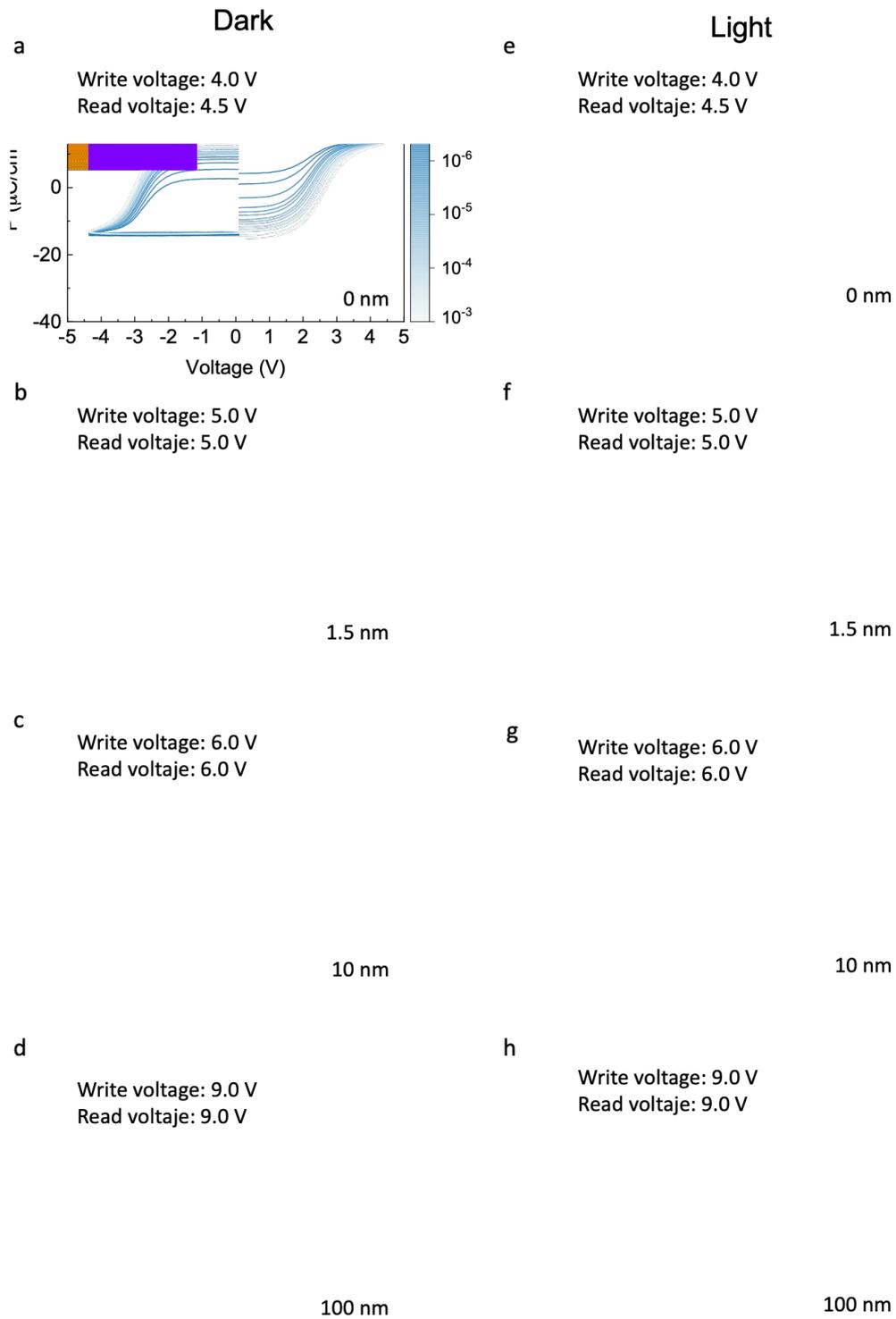
**Fig. S5** (a) P-V loops for 100 nm BTO sample without HZO at increasing voltage. (b) Leakage current for 100 nm BTO sample. (c) Endurance plots at 1 MHz and 10 V cycling voltage for 100 nm BTO sample



