

Electronic Supplementary Information

Large enhancement of ferroelectric polarization in $\text{Hf}_{0.5}\text{Zr}_{0.5}\text{O}_2$ films by low plasma energy pulsed laser deposition

Tingfeng Song,^a Raul Solanas,^a Mengdi Qian,^a Ignasi Fina,^{*a} and Florencio Sánchez^{*a}

^a Institut de Ciència de Materials de Barcelona (ICMAB-CSIC), Campus UAB, Bellaterra
08193, Barcelona, Spain

* ifina@icmab.es, fsanchez@icmab.es

S1: Sketches of the P_{Ar} and P_{O_2} partial pressures used to grow the films

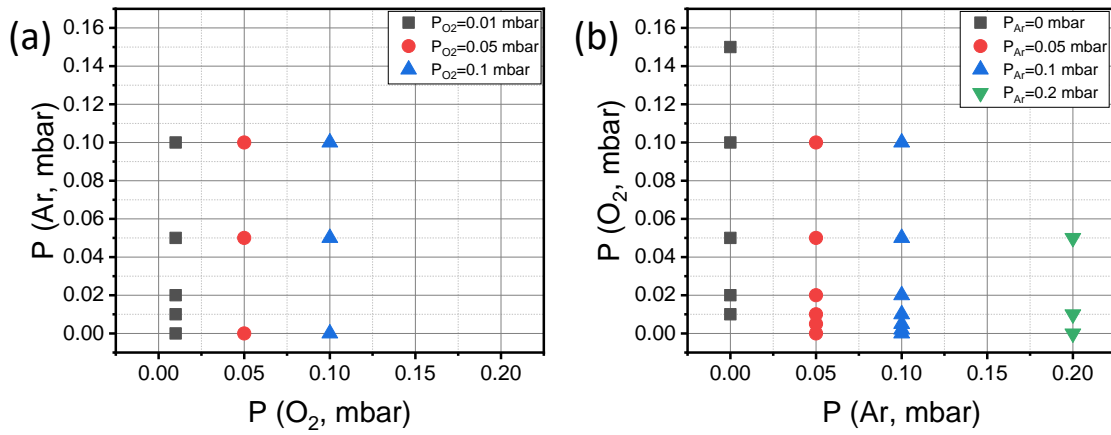


Figure S1. In (a) the three series of films deposited at fixed P_{O_2} are plotted using black squares ($P_{O_2} = 0.01$ mbar), red circles ($P_{O_2} = 0.05$ mbar), and blue up triangles ($P_{O_2} = 0.1$ mbar). In (b) the four series of films deposited at fixed P_{Ar} are plotted using black squares ($P_{Ar} = 0$ mbar), red circles ($P_{Ar} = 0.05$ mbar), blue up triangles ($P_{Ar} = 0.1$ mbar), and green down triangles ($P_{Ar} = 0.2$ mbar).

S2: XRD θ - 2θ scans of the series of films

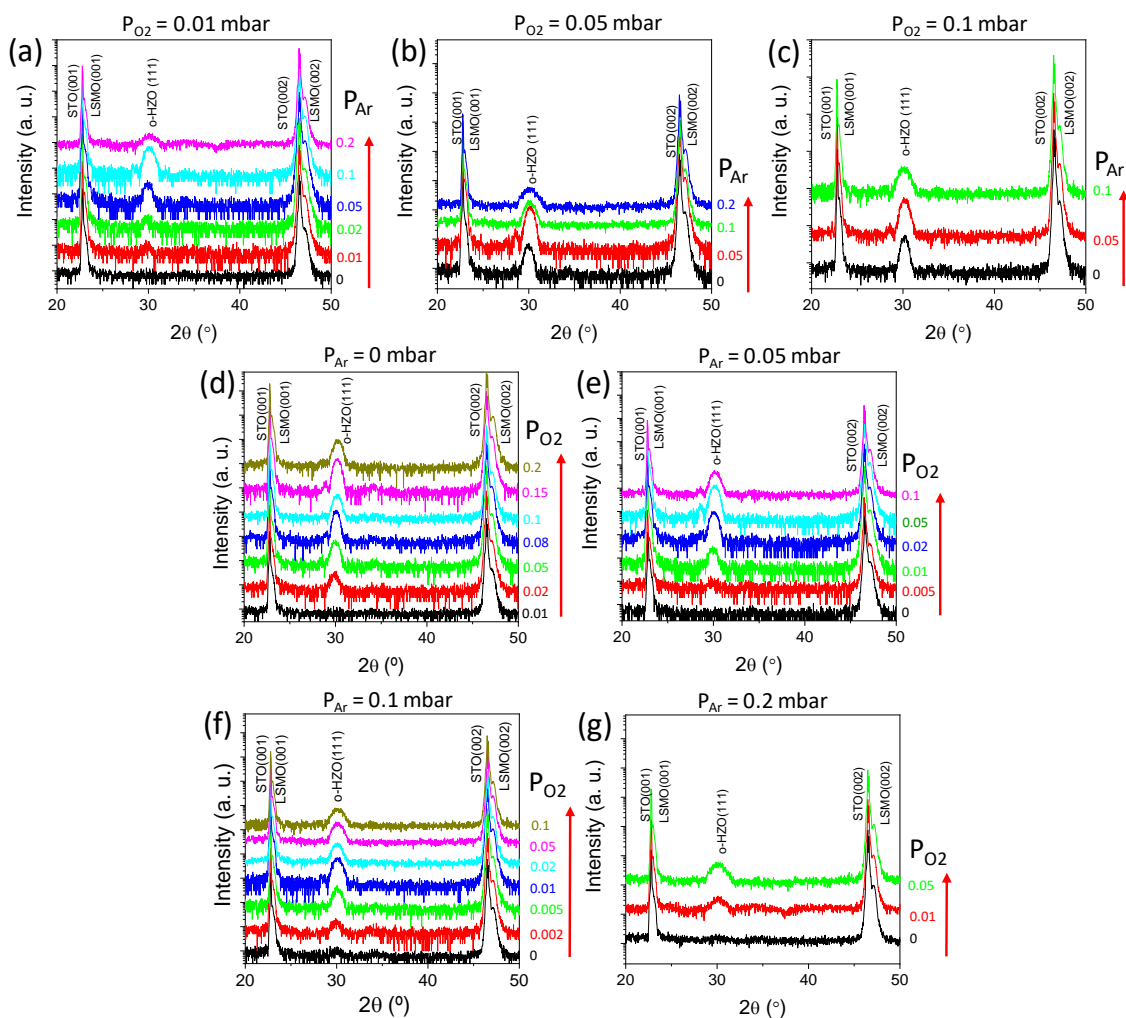


Figure S2: XRD θ - 2θ scans of the series of films deposited under varied P_{Ar} and fixed $P_{O_2} = 0.01$ mbar (a), $P_{O_2} = 0.05$ mbar (b), $P_{O_2} = 0.1$ mbar (c), and the series of films deposited under varied P_{O_2} and fixed $P_{Ar} = 0$ mbar (d), $P_{Ar} = 0.05$ mbar (e), $P_{Ar} = 0.1$ mbar (f), $P_{Ar} = 0.2$ mbar (g).

S3: Simulation of Laue oscillations

Figure S3 shows XRD θ - 2θ scans of selected samples around the o-HZO(111) reflection. The scans were measured using a longer time that the scans show in Figure 1. The o-HZO(111) reflection is simulated (red curves) according to the equation.¹

$$I(Q) = \left(\frac{\sin\left(\frac{QNc}{2}\right)}{\sin\left(\frac{Qc}{2}\right)} \right)^2$$

where $Q = 4\pi\sin(\theta)/\lambda$ is the reciprocal space vector, N the number of unit cells along the out-of-plane direction and c the corresponding lattice parameter.