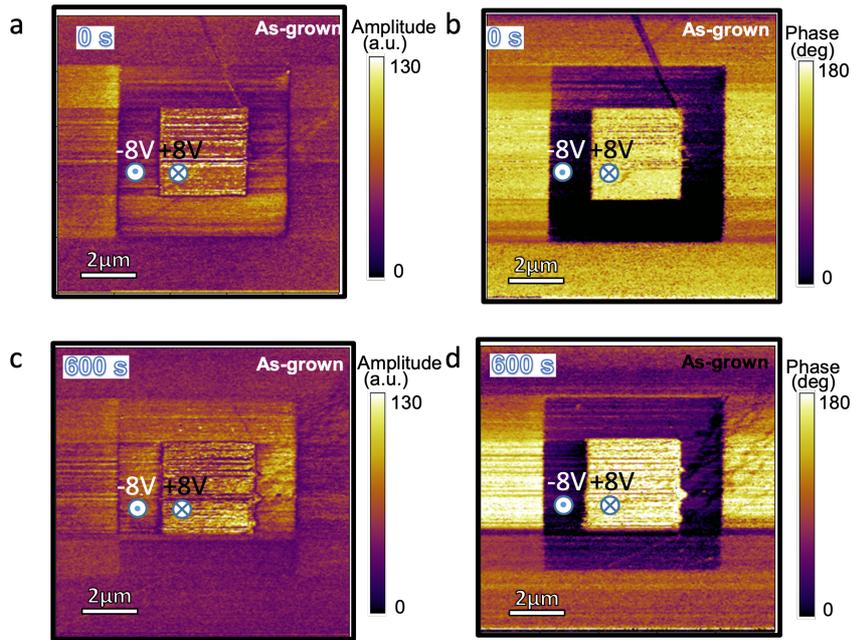
**Fig. S6** (a-d) Corresponding to Fig. 3(a-d), I-V loops of HZO/BTO samples with BTO thickness of 0, 1.5, 10 and 100 nm, respectively with and without illumination and in the pristine state and after  $10^6$  cycles



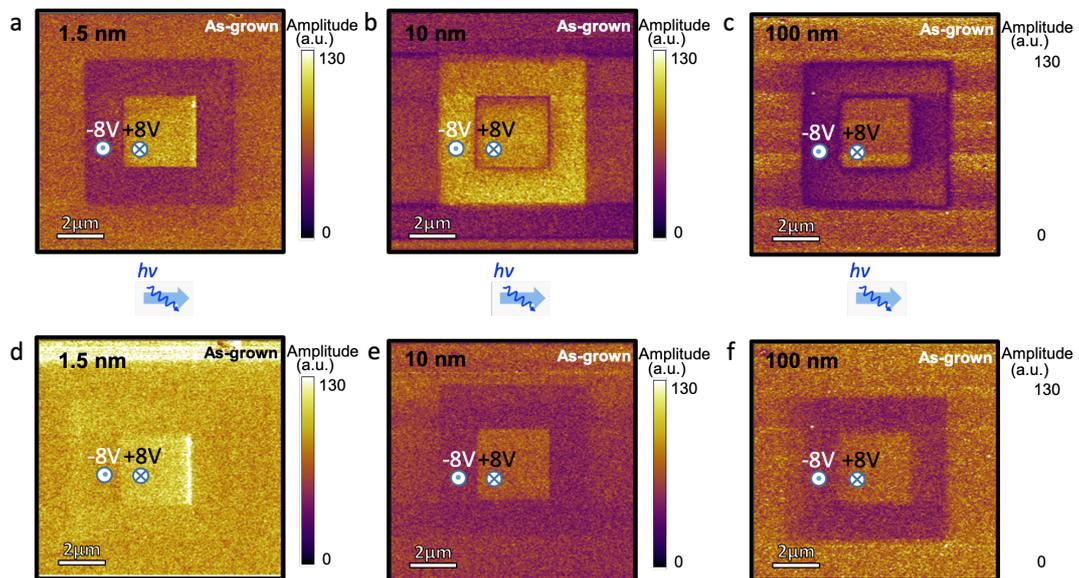
**Fig. S7 (a-e)** Endurance plots at 1 MHz and indicated voltage of samples with BTO thickness of 0, 1.5, 5, 10, 30 and 100 nm. **(f-k)** P-V and **(l-q)** I-V loops corresponding to data of **a-e**