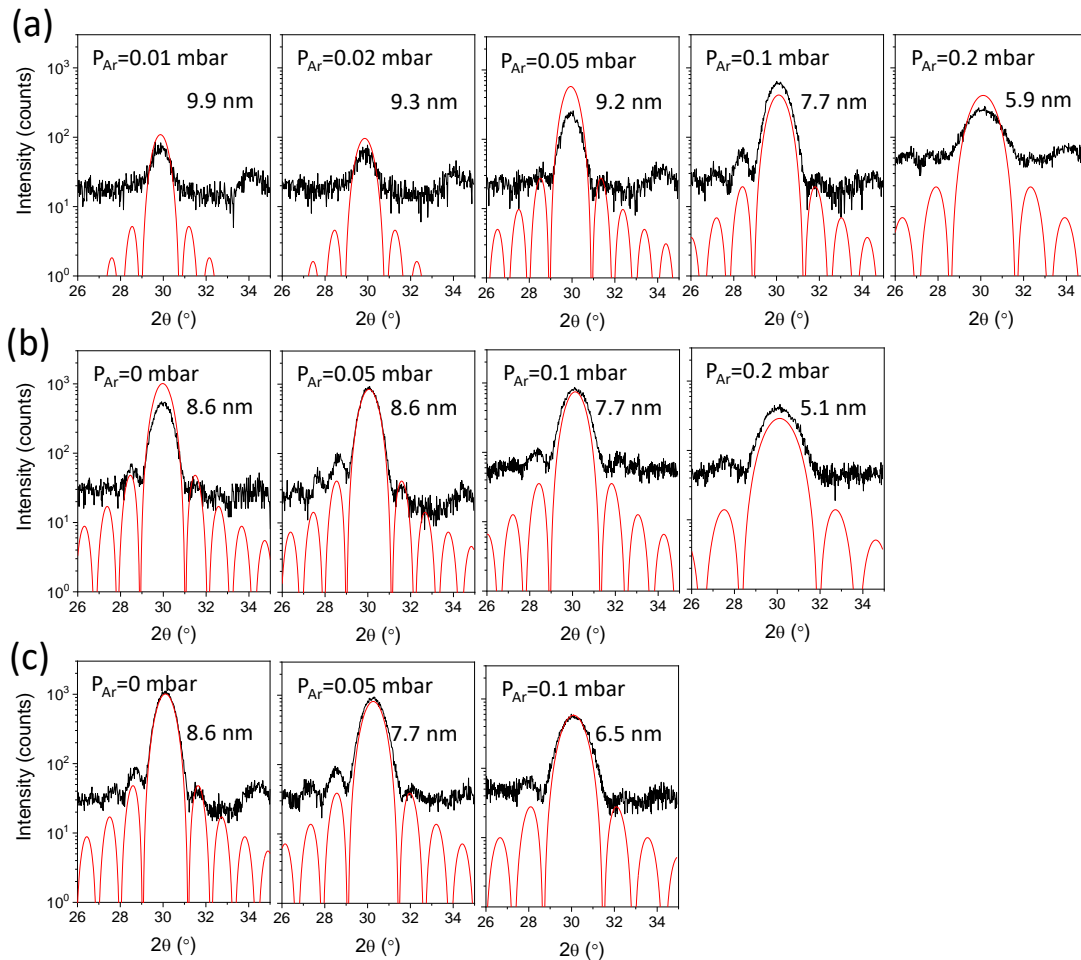


Figure S3. XRD θ - 2θ scans around the o-HZO(111) reflection. (a) scans of films deposited under $P_{O_2} = 0.01$ mbar and the P_{Ar} pressure indicated in each panel. (b) scans of films deposited under $P_{O_2} = 0.05$ mbar and the P_{Ar} pressure indicated in each panel. (c) scans of films deposited under $P_{O_2} = 0.1$ mbar and the P_{Ar} pressure indicated in each panel. Red lines in each panel are Laue fits. The thickness estimated by the fits is indicated in the corresponding panel.

S4: Thickness and growth rate as a function of P_{Ar}

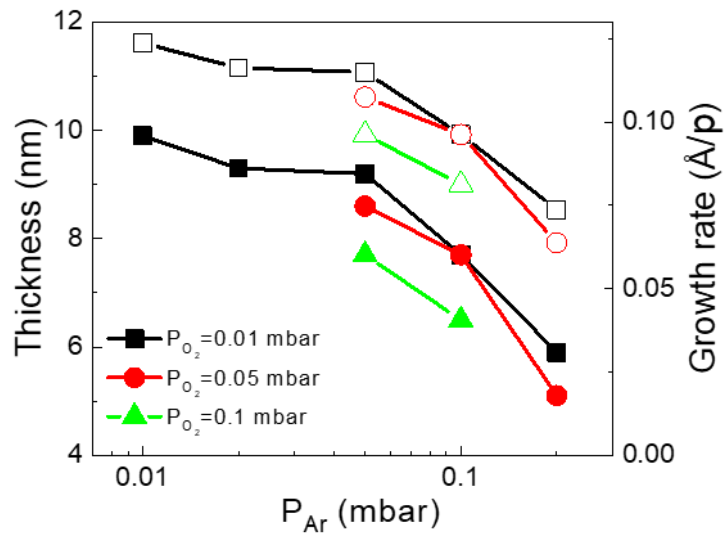


Figure S4. Thickness (solid symbols, left axis) and growth rate (empty symbols, right axis) as a function of P_{Ar} for fixed $P_{O_2} = 0.01$ mbar (squares), $P_{O_2} = 0.05$ mbar (circles) and $P_{O_2} = 0.1$ mbar (triangles). Thickness of the films was estimated by simulation of Laue oscillations as described in Figure S3.

S5: XRD pole figures

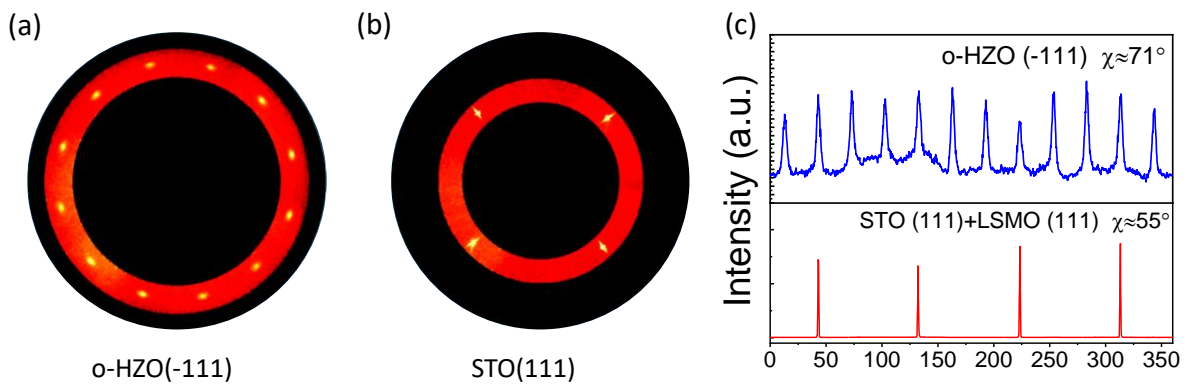


Figure S5. XRD pole figures of the $P_{O_2} = 0.05$, $P_{Ar} = 0.05$ mbar sample, from (a) o-HZO(-111) and (b) STO(111) reflections. (c) Corresponding ϕ -scans around o-HZO(-111) and STO(111). The pole figures confirm that the orthorhombic phase is epitaxial, presenting four families of crystal variants.

S6: Intensity of the o-(111) reflection, normalized to that of the STO(002) peak

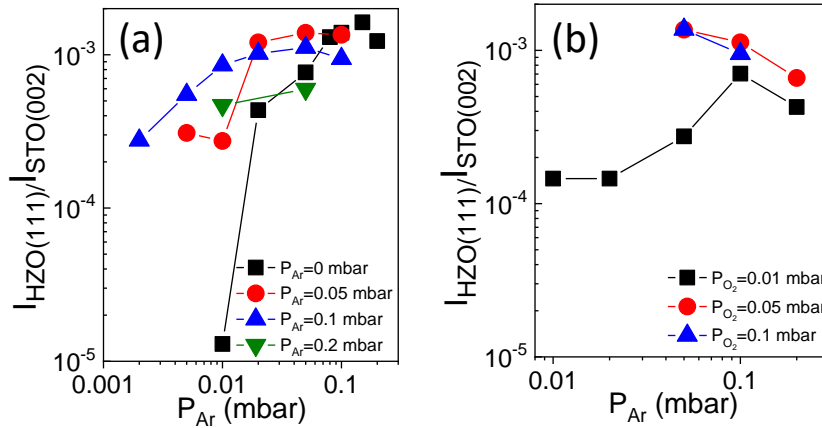


Figure S6. Intensity of the o-(111) reflection, normalized to that of the STO(002) peak, $I_{\text{HZO}(111)}/I_{\text{STO}(002)}$, as a function of P_{O_2} and P_{Ar} . In (a) P_{Ar} is: 0 mbar (black squares), 0.05 mbar (red circles), 0.1 mbar (blue up triangles), and 0.2 mbar (green down triangles). In (b) P_{O_2} is: 0.01 mbar (black squares), 0.05 mbar (red circles), and 0.1 mbar (blue up triangles). The same dependences are obtained normalizing the intensity of the o-(111) reflection to that of the LSMO(002) peak, $I_{\text{HZO}(111)}/I_{\text{LSMO}(002)}$, as shown in Figure 2 of the manuscript.