**Fig. S8 (a-d)** Polarization in dark and **(e-h)** under illumination loops for reading in the switching spectroscopy experiments. Asymmetric loops account for the presence of imprint electric fields

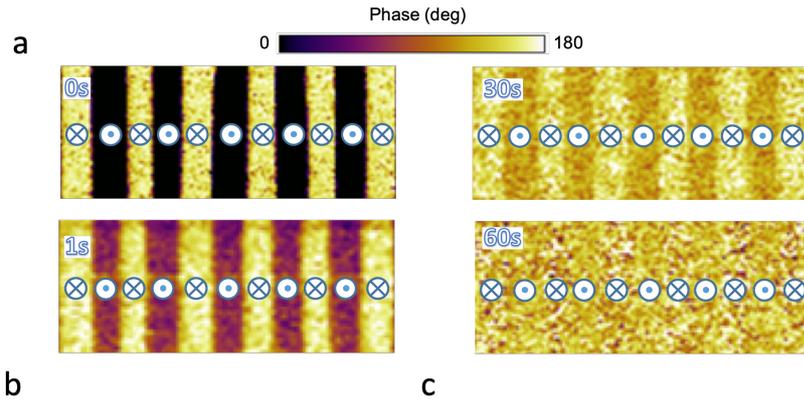


**Fig. S9** (a) PFM amplitude and (b) phase images collected just after poling, respectively, for a HZO single film without BTO. (c, d) Idem collected after waiting 10 min (600 s) under illumination

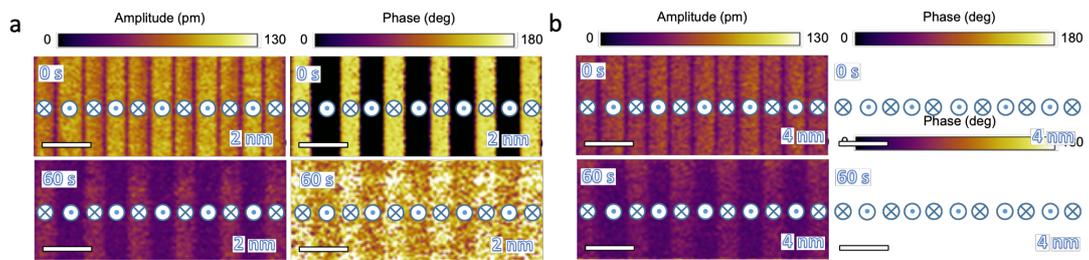


**Fig. S10** PFM amplitude images after (a-c) electrical poling and waiting for 60 s in dark and (d-f) after illuminating the sample for 60 s for 1.5, 10 and 100 nm samples, respectively

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**Fig. S11** (a) PFM phase images collected after indicated illumination time. Arrows indicate the preset polarization state direction. (b) Phase profiles after indicated illumination time obtained from averaging images of panel (a) along vertical axis. (c) DPhase as a function of illumination time evaluated as the phase contrast of phase profiles in panel (b)



**Fig. S12** PFM amplitude and phase images collected just after poling and after 60 s under illumination for (a) 2 nm and (b) 4 nm HZO samples capped with 10 nm BTO