S7: The normalized o-(111) reflection

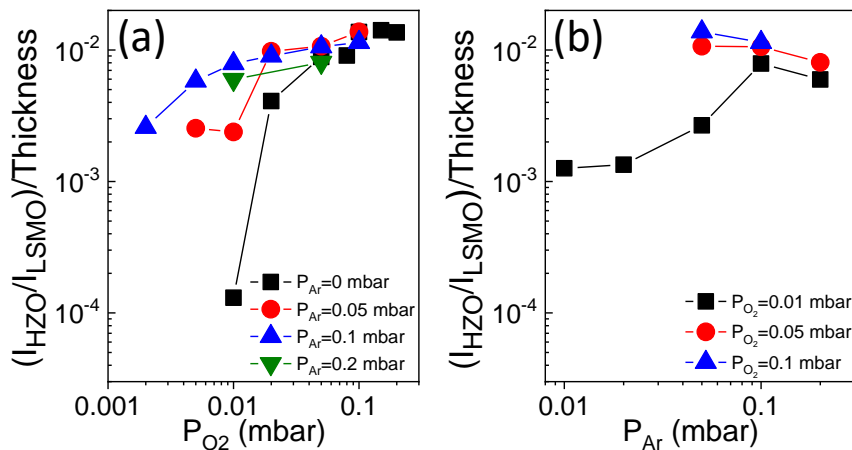


Figure S7. The intensity of the o-(111) reflection, normalized to that of the LSMO(002) peak, $I_{\text{HZO}(111)}/I_{\text{LSMO}(002)}$, shown in Figure 2 of the manuscript as a function of P_{O_2} and P_{Ar} is normalized to the film thickness. In (a) P_{Ar} is: 0 mbar (black squares), 0.05 mbar (red circles), 0.1 mbar (blue up triangles), and 0.2 mbar (green down triangles). In (b) P_{O_2} is: 0.01 mbar (black squares), 0.05 mbar (red circles), and 0.1 mbar (blue up triangles).

S8: Leakage - voltage curves of films

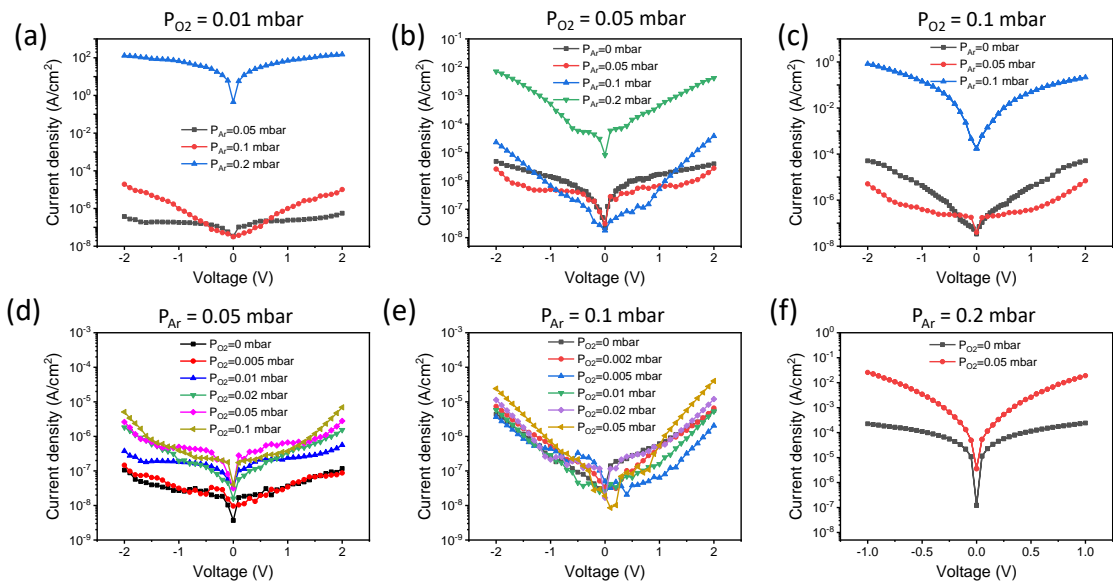


Figure S8. Leakage - voltage curves of films deposited under varied P_{Ar} and fixed $P_{O_2} = 0.01$ mbar (a), $P_{O_2} = 0.05$ mbar (b), $P_{O_2} = 0.1$ mbar (c), and the series of films deposited under varied P_{O_2} and fixed $P_{Ar} = 0.05$ mbar (d), $P_{Ar} = 0.1$ mbar (e), $P_{Ar} = 0.2$ mbar (f).

References

- 1 J. Lyu, I. Fina, R. Solanas, J. Fontcuberta and F. Sánchez, *Appl. Phys. Lett.*, 2018, **113**, 082902.